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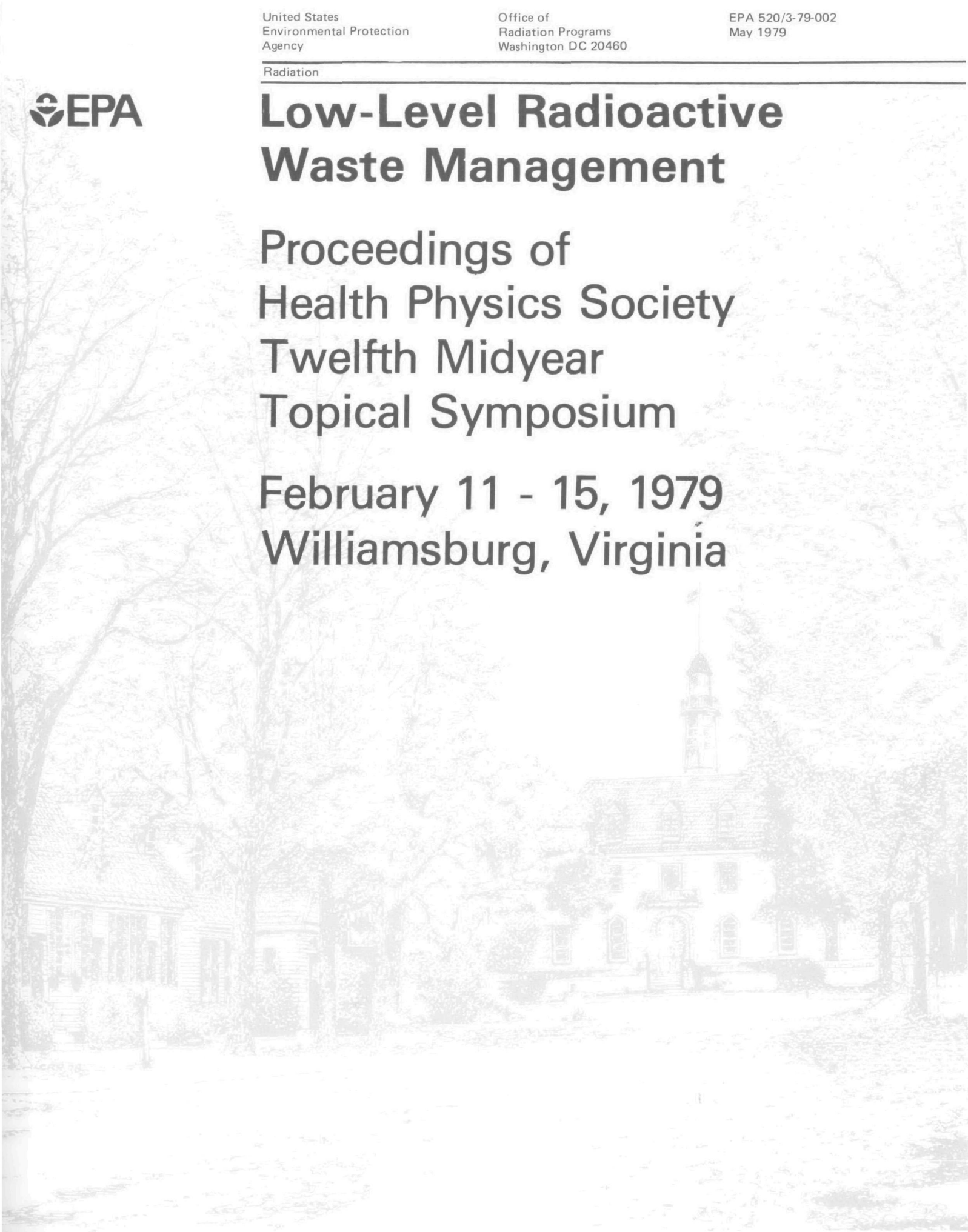

LOW-LEVEL RADIOACTIVE WASTE MANAGEMENT

PROCEEDINGS OF HEALTH PHYSICS SOCIETY TWELFTH MIDYEAR TOPICAL SYMPOSIUM

FEBRUARY 11-15, 1979
WILLIAMSBURG, VIRGINIA

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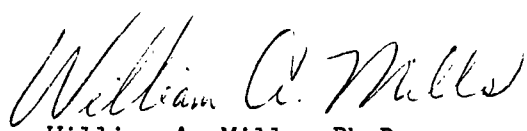
EDITOR

JAMES E. WATSON, JR.

FOREWARD

The management of the Nation's radioactive waste is a matter vitally important to not only the availability of electrical power but also other beneficial uses of radioisotopes for medical and industrial uses. Though not previously considered as radioactive waste, the by-products of many mining and milling operations for other than uranium, e.g., phosphates and metals, contain naturally-occurring radioactive materials which must be addressed in a total waste management program. Often the focus of attention is on high-level radioactive waste management related to nuclear power generation. While obviously this type of radioactive waste merits attention, the Health Physics Society has provided a valuable service in providing an opportunity for scientific and technical discussions to focus in this Mid-year Symposium on the broad concerns of "low-level radioactive waste management." The topics discussed in these proceedings clearly demonstrate that low-level radioactive waste materials should and can be managed for the protection of public health. The Office of Radiation Programs in the Environmental Protection Agency is pleased to have the opportunity to sponsor the publication of these proceedings for the benefit of not only health physicists but also for those interested in assuring that the public health is protected and our environment respected.



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ACKNOWLEDGEMENTS

Appreciation is extended to all authors and chairpersons for their participation in this symposium. In general, papers are presented in these proceedings exactly as submitted by the authors and no editing of content was undertaken. In a few cases figures and/or tables of a particular manuscript were grouped for photo-reduction, and format changes were made in papers 8 and 21.

The symposium proceedings were published through the Office of Radiation Programs, U.S. Environmental Protection Agency. This method of publication made the proceedings available to attendees and other interested persons in a timely manner. Coordination between EPA and the program committee in the preparation of these proceedings was very capably managed by Dr. Stephen T. Bard.

It is impossible to acknowledge all of the many persons who unselfishly gave of their time to work on the preparation for this symposium. Many hours, days and weeks were worked by those persons shown in the section "Symposium Committees" and by other members of the North Carolina and Virginia Chapters. Any credit for this symposium is shared by all members of these Chapters.

A special acknowledgement must be made to my secretary, Ms Frances L. Dancy. Ms Dancy's work covered the preparation of the abstract form, the preliminary technical program, the book of abstracts and these proceedings. During this time she also handled a multitude of correspondence. Her efforts made it possible for the program committee to meet all established schedules.

James E. Watson, Jr.

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* Withdrawn

** Accepted But Unable To Present In Person

*** Added

KEYNOTE ADDRESS

Low-level Radioactive Waste Management - Retrospect and Prospect

J A Lieberman
Nuclear Safety Associates

There are probably two purposes of a keynote address. The first presumably is to set the stage or theme for the conference. However, after looking at the program I think one can fairly argue that just the listing of the papers and the authors does this almost automatically. The scope and nature of this conference is already pretty well identified, and I am sure we all can look forward to a very productive meeting. The second purpose of a keynote address, at least the ones that I have watched, is more or less a mechanism for "clearing the aisles," so to speak, and getting on with the business of the conference so we can hear and discuss the papers. The aisles seem reasonably clear. Nevertheless, I hope the remarks I have to make will help serve both purposes in some small way.

Right at the outset I would state my conception of the keynote of this meeting in fairly simple terms that I think are consistent with the "retrospect and prospect" which is the title of my talk. It goes somewhat as follows.

In the field of management of low-level radioactive waste, we are the beneficiaries of a record and a heritage, if you will, of which I think we can be reasonably proud. This record and heritage are the result of work by many dedicated and competent people who from the very beginning of this industry responsibly recognized the public health and safety and environmental impact potential of the waste associated with the industry. As a matter of fact there are a number of these people who are in the audience this morning. This does not mean that we can be allowed to rest on any self-acclaimed laurels. We should continually direct our efforts in the future to doing the job of managing these materials more effectively and more efficiently, but this continuing effort must be rational as well as prudent. The rationality must stem from a sound scientific and technical base and a realistic public cost-benefit equation, that is, one that doesn't have zero risk or zero release as one of its terms. The institutional control and regulatory framework

should be established on a similar basis, and should also provide better communication to the public, so that they can, if not be assured of, at least be afforded an opportunity to understand the validity of the system. Having stated this theme, two separate statements I ran across recently came to my mind. The first was one that was made by Jim Coulter, Secretary to the Department of Natural Resources for the State of Maryland. I think this statement was made in connection with some problems the states were having in water pollution control and, as I recall, the control of trihalomethanes. Jim said something to the effect that he thought a distinct form of hell was being in heaven, or close to it, and not knowing it. I think that might have some pertinence to our low-level waste management situation. The other statement that I ran across recently was in an article by Aaron Wildofsky, in the recent issue of the American Scientist that some of you might have seen. He is now, I believe, President of the Russell Sage Foundation. In the course of this article he made the remark, that struck a responsive chord as far as I was concerned, that "Chicken Little is alive and well in America."

There is also a little story that comes to mind that I think also has some pertinence, and anyway I think it is sort of an interesting little story. It concerns a Jewish grandmother. It seems that there was this grandmother and her only daughter, who was either divorced or widowed but was alone, and a young grandchild about four or five years old, who obviously was the apple of his mother's and certainly his grandmother's eye. A lot of affection and care was bestowed on this youngster. Now it turns out one day the grandmother wanted to go to the beach and wanted to take the child along. Well, the child's mother was quite reluctant—they were very careful about this youngster, who was the only remaining line in the family—and said no, she didn't think the grandmother ought to take the child, that it may not be safe, and he was a very active youngster who might be too much to handle. But the grandmother said it was a nice day and might be relaxing, and finally the mother relented and grandma took the youngster to the beach. Well, the youngster was playing at the water's edge and all of a sudden a great big wave came in, and lo and behold, took the child out to sea. Well, you can imagine the weeping and wailing of the grandmother and her very fervent supplications to God, to please,

please not take this youngster from her, and she prayed and she wailed and sure enough, another wave came along and deposited the youngster at her feet unharmed. Well, she quickly embraced the child, but then cast her eyes heavenward and, in rather a stern voice and with a stern visage, she complained: "He had a hat!"

Now back to a little reprospection—or perhaps even reminiscences. In my view the roughly thirty-five-year history of the nuclear business is probably categorized or broken down into three phases or milestones. The first one, of course, began with the Einstein letter to Roosevelt that led to the Manhattan district, the weapons program, and subsequently to the establishment, after World War II, of the AEC. The second phase was the passage of the Atomic Energy Act of 1954 which, as we all know, was the National Policy basis for the civilian commercial nuclear power industry. The third, in my view, was the passage ten years ago of the National Environmental Policy Act. Some people crudely refer to this as the Lawyers and Consultants Relief Act. It has resulted, I think it's clear to all of us, in a much greater involvement of the third branch of government, the judiciary, and, what is now increasingly recognized by many as the fourth branch of government, (incidentally not called for in the Constitution) the regulatory agencies, in nuclear related issues.

My first contact or exposure with low-level waste management activities was in the old Atomic Energy Commission in the late 40's, just after the first phase, and it was associated with what was then called the "particle problem" at Hanford and Oak Ridge. At the time there was an organization established called the Stack Gas Problem Working Group, a group of outstanding authorities in matters relating to the subject area, and out of the work of that group came things like the high-efficiency particulate filters, and the publication Meteorology and Atomic Energy, a basis for assessment of atmospheric transport and diffusion, and other developments that I think are still in good stead.

With regard to solid low-level waste, one of my early exposures was to the activities that went on at the Knolls Atomic Power Laboratory (KAPL) and at Argonne. The activities I refer to had to do with the waste compaction and incineration work that was going on there. The compaction work at KAPL,

which involved shipment of the waste to Oak Ridge, I thought was quite effective But I must say that the early experience on incineration, particularly at Argonne, (I can say this because my colleague Walt Rodger was responsible for making that incinerator work) led to my own general disenchantment with incineration as a volume reduction technique. I can be convinced otherwise, however, and I expect there have been some later developments that might change my conclusions from that early experience.

Shallow land burial, such as the operations at Hanford, Oak Ridge, Savannah River, and Idaho, was carried out by first-rate people, and in my view they did a very good job. We have to remember that this was almost nineteen or twenty years ago. I think it was in May of 1960 that the burial grounds at Oak Ridge and Idaho were made available for commercial low-level waste, and it was early in 1961 that the Atomic Energy Commission established the basis for commercial shallow burial operation on federal or state-owned land. Of course, we are all aware that between '62 and '67 there were five such commercial establishments licensed at Beatty, Maxie Flats, West Valley, Hanford, and Sheffield, and the sixth, at Barnwell, was licensed in 1971. While much has been made by some of releases or other so-called "problems" from some of these operations, in my judgement, in terms of safety and environmental protection, these activities rate pretty good marks.

Some of my early contacts with sea disposal were on the East coast, out of what was then called the Naval Ammunition Depot at Earle, New Jersey and off the Farralone Islands on the West Coast. I recall taking a trip in an LST, which was used for transporting the waste from the NAD at Earle. It was in the winter time and the weather was pretty bad but we took off early one morning with the deck of the LST loaded with 55-gallon drums and headed out to the established, designated disposal area off about 100 miles the New Jersey coast. It was a very straightforward operation. When we got to the place where the skipper said his navigation chart showed him the disposal site was located, the sailors proceeded to just roll the 55-gallon drums overboard. They were categorized by source, i e originating institution, and there were a few drums that came from an institution that shall remain unnamed that

didn't sink, and that's how the sailors got their rifle practice that morning. Again, I don't mean to imply that everything that was done in those days was perfect, but nevertheless, as a result of many efforts by some thoroughly competent people, the assessment and the investigative efforts related to sea disposal, both nationally and internationally, I believe showed pretty well that scientifically, technically, and environmentally sea disposal for certain categories of waste was indeed acceptable.

Socially and politically, however, it was an entirely different story. I recall with not much pleasure being a witness before the Joint Committee on Atomic Energy with Senator Pastore the Chairman, and being interrogated—to put it mildly—on questions related to sea disposal. As a result of those kinds of non-scientific or technical considerations which are certainly commonplace now, there was a moratorium placed on new licenses for sea disposal in the early sixties, as I recall, and sea disposal was terminated in the United States in the late sixties or early 1970. The future viability of sea disposal, at least in my mind, is questionable, again on socio-political grounds, although I recognize that this technique is being practiced in other parts of the world. With the shut-down of West Valley, Maxey Flats, and Sheffield, the volume limitations in effect at Barnwell, and withdrawal of the license application at Cimmaron, the current major problem, in my view, is the assurance of an adequate disposal capacity, particularly east of the Rockies. The recent IRG draft report to the President, which we are familiar with, estimates the total acreage required for Department of Energy low-level waste plus commercial low-level wastes by the year 2000 ranges from about 450 to 1,650 acres depending on an assumption of a volume reduction factor of 5 after 1985. The IRG apparently did recognize the seriousness of the low-level waste problem, and stated (and I quote) "That there presently exists neither a coordinated national program for management of these low-level wastes or an institutional mechanism to deal effectively with these issues." It then recommends that DOE assume responsibility for developing and coordinating the needed national plant for low-level waste management with active participation and advice from other concerned federal agencies and input from the states, general public, and

industry. I look forward to Sheldon Myers discussion of this subject this afternoon. The IRG further recommends that states be provided the option to retain management control of existing commercial low-level waste or to transfer such control to the federal government. The draft report goes on to suggest that future sites could be developed either by the individual states or by the federal government, but such action should be taken within the agreed-upon framework of an overall low-level waste siting plan developed through a joint Federal/State partnership.

This leads to the prospective part of my remarks.

Of course, there are a number of issues involved—technical, institutional, and administrative. One of the more important technical issues in my judgement is that of low-level waste classification. In my view the publication of NUREG-0456, titled "A Classification System for Radioactive Waste Disposal - What Goes Where" represents an important and valuable contribution to the rationalization of this subject. I believe we are to hear John Adam on this later in the program. Nuclear Safety Associates, working with the Utility Waste Management Group independently arrived at a similar approach and methodology and we believe it provides a sound basis for the development of comprehensive regulations which are essential for consistent, proper management of low-level wastes. While it is clear, at least to me, that the definition of waste categories based on limiting concentration of specific isotopes makes sense, it is equally clear that such isotopic limits cannot be used in any practicable way in the field to individually classify each waste package. A program of sampling and analysis of representative wastes from particular sources as a basis for <u>generic</u> classification of low-level wastes from particular sources would appear to be in order. I might note that the Utility Waste Management Group has taken some initial steps in this direction.

It should also be pointed out that the analyses of NUREG-0456 and other independent studies, including UWMG, have shown that the water migration pathway is relatively unimportant in terms of shallow land burial ground performance.

Other factors that need to be carefully considered in connection with their effectiveness and cost in terms of burial ground performance include volume reduction and solidification. Considering the critical exposure pathways,

it is not immediately clear that the general need for such operations exists. Systematic cost-benefit analyses are in order.

Another area of possible technical consideration is that of alternative disposal systems or methods. The recent NRC advance notice of proposed rule-making in the low-level waste area begins with the statement that "recent developments at the six commercially operated shallow land burial sites have highlighted the need for an explicit regulatory program for shallow land disposal of such wastes by alternative methods." While I am not sure what was meant by "recent developments" I think it is important to make clear that they do not, or should not, imply that any significant hazard to public health and safety has resulted from the operation of shallow land burial grounds. I, of course, fully agree with the need to upgrade the regulatory program for disposal of low-level wastes. But that does not stem from any previous hazard to the public, i e significant technical deficiencies. Instead, it arises because the current regulatory framework does not make sufficiently explicit the information to be provided by an applicant proposing to undertake such an operation, or the standards by which such application will be judged, and because the public is not properly informed as to the bases for the regulatory programs and thus lacks both a proper perception of the limited hazards and confidence in the regulatory agencies and their programs. Accordingly, what is needed is both an improved regulatory program and better communication thereof to the public. I am glad to see the NRC getting on with at least meeting the first part of that need.

The statement in the advance notice appears to overemphasize the possible need for alternative methods of disposal. I believe that the NRC studies and the independent studies rererred to reasonably demonstrate three major points.

a) Shallow land burial is an acceptable method of disposing of radionuclides up to determinable concentrations;

b) The limiting concentrations for essentially all isotopes are determined not so much by the capability of a properly located, designed and operated burial site to contain the isotopes once they are buried, but by handling procedures upon receipt and burial of the waste and by the possibility that intrusion into the waste by individuals may occur at some later date; and

c) The limiting concentrations are higher than the concentrations in wastes which have traditionally or historically been considered

It follows that the need for alternative methods is marginal at best and I strongly support NRC's stated position that "the regulations (10 CFR Part 61) will initially cover only the currently practiced method of shallow land burial" since this is clearly the primary need.

In this same vein, I have an additional general comment concerning alternatives. The advance notice states, "The Commission believes that development of those parts of its program dealing with regulation of alternative methods of disposal of low-level wastes should be accelerated, since such alternatives offer means of providing additional disposal capacity." Alternatives might indeed offer additional capacity, but so would expansion of existing sites, reopening of closed sites, and opening of additonal shallow land burial sites. I believe that any of these three options could be exercised more practicably and more expeditiously than other alternatives. I would urge that work on other alternatives not be permitted to slow the schedule for putting into place the 10 CFR Part 61 regulations relating to shallow land burial, which, incidentally, I understand will not be available in draft form until April 1980.

In passing, I would also comment that the "stretching" consideration of alternatives in NUREG-0308 might well be counter-productive. Even the passing addressing of putting LLW into space and the atmospheric disposal via a tethered balloon tends to imply a more difficult technical problem than really exists and detracts from the credibility of the overall effort.

Other specific issues that have to be dealt with in the near future include those of:

a) Federal-State relationships.

This is, in my view, a critical area. While some useful ad hoc steps have been taken recently to improve the working relationships I believe a more specifically defined, and perhaps even a more formalized process for State involvement must be established in order to overcome this serious bottleneck to orderly, effective low-level waste management. In any case, I believe the States should have the option to retain control or existing commercial facilities or transfer control to the Federal government, and that LLW disposal sites be licensed either directly by NRC or by the State through the Agreement State process. (A State should not be allowed to restrict use of a licensed facility.)

b) DOE take-over of commercial waste burial grounds and opening of DOE facilities to commercial users.

This, in some respects, is connected to the Federal-State relationships issue. There may well be some "sticky wickets" so to speak, for example, the requirement for licensing of the DOE facilities, that would have to be resolved before one might be able to arrive at conclusions on this issue. Nevertheless, there are sufficient incentives to justify vigorous analysis of the pros and cons of these possible actions. Perhaps Sheldon Myers will also enlighten us in this area.

c) On-site management at reactor sites.

Because of the potential critical nature of disposal capacity availability, prudence would dictate careful examination of feasible actions in this are. The National Environmental Studies Project of AIF has sponsored an investigation by NUS of possible on-site alternatives and a report should be out shortly.

There are, of course, a number of additional pertinent issues or topics that I know merit recognition but I will finish as I began.

We really haven't done all that badly over the past three decades or more in managing low-level radioactive wastes. Let's get on with doing the job more efficiently and effectively but let's do it as I said—with prudence and with some reasonable degree of rationality. Let's not ourselves be Chicken Littles. Let's carry on with professionalism and integrity—after all, that's what the Health Physics Society is all about.

SESSION A

ORIGINS OF LOW-LEVEL RADIOACTIVE WASTE

Session Chairperson

R. J. Stouky

NUS Corporation

SOURCES, AMOUNTS, AND CHARACTERISTICS OF LOW-LEVEL RADIOACTIVE SOLID WASTES

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Abstract

Low-level radioactive solid wastes (LLW) are generated in the nuclear fuel cycle, national defense programs, institutional (especially medical/biological) applications, and other research and development activities. The estimated total accumulation of defense LLW, $\sim 50.8 \times 10^6$ ft^3 ($\sim 1.4 \times 10^6$ m^3), is roughly three times that estimated for commercial LLW, mill tailings excepted. All nuclear fuel cycle steps generate some LLW, but power plants are the chief source. From 1975 through 1977, reactor process stream cleanup generated $\sim 1 \times 10^6$ ft^3 ($\sim 2.8 \times 10^4$ m^3) annually. Spent fuel storage (or reprocessing) and facility decontamination and decommissioning will become important LLW generators as the nuclear power industry matures.

The LLW contains dry contaminated trash, much of which is combustible and/or compactible; discarded tools and equipment; wet filter sludges and ion-exchange resins; disposable filter cartridges; and solidified or sorbed liquids, including some organics. A distinguishing characteristic of LLW is a long-lived alpha-emitting transuranic content of <10 nCi/g; this limit, however, is presently under review by NRC. If it is increased, the amount of LLW would also increase. The nonfuel-cycle waste generation rate in 1975 was estimated to be $\sim 7.6 \times 10^5$ ft^3 ($\sim 2.1 \times 10^4$ m^3)/yr. The majority of these wastes, $>6 \times 10^5$ ft^3 ($>1.7 \times 10^4$ m^3), was medical and academic wastes which usually contained isotopes with induced activities of ≤60-day half-life, neglecting ^{3}H and ^{14}C. The remaining research and development LLW contained a broad spectrum of radioactive species that was relatively small in total volume [probably $\sim 1.5 \times 10^5$ ft^3 ($\sim 4.2 \times 10^3$ m^3)]. The amount of nonfuel-cycle waste that is generated annually has been steadily increasing; thus, these estimates tend to be conservative. The routine power-plant LLW contains varying amounts of activated corrosion (e.g., ^{60}Co, ^{59}Fe, and ^{54}Mn) and fission products (e.g., ^{134,137}Cs and ^{90}Sr); the filter sludges and ion-exchange resins have the highest radiation levels and normally require biological shielding and remote handling.

DISCUSSION

The problems that have been encountered at some shallow land-burial sites for low-level radioactive solid wastes (LLW) have stimulated much interest in finding more adequate ways of treating, handling, and disposing of these wastes. There are several ongoing studies being made by various Governmental agencies, among them the NRC, EPA, USGS, DOE, and some bodies at the state level. Nearly all of these studies seem to point up the importance of characterizing all the kinds of LLW with regard to source, volume generated, the type and amount of

radioactivity contained, and the physical form of the waste and its container. A knowledge of these parameters as they now exist is useful in future planning.

Low-level radioactive solid waste is usually described as having: (1) low enough beta-gamma activity levels so that no special provision must be made for heat removal, and (2) penetrating radiation levels such that only minimal or no biological shielding or remote handling is necessary. In addition, it is generally considered to contain <10 nCi/g of transuranic alpha-emitters. The 10-nCi/g value is currently under review by the NRC and may possibly be changed. Obviously, an increase would instantly decrease the amounts of transuranic (TRU) waste now in retrievable storage while simultaneously increasing the amount of LLW acceptable for burial. Only those wastes containing <10 nCi/g transuranics are addressed in this discussion.

An estimate of the accumulated volumes of LLW now buried in existing burial grounds has recently been reported by the Interagency Review Group in their Report to the President (IR78). The volume of buried LLW due to defense operations is about three times that due to commercial operations [i.e., 50.8×10^6 vs 15.8×10^6 ft^3 (1.4×10^6 vs 4.5×10^5 m^3)]. It should be pointed out, however, that the collection time for the defense wastes is about double that for the commercial wastes (i.e., approximately four decades vs approximately two decades). The annual generation rate of defense LLW has leveled off at $\sim 1 \times 10^5$ ft^3 (2.8×10^3 m^3).

The commercial wastes can be broken down into two categories, namely, fuel cycle and nonfuel cycle. Some fuel-cycle LLW is generated in each step of the cycle: mining, milling, conversion of UO_2 to UF_6, enrichment, fuel fabrication, and reactor operation. Spent fuel storage (or reprocessing if we should ever choose that option) and facility decontamination and decommissioning will become important LLW generators as the nuclear power industry matures. At present, with mill tailings excluded from the LLW classification, the light water-cooled reactor (LWR) power plants themselves are the largest generators of fuel-cycle LLW, and the fuel fabrication plants are second.

It has been pointed out in a recent study at the University of Maryland that most of the nonfuel-cycle LLW results from medical and academic (or institutional) applications (An78). The remaining nonfuel cycle wastes are generated in industrial or other research applications. The medical-type wastes comprised the major fraction of the institutional wastes, with the academic wastes representing about one-eighth of the total.

Of the total LLW shipped to commercial burial grounds, it is estimated that, by volume, the fuel-cycle wastes represent ~60% and nonfuel-cycle wastes make up the remaining 40%. The total volume of fuel-cycle LLW is $\sim 1.2 \times 10^6$ ft^3 (3.4×10^4 m^3), and ~80% of this waste, or $\sim 1 \times 10^6$ ft^3 (2.8×10^4 m^3), is generated at LWR plants. Another estimated 2×10^5 ft^3 (5.7×10^3 m^3) results from fuel fabrication. Compared to these waste volumes, the other commercial LLWs generated in the fuel cycle are almost negligible.

Based on 1975 data (An78), an estimated total annual volume of nonfuel-cycle commercial LLW is in the order of 7.6×10^5 ft^3 (2.2×10^4 m^3). Of this, medical/biological-type wastes make up ~80%, or $\sim 6.3 \times 10^5$ ft^3 (1.8×10^4 m^3).

Other academic-type wastes comprise ∿3 to 4%, or ∿2.7 x 10^4 ft^3 (7.5 x 10^2 m^3), and industrial, research, and other miscellaneous wastes make up the remainder [just under 15%, or ∿1.1 x 10^5 ft^3 (3.2 x 10^3 m^3)].

Up to this point, the three generic classifications of LLW (defense, fuel cycle, and nonfuel cycle) have been broadly described in terms of the estimated annual volumes of each that might be expected. An understanding of the physica chemical, and radiological characteristics of each waste type is also required if improved waste management methods are to be developed.

All generators of LLW have some dry wastes that are compactible and/or combustible (e.g., clothing, rags, paper, plastic, and wood). They also have a relatively small fraction that is not compactible and/or combustible (e.g., contaminated equipment, tools, and glass). Most of the defense and fuel fabric tion plant LLWs that are buried fall in these categories. In the fuel cycle, dry LLW probably represents between 30 and 40% of the total waste shipped to burial sites; dry wastes are estimated to comprise between 40 and 50% of the institutional LLW.

The wet LLWs generated at defense and fuel-cycle installations have many similarities and frequently have been treated in similar ways. Most unit opera tions commonly used in both defense and fuel-cycle process stream cleanup are the same, namely, ion exchange, filtration, and evaporation. The spent ion-exchange resins and filter sludges have, for the most part, been merely de-watered by decantation, filtration, or centrifugation, placed in drums (or shipping cask liners), and sent to the burial ground. Sometimes absorbent mate rials such as vermiculite are added to take up any free liquid that may remain. The spent resins and filter sludges together probably account for 10 to 20% of the fuel-cycle LLW sent to commercial burial grounds. Recently, more stringent requirements have been imposed on nuclear power plants, the largest generators of these types of waste within the fuel cycle. The current trend is toward immobilization of resins and sludges by incorporating them in a solidification agent such as cement or urea-formaldehyde resin. In the future, asphalt or unsaturated polyester resins may be used as solidification agents. In any case solidification of the resins and sludges will increase the LLW volume to be shipped by a factor of ∿1.2 to 2 (or greater), depending on the type of solidi-fication agent.

The evaporator concentrates at both defense installations and LWRs contain decontamination solutions and laboratory wastes. The main chemical differences appear to be the rather high concentrations of nitrates in the defense wastes, the borates that are the dominant constituent of pressurized-water reactor (PWR) wastes, and the sodium sulfate that characterizes boiling-water reactor (BWR) wastes. Evaporator concentrates are incorporated into a solidification agent prior to burial. The defense LLWs are usually solidified with cement. Up to the present, in the United States, either portland cement or urea-formaldehyde resin has been used for the LLW fuel-cycle liquids. Cement chemi-cally binds the water into the solid matrix, whereas urea-formaldehyde resin merely encapsulates it within the pores of the matrix material. Solidified evaporator concentrates account for an estimated 40 to 50% of the total LLW from the fuel cycle. Any fuel-cycle liquids that are solidified at the burial site are not included in this estimate. Before continuing, it should be mentioned that borates in high concentration may inhibit the setting of cement,

whereas high concentrations of sodium sulfate can interfere with urea-formaldehyde resin solidification.

The cartridge filters, predominantly used for stream cleanup at PWRs, are noncompactible, noncombustible solids that are treated as wet wastes. They are usually collected in a shielded shipping container, and after several have accumulated, the solidification agent is added. This type of waste probably represents <5 vol % of the total fuel-cycle waste shipped annually. The cartridge filters used in nuclear power plants are fabricated from such materials as cotton, synthetic fibers, polypropylene, matted paper, or porous ceramic.

A summary of the estimated proportions of the various types of LLW currently being shipped to burial grounds from LWRs each year shows that solidified concentrates are the largest fraction (∿40 to 50%), closely followed by the dry wastes (∿30 to 40%). The filters, filter sludges, and spent resins together comprise <25% of the ∿1 x 10^6-ft^3 (2.8 x 10^4-m^3) total waste volume shipped annually.

Historically, the BWRs have quite consistently generated more LLW per unit of thermal power output than the PWRs. A recent ORNL study of solid radwaste practices at nuclear power plants (Ki78) showed that the overall average, through 1977, for PWRs is ∿1.1 x 10^{-3} ft^3 (3.1 x 10^{-5} m^3) per MWh(t); the average for BWRs is slightly more than double this value [i.e., ∿2.3 x 10^{-3} ft^3 (6.4 x 10^{-5} m^3) per MWh(t)]. The number of operating PWRs is about double the number of operating BWRs.

As mentioned earlier, the dry solids in institutional LLW represent an estimated 40 to 50% of the total volume. Another 50% or so is comprised of analytical laboratory wastes, about half of which is solidified and absorbed liquids. Probably <10% is biological waste.

The industrial, research, and other nonfuel-cycle LLW, which is estimated to be slightly <15% of the total volume [i.e., ∿1.1 x 10^5 ft^3 (3.2 x 10^3 m^3)/yr], is not so well defined. It is made up of a miscellaneous mixture of assorted debris.

The radiological characteristics of the LLW vary in accordance with their point of origin. The activity in defense and fuel fabrication LLW is mostly due to uranium and its daughters although, in some cases, mixed fission products may be present. For the most part, however, the distinguishing characteristic of these wastes is the dominance of the naturally-occurring, long-lived alpha emitters.

The nuclear power plant wastes are characterized by their beta-gamma activity. Corrosion products circulating in the coolant are activated in the reactor core. The minute traces of uranium remaining on the fuel cladding after fabrication and/or failed fuel during reactor operation introduce mixed fission products into the coolant stream. The activities of most concern in these wastes are those which have half-lives of several years, for example, Co-60, Cs-134 and -137, and Sr-90. The filter sludges and ion exchange resins that arise from coolant cleanup operations are the most radioactive of these wastes, and they require biological shielding when being handled. The solidified evaporator concentrates and dry wastes generated at nuclear power plants are generally much lower in activity level.

The radioactive species associated with medical/biological LLW usually have relatively short half-lives, generally being <60 days if ^{3}H and ^{14}C are neglected. The other academic, industrial, research, and other miscellaneous LLWs contain a broad spectrum of radioactive species which are not easily classified.

Over the last five years or so, the characterization of LLW has been receiving increased attention that now seems to be culminating in a new National Low-Level Waste Management Program. Several institutions and agencies are now developing data bases that should provide a substantial foundation for making the necessary decisions we face in the near future regarding the treatment, storage, transport, and disposal of low-level radioactive solid wastes.

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A PERSPECTIVE ON THE RELATIVE HAZARD OF LOW-LEVEL RADIOACTIVE WASTE DISPOSAL

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Abstract

Low-level radioactive waste disposal is presently one of the major problems facing most of the industries and institutions which use and produce radioactive materials. A more widespread but less publicized waste disposal problem may, however, be facing almost all major industries. This problem is the disposal of other hazardous wastes.

This paper will examine the problem of radioactive waste management and the management of other hazardous wastes by comparing their relative toxicities. The relative risks of the total problem will then be assessed by comparing the generation rates of each. In addition, the radioactive wastes produced by the nuclear fuel cycle will be compared with the potentially hazardous waste produced by alternate energy sources, namely, the coal fuel cycle and the manufacturing of equipment for the collection of solar energy.

These comparisons suggest that the consequences of the disposal of other hazardous waste could result in risks to current and future generations which are comparable with or which may exceed that due to radioactive waste disposal.

Other aspects of the problems inherent in the safe management of hazardous waste compared to radioactive waste will also be explored.

Introduction

The safe disposal of radioactive waste has been called such things as the problem of centuries, a dilemma, unprecedented, the problem without solution and the monster. All these descriptions suggest that the problems involved with radioactive waste disposal are unique and most difficult to solve. To the contrary, there may indeed be a more difficult waste disposal task facing many industries other than those which use and produce radioactive materials. This task is the safe disposal of non-radioactive industrial hazardous waste.

What Is Hazardous Waste?

Almost all industries produce byproduct materials which have no commercial value and therefore must be disposed of as waste products. What makes this a very serious problem is the fact that about 10% of these industrial wastes are considered to be a hazard to both public health and the environment. These hazardous wastes include toxic heavy metals, primarily chromium, lead, arsenic and cadmium, and also highly dangerous chemicals and pesticides.

Table 1 lists those industries which were the major contributors to the hazardous waste disposal problem in 1977. The highly dangerous chemical and pesticide wastes were produced primarily by the organic chemical industry, while the waste from the other industries was considered hazardous mainly because of its toxic heavy metal content.

The heavy metals and chemicals that make the waste hazardous can cause adverse effects upon human health in a variety of ways which are much the same as the adverse effects of the radioisotopes in radioactive waste. Some of these toxic substances, if ingested in sufficient quantities, can cause immediate or acute harm, much like the hazards of ingesting or being exposed to high-level radioactive waste. Furthermore, many of these same toxic substances, if ingested in lesser quantities, can cause long term chronic effects, such as cancer and genetic damage, which is similar to the hazards of low-level radioactive waste and uranium tailings.

Another measure of the hazard of a substance is its persistence in the environment. All of the toxic heavy metals are stable elements and therefore remain hazardous forever, assuming that a possible change in chemical form will not affect their toxicity. Likewise, many of the highly dangerous chemicals are also extremely stable and their decomposition to less harmful substances may be uncertain over the long period of time that containment is necessary. Radioactive waste, on the other hand, does obey the law of radioactive decay. It will systematically decay to a relatively harmless material in a known period of time. For low-level radioactive waste, this period is a few hundred years. For high-level radioactive waste, the transuranic isotopes which are present may extend this period to many thousands of years. Therefore, from the standpoint of its persistence, much of the hazardous waste is comparable to the transuranic isotopes in high-level radwaste or the long lived naturally occurring isotopes in uranium tailings.

Toxicity of Hazardous Waste Versus Radioactive Waste

When comparing hazardous with radioactive waste, one of the most difficult questions to be addressed is the identification of an appropriate basis for comparing the relative toxicity of the two. At least one other study (Co77) has compared the relative toxicity of naturally occurring toxic heavy metals to high-level radioactive waste. That study concluded that the EPA safe drinking water regulations (En76) for toxic heavy metals was equivalent to the 10 CFR 20, Appendix B, Table 2, Col. 2 (MPC_w) limits for radioactive isotopes.

Since limits for radioactive isotopes are also given in the EPA safe drinking water regulations and since this is one of the few Federal regulations which addresses both, it may be more appropriate and definitely much more conservative to use this standard exclusively for comparing radioactive and hazardous waste. These regulations are also appropriate because they address the problem of public drinking water contamination, which is the most likely route of ingestion by man of any buried toxic material.

When looking in detail at the EPA drinking water regulations, it becomes apparent that the limits for heavy metals and chemicals are based on criteria which is at best vaguely defined. The limits are typically set at background concentrations or at levels where no adverse effects are known to have occurred. This is basically due to the fact that relatively little reliable scientific data exists on the adverse effects and allowable levels for heavy metals and chemicals in the environment.

On the other hand, as you are well aware, the effects of ionizing radiation are probably the most studied and best understood of all environmental insults. The allowable concentrations of radioactive isotopes, as given in

the EPA drinking water regulations, are based on a maximum allowable whole body or internal organ dose of 4 mrem per year for most isotopes. The exceptions to this are the bone seekers, such as radium-226, whose limits are based on a cancer risk of approximately 1 x 10^{-6} per person per year. This risk is about the same as that due to a whole body exposure of 4 mrem per year.

In order to establish a firmer basis, mainly for the toxic heavy metals and chemicals, the National Academy of Sciences (NAS) recently completed a study (Na77) which reviews in detail the current interim EPA drinking water limits. The major conclusions of that study which are of interest here is that: (1) the limits for at least arsenic and lead are probably too high to protect the public adequately, (2) the limit for chromium should probably be based only on the Cr^{+6} or chromate ion, and (3) the limits for radioisotopes are probably adequate to protect the public. This NAS study also confirms that for at least one of the chlorinated hydrocarbons, which has a limit set by the EPA drinking water regulations, the expected cancer risk from that limit is comparable with the cancer risk for the allowable levels of radioisotopes. Consideration of these additional factors tends to confirm the assumption that the limits prescribed by the EPA drinking water regulations provide equal protection from toxic heavy metals, chemicals and radioisotopes. They are therefore probably the best currently available guidelines with which to compare the relative toxicity of hazardous and radioactive waste.

By using the limits as set by the EPA safe drinking water regulations and by extending the concept of a radiotoxic hazard index* to include toxic heavy metals and chemicals, we can develop a relative toxicity index, expressed as m^3 of water required to dilute a given quantity of toxic material to safe drinking water levels. With this index we can make a direct comparison of the relative potential hazard of radioactive versus hazardous waste. This comparison per metric ton of the various types of waste is shown in Figure 1. It should be noted that this is only a comparison of the potential for groundwater contamination, and as such does not include possible mitigating factors which may retard or enhance its ultimate uptake by humans.

When examining Figure 1 in detail, a few important points should be mentioned. In order to provide a baseline for comparisons with background, the average toxic heavy metal and naturally occurring radium concentration (Co77) per metric ton of the earth's crust is shown in broken lines on the figure. The radioactive waste potentials are taken from expected concentrations as given in various NRC reports (Nu78a) (Nu76). The transuranic potential of spent fuel/HLW was developed by comparing its cancer risk with that of radium. The increase in the potential long term toxicity of low-level radwaste above the essentially stable component primarily due to iodine-129 is caused by the ingrowth of the daughter products of uranium-238 which is disposed of as source material. The hazardous waste potential is a composite of various EPA reports (Ba76) (Ca75) (Gr75) (Ja76) (Ve75) which have reported the toxic material concentration of the waste products of those industries listed in Table 1. The toxic heavy metal content accounts for the non-decaying portion of the hazardous waste curve. The

*The radiotoxic hazard index is defined as a measure of the amount of water required to dilute a certain quantity of radioisotopes to permissible concentration and is determined by dividing the initial quantity in curies by the permissible concentration in Ci/m^3 (Nu76).

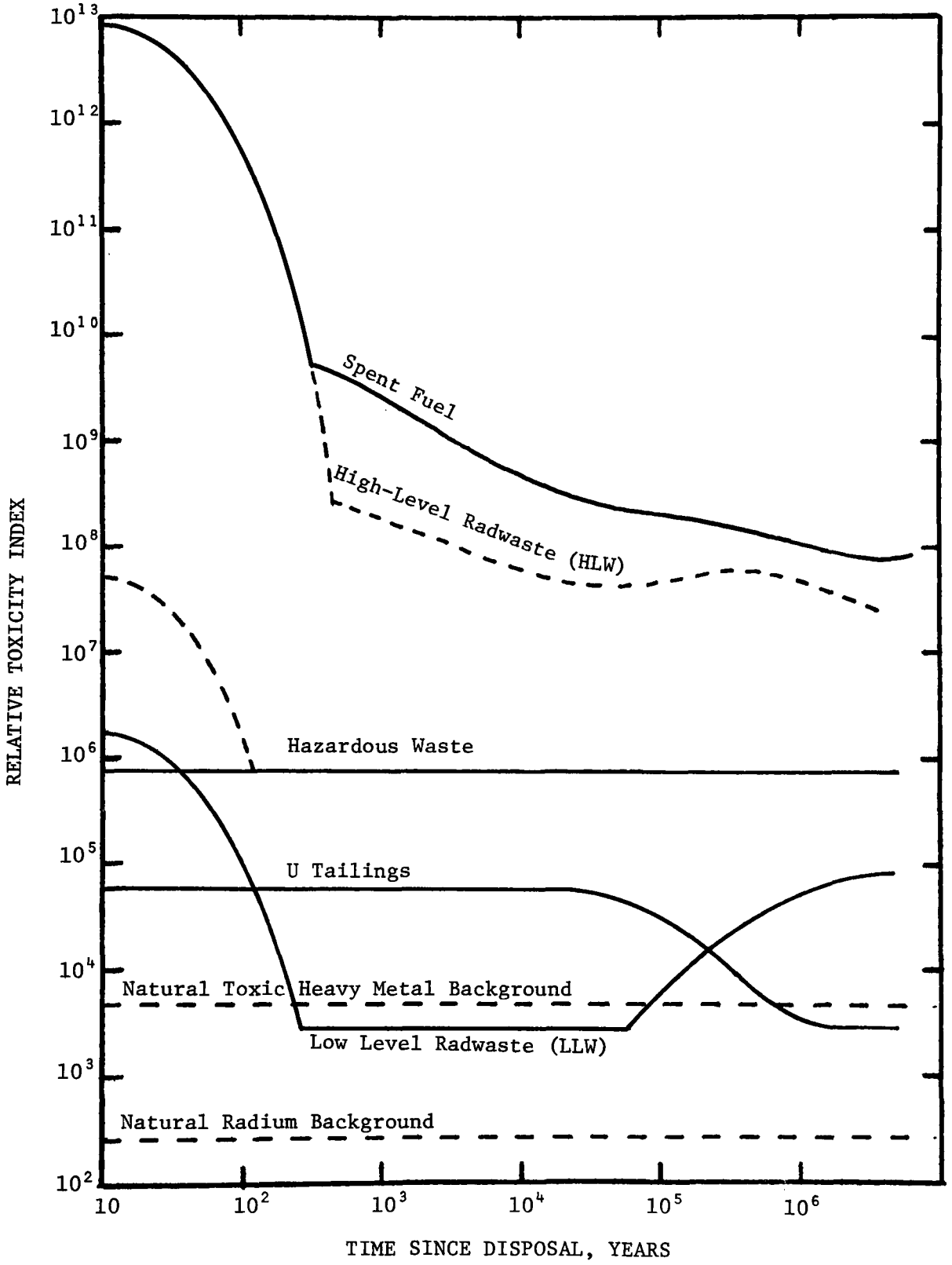

Figure 1: Relative Toxicity of A Typical Metric Ton of Hazardous Versus Radioactive Waste

broken line decaying portion is an upper bound to account for the highly dangerous chemicals which do not currently have safe drinking water limits. The exceptions to this are a few chlorinated hydrocarbons, whose limits are used as representative of all the highly dangerous chemicals.

Since Figure 1 only compares a typical metric ton of each type of waste, the total annual production rate of each must also be considered to give a truly indicative perspective as to the total potential for groundwater contamination from radioactive and hazardous wastes. These annual production rates for 1977 are shown in Table 1. Including this consideration, the total relative toxicity of hazardous versus radioactive waste is shown in Figure 2. This comparison shows that based on current production rates the total long term potential of hazardous waste is comparable with that of spent fuel and several orders of magnitude higher than the long term potential of low-level radwaste.

Comparison of Wastes Produced by Alternate Energy Sources

It is well known by the public that most of the radioactive waste is produced by the nuclear fuel cycle. On the other hand, it is a little known fact that most other alternate energy sources also produce large quantities of hazardous waste. This can be shown, by looking at the two major short and long term competitors with nuclear, namely coal and solar.

The coal fuel cycle can produce hazardous waste due to the fact that during combustion most of the toxic heavy metals which are contained in coal in essentially background concentrations are carried over into the fly ash and bottom ash. Here they are concentrated by about a factor of ten and become much more available for leaching due to chemical changes. In addition, large quantities of sulfur dioxide scrubber sludges are generated by most modern coal fired plants. This sludge is produced in a form that is not only difficult to dispose of properly, but may be hazardous due to its sulfur content or from the carry-over of some toxic heavy metals.

Solar energy facilities, and for that matter almost all renewable energy sources, do not generate any waste products during operation. However, because these energy sources are very dilute, much larger quantities of materials compared to the more conventional energy sources are required for the manufacturing of equipment which is necessary to collect this diffuse energy. With this requirement for materials, mainly primary metals, comes the generation of large quantities of hazardous waste. This waste is produced mostly from the smelting and refining of the ores and the finishing of the metal surfaces.

Table 2 compares the total lifetime radioactive waste production of a 1000 MW nuclear plant with the lifetime hazardous waste production of a 1000 MW coal plant and 1000 MW of solar power. The solar contribution is assumed to be half flat plate collectors (heating and cooling) and half solar thermal (electric). The potential relative toxicities of these wastes are then compared graphically in Figure 3. This figure suggests that the wastes from coal and solar power are of comparable hazard to the wastes from the nuclear fuel cycle over the long term.

It should also be pointed out that the above indicated waste potential is maximized for nuclear since the quantity of uranium tailings is the greatest for the once through LWR fuel cycle. However, since the coal and solar potentials

Table 1: Total Quantity of Hazardous and Radioactive Waste Produced in the U.S. in 1977

Type of Waste	Quantity Produced (Metric Tons/Year - Dry Weight)
Hazardous (by type of industry)(1)	
Primary metals production	4.73×10^6
Organic chemicals and pesticides	3.50×10^6
Inorganic chemicals	2.30×10^6
Electroplating and metal finishing	1.32×10^6
Petroleum refining	0.72×10^6
Other industries	1.23×10^6
Total Hazardous Waste	13.80×10^6
Spent Reactor Fuel(2)	9.40×10^2
Low-Level Radwaste(3)	1.09×10^5
Uranium Mill Tailings(4)	7.30×10^6

(1)En77
(2)Nu78
(3)Mu76
(4)Assumed to be 0.1% uranium ore (Nu76)

Table 2: Quantities of Lifetime Waste Generated by 1000 MW of Various Alternate Energy Sources

Type of Waste and Energy Source	Lifetime(1) Generated Quantity (Metric Tons - Dry Weight)
Nuclear	
Spent Fuel(2)	1.05×10^3
Low-level radwaste(3)	5.67×10^4
Uranium tailings(2)	8.16×10^6
Coal(4)	
Fly ash and bottom ash	2.03×10^6
Scrubber sludge	3.57×10^6
Solar	
Hazardous waste(5) from the manufacturing of solar collectors(6)	4.28×10^4

(1)Assumed to be 30 years in all cases
(2)Nu76
(3)Mu76
(4)Assuming Northern Appalachian coal which has been washed (Dv77)
(5)Hazardous waste production rate and constituents (Ba76) (Ca75)
(6)Material required for 1000 MW solar installation (In78)

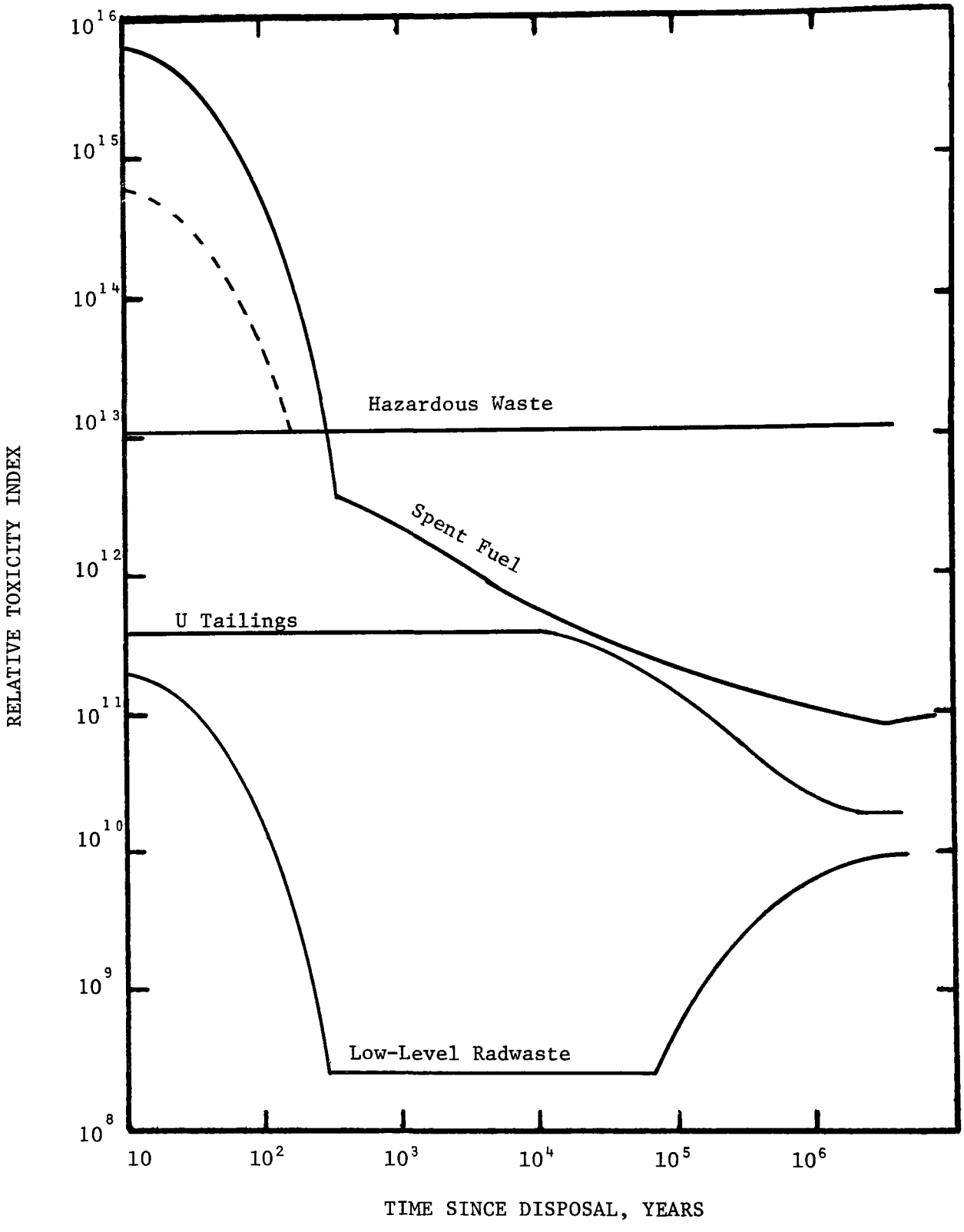

Figure 2: Relative Toxicity of the Total Quantity of Hazardous Versus Radioactive Waste Produced in 1977

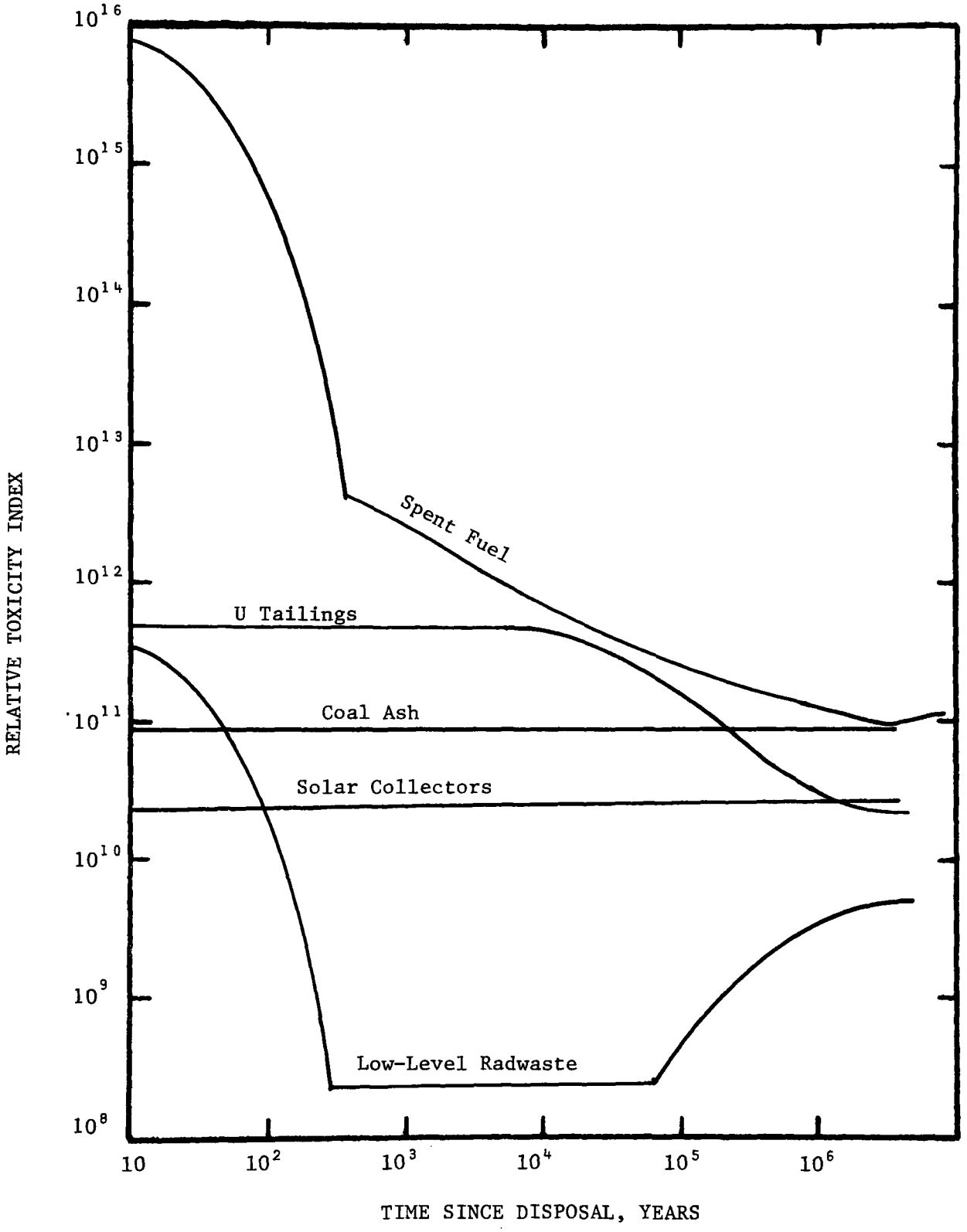

Figure 3: Relative Toxicity of the Lifetime Radioactive Waste From A 1000 MW LWR Nuclear Fuel Cycle Versus the Lifetime Hazardous Waste From A 1000 MW Coal Fuel Cycle and the Manufacturing of Collectors for 1000 MW of Solar Energy

only account for toxic heavy metals, the additional consideration of naturally occurring radioactive isotopes could make the relative toxicity of their wastes at least equal to the uranium tailings potential. This follows from the fact that some coals may contain over 40 ppm uranium (Nu78a) and by assuming all copper solar collectors and noting that some copper ores may contain 50 ppm uranium (Co78).

Other Factors Which Make the Hazardous Waste Problem Difficult to Solve

In 1976 Congress passed the Resource Conservation and Recovery Act (RCRA) which mandates that EPA develop regulations which will insure proper handling and disposal of hazardous wastes. These regulations, which have just recently been published in draft form, (En78a) are about nine months behind the Congressionally mandated deadlines, and are not scheduled to be finalized until January 1980. In the meantime, EPA has estimated (En77) that about 90% of the total quantity of hazardous waste which is currently generated is being handled and disposed of in a manner which may not be adequate to protect public health and the environment. In fact, it has recently been reported by the EPA that there are about 32,000 sites in the U.S. which contain potentially dangerous amounts of hazardous waste, of which at least 638 may contain quantities which could cause "significant imminent hazards" to public health (En78b).

There are many potentially troublesome problems inherent in the management of hazardous waste which may make it a more difficult task than the safe management of radioactive wastes. A few of these potential problems are the following.

Hazardous waste is much more difficult to account for and control at the source. This is mainly due to the fact that hazardous waste generators will not be regulated as stringently as are all radioactive waste generators. Also complicating this is the fact that many industrial processes are proprietary and therefore even information concerning the composition of the waste products may be very difficult to obtain. This problem with accountability then leads to inexpensive solutions such as illegal dumping in waterways and sewers.

The constituents of hazardous waste are much more difficult to detect and measure accurately than are radioisotopes. Very sophisticated and time consuming procedures are required to achieve low detection thresholds, and even then they do not compare with the thresholds which are achievable for radioisotopes. In addition, some toxic chemicals may change to other toxic compounds and therefore an individual analysis must be initiated for each suspected contaminant. There is no simple screening procedure such as a radioactive gross alpha or beta measurement, which is very useful for quickly determining the effectiveness of radioactive waste management.

Much of the industrial waste, particularly from primary metals production, was considered nonhazardous by the EPA reports on hazardous waste production if it did not leach toxic materials in appreciable quantities when mixed with distilled water. This was true even if the industrial waste contained large quantities of toxic materials. In addition, there were many other toxic constituents of the industrial and hazardous waste, such as cyanide, phenol, oils and greases, which were not considered in the comparisons of potential toxicities because they currently are not included in the EPA drinking water regulations. By contrast, the radioactive waste potentials include all the radioisotopes which are generated regardless of their chemical or physical forms.

Like the situation with low-level radioactive waste disposal there currently is a very limited number of licensed hazardous waste disposal sites in the U.S. This fact is proving to be very troublesome to hazardous waste generators since currently they produce about 200 times more waste by weight than the generators of low-level radwaste. In addition, the publicity of recent problems with some of the abandoned hazardous waste disposal sites may make future licensing of these sites extremely difficult.

Even considering all of the above, probably the most disturbing inequality in the situation is that, in the opinion of many, the proposed EPA regulations for hazardous waste disposal will be much less stringent than even those which are being developed for low-level radioactive waste disposal. These inconsistencies follow from a long standing tradition whenever radioactive materials are involved.

As a prime example, the proposed hazardous waste regulations consider solid waste to be hazardous only if the contained toxic materials can be leached from the wastes in concentrations exceeding 10 times the EPA safe drinking water standards. The regulations go on to exempt those producers which generate less than 100 kilograms of waste per month from all the regulations except those dealing with disposal in approved facilities. (The waste producers themselves are to determine if they exceed this threshold.) In contrast with this are the recently proposed EPA criteria for all radioactive wastes (En78a) which precludes the establishment of any "de minimus" concentration of radioisotopes for a waste to be deemed radioactive. Furthermore, a recent NRC proposed rule change to 10 CFR 20 (Nu78b) would discontinue the practice of burial of small quantities of licensed materials at sites other than licensed disposal facilities.

Causing the situation to be even more inconsistent is the fact that radium-226, which has the lowest NRC permissible drinking water limit of any radioisotope, will be regulated under the proposed EPA rules as a hazardous waste. Incredibly, the EPA is proposing as a "de minimus" level for radium-226 (a concept already disallowed for radioactive wastes), the same limit which the NRC has deemed unacceptable for unlicensed disposal of this same radioisotope.

Summary

In the past radioactive waste disposal has been viewed by a majority of the public in a complete vacuum because it was felt that the problems involved were not comparable to any other environmental insult. However, when considering some of the problems which are inherent in the safe disposal of hazardous waste, it must be concluded that radioactive waste disposal is not a unique problem for this country to solve. In fact, when directly comparing the two problems, radioactive waste seems to be the more manageable and therefore the easier to implement successfully.

The responsible Federal agencies should also take note of the inconsistencies which prevail in their regulatory efforts and begin treating equal hazards equally. The intent here is not to ease the strict requirements that will be necessary for safe radioactive waste disposal, but to determine if the standards for hazardous waste disposal will be adequate in comparison with those stringent standards. This will assure that public health and safety is being equally protected from all toxic waste, regardless of the source.

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A PROFILE OF INSTITUTIONAL RADIOACTIVE WASTES GENERATED IN 1977

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Abstract

A national survey of radwaste volumes and characteristics generated by large medical and academic institutions in 1977 was performed. This is a followup to a survey which obtained 1975 data. The estimated total waste volume generated by the survey population in 1977 was 7771 m^3 (274,400 ft^3). These data and those from the previous survey show that the volume is increasingly linearly and consistently accounts for approximately 11% of the total volume of low level radwaste buried commercially. Most of the waste shipped by respondents (78%) was shipped to the shallow land burial site at Barnwell, South Carolina.

Included in this profile of institutional radwastes are: a report of the principal radionuclides present as waste contaminants, and a breakdown of waste by form; an estimate of the effect of mechanical compaction of dry waste and a review of the extent to which alternative disposal methods are used by the study population for the various waste forms.

Performed under USNRC Contract #NRC 04-76-0344.

Introduction

Among the most significant sources of non fuel cycle low level radwastes, are medical and academic licensees. In 1975 we showed that the larger medical and academic institutions accounted for approximately one-third of the non fuel cycle wastes buried that year in the commercial shallow land burial sites (An78).

We are at present concluding a followup to the 1975 survey, after having obtained waste data from the same population for the calendar year 1977. As in the preceding survey, the study population was selected from NRC and agreement state licensee lists, to meet the following criteria:

- Large hospitals with ~450 beds or more, excluding mental health and other extended care facilities.
- Schools of medicine (hereafter referred to as medschools).
- Four year colleges and universities within excess of 5000 students (hereafter referred to as colleges).

The data that we sought from these institutions was generally compiled by the individual within the institution who managed the license. Therefore, we actually surveyed radiation control programs.

Radiation control programs are often, but not always, consolidated within the various parts of an academic institution and also with neighboring institutions. To get a better idea of the fraction of the study population represented by the data, we divided the population into entities. We define a population entity as a college, a medschool or a hospital. Some members of the study population may contain more than one entity due to the consolidation of radiation control programs. Based on responses, the population was categorized into the following groups of entities:

- Hospitals only
- Hospitals and medschools
- Hospitals, medschools and colleges
- Medschools only
- Medschools and colleges
- Colleges only

The numbers of entities within each category for the total population and for respondents are shown in Table 1. The percent responses in the right most column were used for data extrapolation. Overall the reponses accounted for 59% of the hospitals, 66% of the medschools, and 56% of the colleges for which the totals are 348, 116, and 323, respectively. Geographically, we obtained data from 48 states and the District of Columbia, only Nevada and Alaska were not represented.

Results

The estimated total waste volume shipped for burial by the study population in 1977 was 7771.1 m^3 (274,400 ft^3). This constitutes approximately 11% of the total low level waste volume shipped during the study year (Ho79). The institutional waste fraction reported in 1975 was also 11% suggesting similar growth rates (An78). We also obtained shipment volume data for 1976 in the current study; and have data for the years 1972 to 1975 from the previous survey. The latter volume estimates have been recomputed using the present population breakdown scheme, and differ somewhat from that previously reported (An78). These six data points are plotted in Figure 1. The waste volume appears to be increasing linearly: the correlation coefficient for the least squares fitted line is .96. The equation of the line is:

$$V = m x + b$$

where:

V = volume in m^3

$m = 639.74$

X = (year - 1900)

$b = -41621.63$

Five commercial burial sites were in operation during the study year. The breakdown of the data reported by respondents by destination is shown in Table 2. The dominant burial site is again Barnwell, South Carolina, which received 78% of the volume. This contrasts with 1975 when 30% of the volume was shipped to this site (An78).

In 1977, respondents shipped a total of 372 Ci of non sealed source activity for burial. Of this total, 99.1% was identified; the breakdown of the identified activity by half-life category is shown in Figure 2. Most of the activity in the category with half-life of 90 days or more is Tritium (91.5%). Of the remaining activity in this category, 5.7% is Carbon 14. Again, it can be concluded that the significant nuclides contaminating the waste of the study population are ^{3}H and ^{14}C. The major part of the remaining activity shipped by this population in 1977 would be undetectable if one assayed that waste today.

Sealed sources shipped for burial were treated separately. The total reported sealed source activity is broken down by nuclide in Table 3. Again, the majority of the activity shipped is Tritium, generally consisting of spent neutron generator targets. Most of the remaining sealed source activity is ^{137}Cs, ^{192}Ir and ^{226}Ra.

The types of waste shipped by the study population were categorized by waste form:

- Dry, solids - syringes, vials, test tubes and other disposable labware, absorbent papers, gloves, etc.
- Adsorbed liquids - aqueous and organic liquids dispersed in an adsorbent media.
- Liquid scintillation vials - full scintillation vials packed in adsorbent material.
- Biological wastes - predominantly carcasses of laboratory animals, also including animal tissues, bedding, excreta and labeled culture media.

The breakdown of the wastes by type are shown in Figure 3. It is noteworthy that the scintillation vial waste fraction has increased from 29% in 1975 to 43% in 1977 (An 78).

Volume reduction techniques are frequently touted as an important adjunct to waste processing in this population. In 1975, 23% (N = 29) of the respondents who shipped waste compacted dry waste prior to shipment (An79), in 1977 24% (N = 47) compacted.

To obtain an estimate of the impact of the use of mechanical compactors for reduction of dry waste volume, the dry waste fractions of those institutions compacting waste was multiplied by the reported compaction ratio. A hypothetical total volume was then obtained by summing the "expanded" dry waste with the other waste fractions. The total volume thus obtained is 41% greater than that without compaction. This is undoubtedly an optimistic assessment of the impact of waste compaction, but could be considered to be an upper limit. It can be stated, however,

that those compacting dry waste are the largest waste producers; therefore, a relatively small number of compactors will have a major effect.

Of the 342 coded responses, 316 or 92% received unsealed sources of radioactive material in 1977 and could be considered to be potential waste producers. Of these, 292 indicated that radioactive wastes of some type were diposed of in the study year. Although we only quantitated wastes shipped for burial, data regarding the use of alternative disposal methods was also obtained. Table 3 reports the numbers and percentages of respondents disposing of the following waste types:

- Liquid scintillation vials and fluids
- Other (non scintillation fluid) organic liquids
- Aqueous liquids
- Biological wastes

  Dry solids

The percentage breakdowns, showing the disposal methods utilized for each waste type, are shown in Table 4. The alternatives shown are not exclusive, often more than one disposal method was used for a given waste type.

The disposal of scintillation vials and fluid, generally contaminated with ^{14}C and ^{3}H, perhaps present the greatest disposal difficulties. Although shipment for burial is the most prevalent alternative, the use of other methods is common. A relatively common method is to dispose of scintillation fluids via the sanitary sewer and dispose of the empty vials in the common refuse. Much of the dry wastes which appear in the common refuse are apparently contaminated with short-lived nuclides; they are thrown there after the nuclide is "decayed off".

The use of incineration as a disposal alternative is most common with biological wastes, and is used to a lesser extent with the other waste types.

In the survey questionnaire we asked respondents to give the total cost, excluding labor, for shipping waste in 1977. The total cost for the 197 respondents who shipped waste in 1977 was approximately $1.4 million. To put this figure in perspective the costs per cubic foot of wastes averaged $10.90 ($\pm$\$13.20) and ranged from a maximum of $125 per cubic foot to $0.38 per cubic foot shipped. Obviously, the shipment costs vary widely depending on location of the institution, its shipment frequency, volume and other factors.

This presentation is essentially a very broad stroke profile of the data in the 1977 survey. In the near future, we expect to examine closely the differences in waste type among the various types of institutions within the population. We are currently pursuing a waste stream approach which we hope will correlate the waste types and disposal alternatives to the type of institution and the specific uses of radioactive materials.

This work was performed under contract to the U.S. Nuclear Regulatory Commission (NRC 04-76-0344).

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TABLE 1. BREAKDOWN OF SURVEY POPULATION BY CATEGORY

	NUMBERS OF ENTITIES		
	TOTAL	RESPONDING	% RESPONSE
HOSPITALS ALONE	267	142	53.0%
HOSPITALS AND MEDSCHOOLS	77	55	71.4%
HOSPITALS AND MEDSCHOOLS AND COLLEGES	124	103	83.1%
MEDSCHOOLS ALONE	13	7	53.8%
MEDSCHOOLS AND COLLEGES	52	22	42.3%
COLLEGES ALONE	254	134	52.8%

TABLE 2. BREAKDOWN BY DESTINATION OF WASTE VOLUMES SHIPPED

DESTINATION	PERCENT OF TOTAL VOLUME
SHEFFIELD, IL	8.42%
MAXIE FLATS, KY	1.06%
BEATTY, NV	6.66%
BARNWELL, SC	77.64%
RICHLAND, WA	5.50%
OTHER SITES	0.71%

TABLE 3. SEALED SOURCES SHIPPED FOR BURIAL

SEALED SOURCE	N	TOTAL ACTIVITY (Ci)	PERCENT TOTAL ACTIVITY
^{3}H	10	71.550	80.9%
^{137}Cs	11	11.080	12.5%
^{192}Ir	7	1.194	1.3%
^{226}Ra	17	3.435	3.9%
Others	33	1.207	1.4%

TABLE 4. WASTE TYPES DISPOSED BY STUDY POPULATION

WASTE TYPE	%DISPOSING*
SCINTILLATION VIALS AND FLUIDS	71%
OTHER ORGANIC (NON SCINTILLATION) FLUIDS	48%
AQUEOUS LIQUIDS	78%
BIOLOGICAL WASTES	59%
DRY SOLIDS	86%

*Percent of those who disposed of radwaste in 1977, (total N=292).

TABLE 5. DISPOSAL ALTERNATIVES*

WASTE TYPE	PERCENT OF RESPONDENTS USING DISPOSAL METHOD**					
	SEWER	COMMON REFUSE	INCINERATION	SHIP FOR BURIAL	BURY ON SITE	TRANSFER TO OTHER INST.
SCINTILLATION VIALS AND FLUIDS	29.0%	23.7%	4.3%	80.7%	7.7%	1.0%
OTHER ORGANIC LIQUIDS	46.0%	-	2.9%	74.8%	7.9%	1.4%
AQUEOUS LIQUIDS	68.9%	-	-	54.4%	5.3%	1.8%
BIOLOGICAL WASTES	7.0%	4.1%	25.7%	71.9%	9.4%	2.3%
DRY SOLID WASTES	-	27.5%	12.0%	72.1%	7.2%	6.4%

* Alternatives are not exclusive; many institutions disposed of a waste type by more than one method.

** Percent of those who disposed of that type of waste.

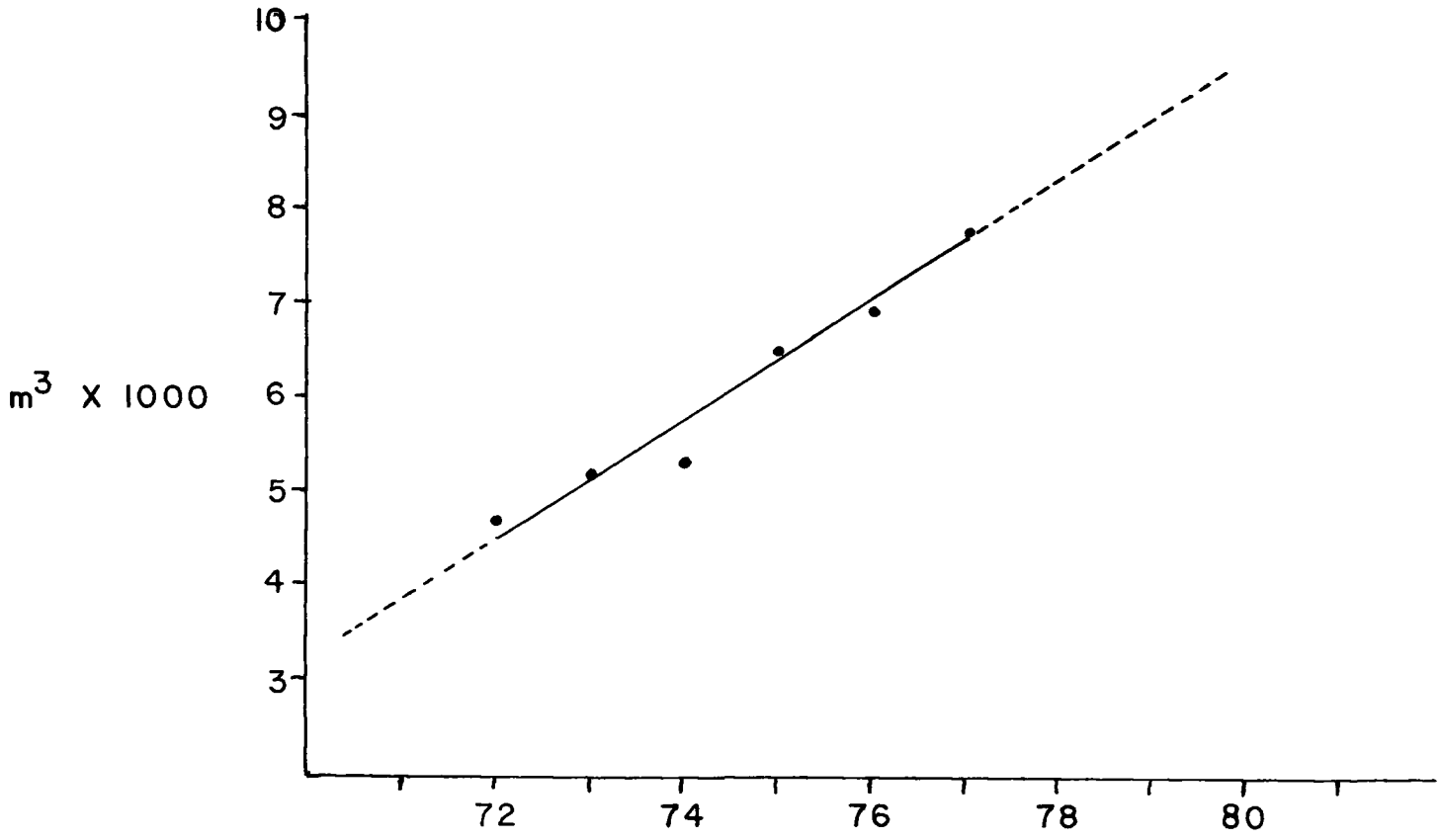

FIGURE 1. INSTITUTIONAL RADIOACTIVE WASTES SHIPPED FOR BURIAL 1972 - 1977

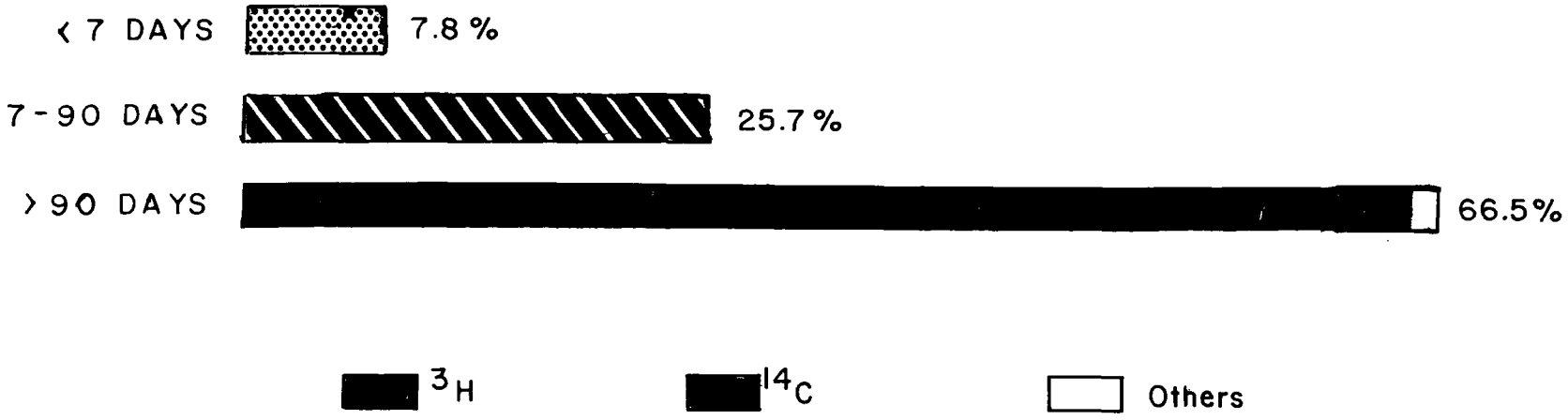

FIGURE 2. NON SEALED SOURCE ACTIVITY SHIPPED FOR BURIAL BROKEN DOWN BY HALF-LIFE CATEGORY

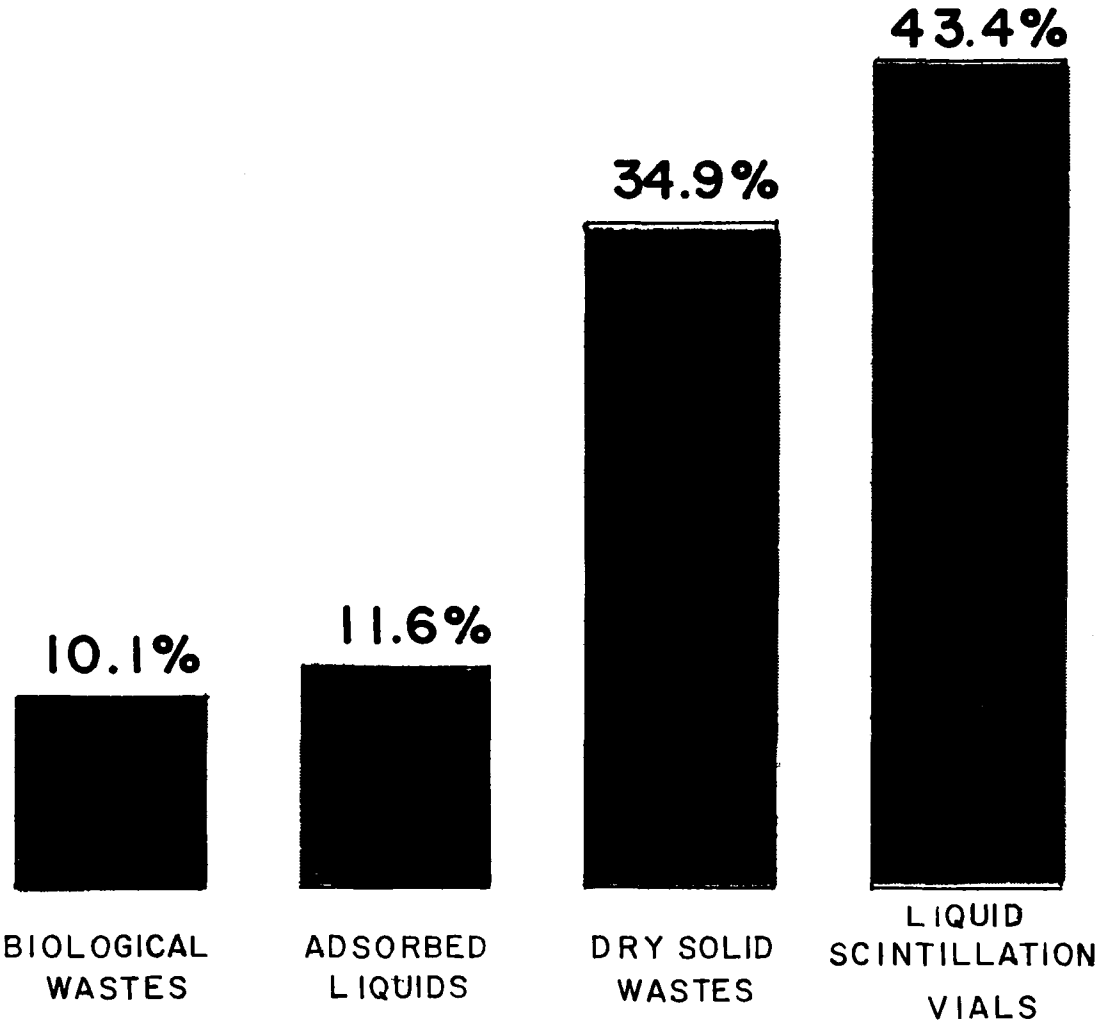

FIGURE 3. BREAKDOWN OF WASTES SHIPPED FOR BURIAL BY FORM

HEALTH PHYSICS CONSIDERATIONS IN ACCELERATOR DECOMMISSIONING AND DISPOSAL

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ABSTRACT

There are perhaps as many as 1,200 particle accelerators in the United States, ranging in size from the very small Cockroft-Walton and electron linear accelerators to the multi-GeV research synchrotrons. When an accelerator has reached the end of its useful life the radioactivity induced in the components then presents several disposal problems. At least fifty accelerators produce significant induced activation, and several hundred more are capable of producing fluxes of neutrons which could result in activation of various components of the accelerator facility. This is generally of low level except for the very high energy/high intensity machines.

In most cases, the induced radioactivity is confined to relatively few parts of the machine and the disposition of these is through the normal radioactive waste channels without complication. However, some machines leave a legacy of low level induced radioactivity in massive components. Examples of massive items are large magnets, shield blocks and beam stops of concrete, earth or iron, and even the walls and floors of the building itself. These need to be dealt with in a manner so as to pose no potential health hazards to persons in the vicinity of the public at large.

The disposition of radioactive waste from major past decommissionings, including the Cambridge Electron Accelerator (CEA) and the Brookhaven Cosmotron, will be discussed. The extent of the induced radioactivity problems in decommissioning of smaller accelerators will also be discussed.

INTRODUCTION

Over the past several years, there has been increasing concern over the accumulation of radioactive materials at various scientific, industrial and other facilities in the United States, and increasing pressure to ascertain what is being done to assure that any potentially serious waste disposal problems are not being overlooked. Of course, the nuclear power industry has the major share of the problem and, therefore, commands most of the attention. However, there exists, in addition to the power industry, a sizable variety of users of radioactive materials in medical products, industrial products, as sources for radiation processing and sterilization and, as by-products, from the operations of particle accelerators. The subject of nuclear facility decommissioning has recently been addressed by the Comptroller General in a June 2, 1977, report to the Congress entitled "Cleaning Up the Remains of Nuclear Facilities - a Multi-billion Dollar Problem" (Co77). The primary thrust of the report is toward the nuclear power industry; however, other aspects of the problem, which include isotope usage and accelerator facilities, are recognized as potential problems.

The Department of Energy has initiated a comprehensive study of the quantities and types of radioactive materials in existence both at its

existing facilities and at the facilities formerly utilized as part of the Manhattan Engineer District/Atomic Energy Commission (MED/AEC) program. The Division of Environmental Impact Studies at Argonne National Laboratory was requested to perform a comprehensive study of the portion of the decommissioning problem that concerns the dismantling and disposal of all types of particle accelerators in the United States.

The questions that this study relates to are contained in the report to the Congress and are as follows (Co77):

"What is the extent of the decommissioning problem for accelerators?

"Are standards needed for induced radiation?

"What should be the limits on acceptable radiation levels?"

Only the first of these questions is directly pursued in the review effort.

In order to develop information relative to accelerator decommissioning, the following tasks have been initiated. Since some of these tasks are not yet complete, only progress can be reported in some areas. Also, the aspects of the study that do not relate to the waste problem will not be discussed. The tasks are:

1. Compile a census of the accelerator population in the United States and categorize the machines according to their potential inventories of active material.

2. Review the history of past accelerator decommissionings in regard to technological, environmental, health and economic aspects.

3. Survey the quantity of radioactivated material at existing particle accelerator facilities with emphasis on the high energy machines (e.g., AGS, Bevatron, FNAL, LAMPF, SLAC, ZGS, etc.).

Lower energy machines have not been ignored, since there is a concern about the neutron production by medical electron accelerators.

4. Develop a generalized decommissioning scenario for the various categories of accelerators. Included in this effort will be such things as estimate of costs, determination of the volume of radioactive waste to be expected, and the estimation of the degree of reusability of various classes of components.

5. Study the alternative methods of decommissioning as they relate to accelerators. The alternatives are:

   a) complete dismantling and removal of the accelerator and its building to a waste burial site, temporary storage, or immediate recycle.

   b) removal of the accelerator only to a waste burial site, temporary storage or immediate recycle, leaving the building for research or office space.

c) mothballing in place with eventual performance of a) or b) above.

d) entombment in place.

Mothballing and entombment may, in fact, be necessary in cases where radiation levels around accelerator components are comparable with the nuclear reactor environment.

PRODUCTION OF INDUCED RADIOACTIVITY

Induced activity is due to both the interactions of the primary accelerated beam with a target and to interactions of any secondary particles produced by the primary beam interactions. The governing factors for material in the vicinity of the target, generally are related to the neutron production capabilities of the accelerated beam. The pertinent factors are as follows:

1. Species of particle(s) accelerated. Generally deuterons and tritons generate more secondary neutrons than protons, while electrons generate much fewer, other factors being equal.

2. Energy of the accelerated particles. The induced activity is a function of the particle energy and the appropriate reaction cross sections once the threshold energy is exceeded.

3. Beam intensity or current. Induced activity is proportional to the number of particles accelerated.

4. Duty factor. The ratio of operating time to shutdown time determines the actual value of the effective long-term average beam intensity.

5. Primary usage of the accelerator. This has a direct impact on the other parameters, e.g., a high energy research accelerator or an isotope production facility is generally run at maximum attainable currents and as continuously as operational and fiscal constraints allow. At the other extreme, some research accelerators are used exclusively for short-term intermittent sample irradiations or for low intensity scattering experiments.

Table 1, found in the National Bureau of Standards Handbook 107 (NBS70), summarizes the potential for induced radioactivity expected for various accelerated particles and energies.

Until the last few years, medical and industrial accelerators (almost exclusively electron accelerators) have been of energies less than 10 MeV. However, the use of medical linacs in the energy region above 10 MeV is now on the increase. The rate of growth of medical linac installations is presently in the range of 200 to 250 units per year with an estimated eventual population of approximately 2000 units in the United States (Ro78). Heavy ion and neutron therapy are both receiving increased attention from medical researchers and may eventually add significantly to the neutron producing accelerator population. The number of compact neutron generators used as analytical tools now exceeds 200 in the United States. Neutron generators were specifically not part of this review because they are generally of very

small size with very low levels of activation.

PAST DECOMMISSIONINGS

A number of accelerators of all types have already been decommissioned. Some of the earliest cyclotrons and betatrons were simply disassembled and the components reused for other purposes or sold as scrap metal. The beam energy and intensity of the early machines were generally very low so that any induced radioactivity would have been essentially undetectable except by very sensitive survey techniques. There are essentially no records regarding these very early decommissionings.

The major decommissionings which have been reviewed in some detail are the BNL Cosmotron, the Cambridge Electron Accelerator at Harvard, the 142 inch Carnegie-Mellon synchrocyclotron, and the University of Rochester synchrocyclotron. The Brookhaven Cosmotron, a 3-GeV proton synchrotron, was shut down December 31, 1966. The machine was kept in standby condition for 1 year after shutdown during which time the experimental area was dismantled and much of the equipment was transferred to the Alternating Gradient Synchrotron (AGS) facility at BNL. After the year had elapsed authorization to proceed with the dismantling of the Cosmotron itself was granted by the AEC, the owner of the machine. The removal of reusable equipment and components was performed on a spare-time basis by BNL personnel. The actual disassembly of the synchrotron ring magnets was done by contract technician labor over a 3 or 4 - month period. The one-year waiting period resulted in a significant reduction of the induced activity levels. The iron magnet segments, copper windings, vacuum chambers and vacuum pumps were placed in the radioactive material storage yard where most of them remain today. The presence of induced radioactivity in these items precluded their release to scrap dealers. A number of the magnet blocks have been used as shielding at the ZGS and more recently by the Fermi National Accelerator Laboratory. Because of the difficulty in removing the epoxy resin and fibre glass insulation bonded to the copper magnet windings, very few of these have been reused.

The Cambridge Electron Accelerator operated for the last time on May 31, 1973. The initial decommissioning plan, based upon the conditions of the contract between Harvard and the AEC, included complete removal of the accelerator along with all underground structures and other buildings and restoration of the land to its original status. A modified plan was agreed upon and only the underground accelerator tunnel was scheduled for demolition and removal. The accelerator itself was dismantled and the magnets sent to BNL for possible reuse in an AGS experiment. They are still in storage at BNL. The rest of the experimental equipment was either absorbed by other programs at Harvard or by other laboratories and universities. The demolition of the underground tunnel was more difficult than expected because of the heavy reinforcing and was not completely accomplished. Since the shock waves from the wrecking ball were endangering the nearby private residences, segments of 6 foot thick reinforced concrete retaining walls were left buried in place. The site presently has the appearance of a vacant lot. Radioactive waste volumes and radiation intensities were minimal from this effort since this was an electron accelerator with low average beam current. A radiation survey made within 24 hours after the final shutdown indicated only one area with radiation intensity as high as 100 mR/h at contact (Sh73).

The Carnegie-Mellon Synchrocyclotron decommissioning is an example of a case in which it was desired to save the building, belonging to the University, and offer it for sale on the open market. The complete accelerator and all associated equipment were disassembled and removed from the building, and the building itself decontaminated to levels which did not exceed two times natural background. In order to remove the magnet iron (~ 310 tons) from the accelerator vault, it was cut up in place with torches using appropriate contamination control measures. Approximately half of the iron was transferred to MIT and the other half to the Los Alamos Meson Facility to be used for shielding. The coils are in storage at BNL. After removal of all the accelerator hardware, it was found that in certain areas of the concrete walls and floor, there were detectable levels of induced radioactivity which necessitated the removal of a layer of concrete several inches thick. This effort generated several truck loads of concrete and several hundred barrels of dirt and rubble which required transportation to a commercial low-level waste burial ground.

RADIOACTIVITY LEVELS AT ACCELERATOR DECOMMISSIONING

In discussing the types and quantities of radioactive materials that are generaged by an accelerator D&D effort, this report will focus on accelerators with beam energies of tens of MeV and higher. There are a limited number of components in any given accelerator that will become highly radioactive. These will be portions of the primary beam transport systems, target stations and beam stops which are directly struck by the accelerated beam as part of normal operations. For the very high energy and high intensity accelerators, those components and structures in the vicinity of points of primary beam interaction will also be highly activated by secondary particles. In addition to these localized "hot spots", the main structure of the accelerator, primarily the magnet iron along with its copper or aluminum windings for circular machines, and copper drift tubes and tanks for linear accelerators will all contain a highly variable volume distribution of activation products. It can generally be assumed that all the permanent components within the primary shield enclosure or vault will have some degree of induced radioactivity. Additionally, the walls of the shield vault itself may contain significant quantities of induced radioactivity. Other components can contribute to the radioactive waste volume. For example, cooling water systems may accumulate and concentrate radioactivated corrosion products. Vacuum systems, ventilation systems, and target transport systems also could present additional radioactively contaminated material.

The detailed distribution of isotopes produced in accelerator materials is a complicated function of the type and energy of the incident particle(s), beam intensity, beam transport efficiency, target elements, and the cross-sections for the various reactions involved. To illustrate the situation, Figure 1 shows production cross-section curves for a Bi target bombarded with protons of three different energies (Pa73). For machines of particle energy in the 10's MeV, the isotopes produced are clustered very near the mass number of the target material. For machines with higher energies (100's of MeV), spallation and high energy fission processes result in a much broader spectrum of product isotopes. The curve for 500-MeV particles shows two humps in the production curve. The hump at the higher mass numbers is due to spallation reactions and the hump at approximately one half of the target mass number

results from high energy fission. In the GeV region, the production of all isotopes becomes almost equally probable, with all types of production reactions becoming energetically possible. Similar distribution curves would be obtained with any other heavy element. In this report, the term "target material" is used in the broad sense meaning any material with which the primary beam interacts.

Most of the major components of particle accelerators consist of either iron or copper with minor amounts of other materials. Major exceptions to this are the use of depleted uranium and lead for certain shielding and collimation applications, and the use of aluminum for magnet windings. Activation products in iron and copper are primarily short-lived with half-lives of less than a few days. Table 2 summarizes the important long-lived isotopes found in activated accelerator components. It is seen that ^{60}Co, ^{22}Na and ^{54}Mn will be the controlling isotopes.

The longer an accelerator is operated, the closer will be the approach to saturation activity for the long-lived products. The total quantity of the long-lived activities present at shutdown depends on the gross long-term average operating conditions of the accelerator. Short-lived activity is due only to the operations during that operating period just prior to shut off.

Qualitatively, there is an initial rapid decay of the short-lived components in the mix followed by a slower decay governed by the long-lived isotopes. Some generalizations can be made in regard to the shape of the decay curve. For positive ion accelerators with beam energies less than approximately 100 MeV, the initial decay is very rapid and essentially lasts for about a week after shutdown. The long-lived decay tail is then controlled by the decay of the ^{59}Fe and ^{65}Zn with half-lives of 45 and 245 days respectively. In higher energy accelerators, i.e., above 100 MeV, the slow decay component is controlled by such additional isotopes as ^{54}Mn, ^{57}Co and ^{60}Co, with half-lives ranging from 270 days to 5.26 years. ^{60}Co is the longest lived gamma emitter and is generally found in the iron and copper, which make up the bulk of the mass of most large particle accelerators. A few accelerators have used aluminum windings for the main magnet coil and ^{22}Na, with a 2.6 year half-life, may be the controlling long-lived activity.

A generalized decay curve of accelerator-induced radioactivity can be derived using an analytical expression developed by Sullivan and Overton (Su65) for high energy accelerators.

$$D(t) = a\ G\ \ln\left[\frac{T + t}{t}\right]$$

where $D(t)$ - the dose rate
a - machine dependent parameters
G - cross section and other physical constants
T - length of irradiation time (age of the accelerator)
t - length of decay time after shutdown

Figure 2 shows the relative value of $D(t)/aG$ for two values of T as a function of decay time t. The upper curve is for T = 25 years and the lower is for T = 100 days.

Two empirical decay curves are also plotted for comparison. These curves were obtained as follows. Samples of reagent grade copper and iron were exposed to the particle flux near a high beam loss area of the ANL 12.5-GeV synchrotron for a three-month period. The decay of the gamma activity of the samples was then monitored using a large NaI gamma counter for a period of four years. The theoretical 100-day irradiation curve is seen to be in reasonable agreement with the decay curves observed for the copper and iron. Data obtained from the LBL 184 inch synchrocyclotron on the decay of radiation levels during an 11-day undisturbed shutdown in 1971 falls between the curves for Cu and Fe (Ri71). It appears, then, that it would be reasonable for planning purposes to use a decay curve derived using the actual age of an accelerator in the Sullivan and Overton formula. It can be seen from the upper curve, for an assumed 25-year old accelerator, that ~30% of the radioactivity will remain two years after shutdown. From this point on, the decay could be assumed to be due primarily to the ^{60}Co in the material. Based on this model, a block of magnet iron from a high energy accelerator reading 100 mR/h at 1 day after shutdown will read 30 mR/h after two years of decay. Then, using the 5.26-year half-life of ^{60}Co, it will take 61 years to reach a radiation intensity of 0.02 mR/h, which is on the order of two times natural background. It can, therefore, be argued that 100 years would be a reasonable upper limit on the lifetime of accelerator-induced activity.

ESTIMATION OF QUANTITY OF RADIOACTIVE MATERIAL

Proton Accelerators

Estimates can be made of the total quantity of radioactivity contained in a proton accelerator by using approximations discussed in a paper by Gollon (Go76).

The method for estimating the total radioactivity in an accelerator is based on the fact that an equilibrium for constant conditions of operation, the decay rate of radioactivity is equal to the production rate. The production rate is related to the accelerated beam intensity and energy. As a first approximation, for accelerators of energy on the order of a few hundred MeV, the saturation activity is numerically equal to the beam intensity. Using the basic relationships of

$$1\ \mu a = 6.025 \times 10^{12} \text{ protons/sec and}$$
$$1\ Ci = 3.7 \times 10^{10} \text{ dis/sec},$$

we can calculate a value of 160 Ci/μA. This radioactivity is distributed among the various machine components and the experimental apparatus which intercepts the beam. For example, the fraction of the beam that results in the activation of a cyclotron magnet is probably in the range of 1% to 10%. Therefore, we could expect to see approximately 1.6 to 16 Ci of saturation activity from operations with a 1-μA beam of protons. For machines of lower energies, this relationship will overestimate the long-lived activity present.

For higher energy accelerators, the phenomenon of spallation, or star production, in which a multiplicity of secondary particles are produced, can result in activation products. For example, a beam of 10^{12} 400-GeV protons/sec is estimated to produce approximately 10 kCi of long-lived activity (Go76), which yields a ratio of about 1600 Ci/μA of beam.

Electron Accelerator

The estimation of radioactivity induced by high energy electron beams and the associated bremstrahlung has been reviewed in a paper by Swanson (Sw79). The photons produced by electron beams produce radioisotopes by photonuclear reactions, the quasi-deuteron effect and photo spallation at high energies. The components which absorb most of the electron and photon beams are subject to the highest activation levels.

Table 3 is a summary of the long-lived isotope production data calculated for electron beams of energy 35 MeV and greater. The percent of saturation attained for each isotope after an assumed 25-year operating life has been calculated and is shown along with the expected production in terms of curies per kilowatt of beam power. The long half-life isotope ^{26}Al is included to illustrate the fact that products with very long half-lives will generally not be a problem.

SUMMARY

Table 4 partially summarizes the results of this study to date on the estimation of the eventual quantity of radioactive material that will require proper disposal. Emphasis is upon the largest accelerators with the specific goal of arriving at the total masses and volumes of radioactive waste that they represent. Most of the accelerator material involved is iron or copper. The shielding included in these totals is primarily steel or concrete. While the total quantities involved are considerable, it must be remembered that all accelerators will not be decommissioned and dismantled as waste at one time.

In keeping with the philosophy of maintaining all radiation exposure to levels which are As Low As Reasonably Achievable (ALARA), it can be argued that any radioactivity which would add to natural background should not be released to the world at large for unrestricted use. It can also be argued that dwindling natural resources, including metals such as copper and iron, need to be reutilized to the maximum extent possible for both ecologic and economic reasons. Consequently, if the recycling of these materials is contemplated, all potential exposure pathways need to be considered, including the internal exposure possibilities (e.g. from melting, grinding, cutting, or welding operations) as well as the external exposures potential from usage of the material. In addition, there are a number of other potential detrimental effects aside from health effects which would need careful consideration. Two examples of these are the possible effects on the photographic film industry and the disruption of analytical techniques used in geology, archeology, medicine, crime detection, etc.

It seems that it would be possible to mothball in place or temporarily store the accelerator components long enough to allow the induced activity to decay to levels which are essentially indistinguishable from natural background by sensitive detectors such as gas proportional counters or μR meters. Based on the results of this study, it is probable that this period of time would not be longer than 100 years for the major components of the present high intensity machines. Millions of tons of material would be a definite burden to bury and perhaps be a treasure worth eventual recycle.

TABLE 1

RELATIVE PRODUCTION OF INDUCED RADIOACTIVITY IN PARTICLE ACCELERATORS

Accelerated Particle	Energy Range (MeV)	Induced Radioactivity In	
		Target	Vicinity
Electrons	<1.67	None	None
	1.67 - 10	Limited	Very slight
	>10	Probable	Suspect
Protons, Helium ions	<1	Limited	None
	1 - 10	Limited	Suspect
Deuterons, Tritons	Any Energy	Limited	Suspect
All ions of light atomic weight	>10	Certain	Suspect

TABLE 2

RADIOACTIVE ISOTOPES OF CONCERN IN ACCELERATOR DECOMMISSIONING

Isotope	Half Life	Produced In
^{22}Na	2.6 a	Aluminum
^{54}Mn	303 d	Iron
^{57}Co	270 d	Iron
^{60}Co	5.26 a	Stainless steel & copper
^{65}Zn	245 d	Copper

TABLE 3

INDUCED RADIOACTIVITY FROM ELECTRON BEAMS

Target Material	Isotope Produced	Half Life	Saturation* Activity, Ci/kW	% of Saturation for 25 Year Operation	Actual Expected Activity, Ci/kW
Concrete	^{22}Na	2.6 a	0.1	99.9	0.10
Aluminum	^{22}Na	2.6 a	0.28	99.9	0.28
"	^{26}Al	$.4 \times 10^{5}$ a	8.8	0.0023	2.04×10^{-4}
Iron	^{54}Mn	303 d	0.59	100.0	0.59
Nickle	^{57}Co	270 d	5.9	100.0	5.9
"	^{60}Co	5.26 a	0.1	96.2	0.096
Copper	^{60}Co	5.26 a	0.65	96.2	0.63
"	^{63}Ni	92 a	0.45	17.0	0.077

*Data from (Sw79)

TABLE 4

ESTIMATED MASS & VOLUME OF ACCELERATOR RADIOACTIVE WASTE

Type of Accelerator	Approximate # of Machines	Estimated Mass of Active Waste, tons per accelerator	Estimated Volume, ft^3/accelerator
Cyclotrons <20" diam. & e^- linacs	1000	$\leq$ 30	$\leq$ 60
Cyclotrons 20" to 50" diam.	15	30 - 50	260 2200
Cyclotrons 50 to 100" diam. e^- linacs $\geq$ 600'	16	250 800	2200 - 7000
Cyclotrons diam. > 100"	4	800 - 2300	7000 - 20,000
Proton synchrotrons 60' - 200' diam.	3	2300 16,500	20,000 - 140,000
Proton synchrotrons > 200' diam. proton linac > 500' length	3	16,500 220,000	140,000 - 2×10^6

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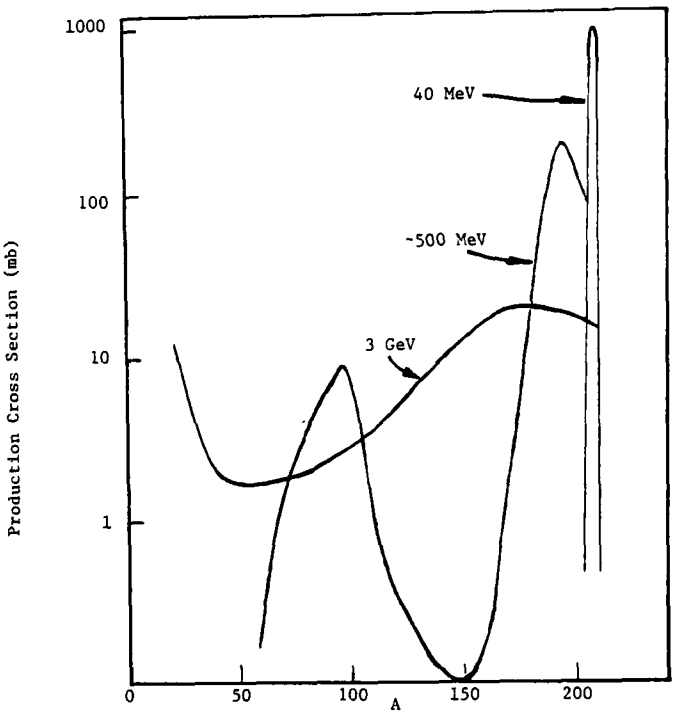

Fig. 1. Mass Yield Curves for Protons on Bismuth (Pa73)

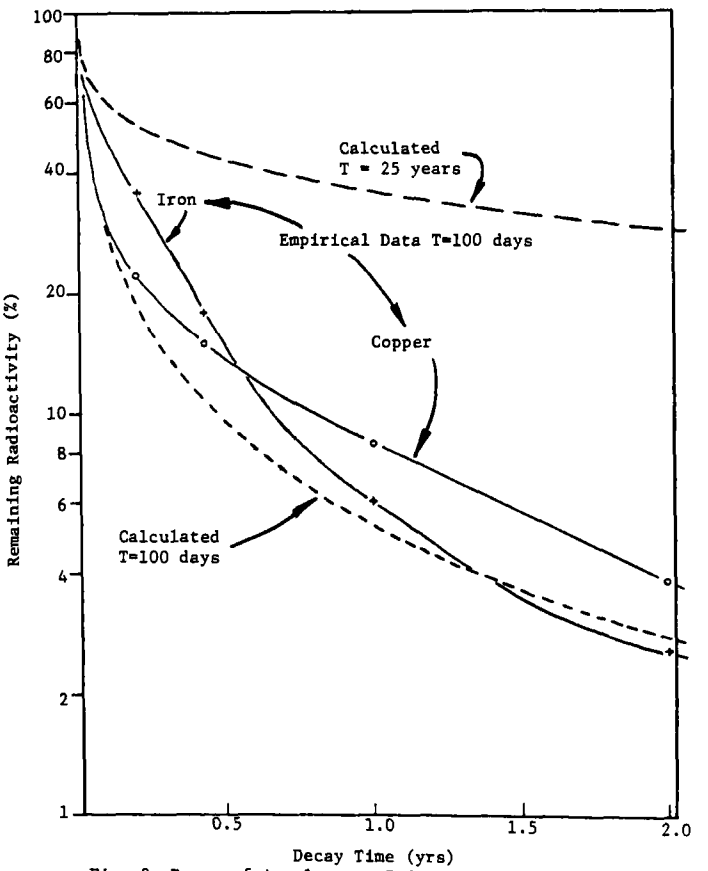

Fig. 2. Decay of Accelerator Induced Radioactivity

LOW-LEVEL RADIOACTIVE WASTE FROM RARE METALS PROCESSING FACILITIES

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Abstract

In the past year, problems of disposal of byproducts, tailings, and wastes from rare metals and thorium producing facilities have received the attention of radiological agencies. Many of the raw ores used by these processing facilities were rich in natural radioactivity and the residues of production were often not disposed of properly. Mill tailings from the uranium mining industry have only recently come under federal regulation. It can be expected that similar attention will be focused on the environmental impacts of the rare metals processing industry as illustrated by interest in the problems of Parkersburg, WV, and Albany, OR. One other site in Akron, NY, does not appear to be an immediate problem, but its situation is typical of the many yet unsurveyed inactive rare metal facilities in the country. The radiological problems presented by and tydecontamination activities which may be required of these rare metal facilities are examined.

Introduction

The federal government has recognized that companies which process thorium and uranium ores require regulatory controls in order to protect man and the environment from unnecessary radiation. The recent passage in November, 1978 of the Uranium Mill Tailings Bill (H.R. 13650) demonstrates the government's recognition that the front-end of the uranium fuel cycle, i.e., mining and milling of uranium, had been neglected. The bill defines procedures for a remedial action program at inactive mill sites and regulations for active mill sites.

Companies which provide titanium, phosphorus, rare earths, and rare metals for industrial and chemical use are not normally regarded as possessors of large quantities of radioactive materials. In fact there appears to be a historical laxity in documenting the processing and waste disposal activities of these industries. A recent EPA publication reviews the available literature on technologically enhanced natural radiation due to mineral extraction industries (Bl 78). It is only recently that phosphate industrial wastes have been listed as hazardous radioactive wastes in the U.S. Environmental Protection Agency's proposed Hazardous Waste Guidelines and Regulations (Co 78). This paper will review the situations at the existing Teledyne Wah Chang, Co., Inc. located at Albany, Oregon, and the former Carborundum Corp./Amax Specialty Metals, Inc., facilities located at Parkersburg, West Virginia, and Akron, New York, in order to show the extent of the radioactivity problem at rare metals processing facilities and the need to identify for radiological review other rare metal and rare earth processing sites.

As shown in Figure 1, the unusual grouping of rare earth and rare metal processing industries stems from their common ore origin. The ores used in rare earth and metals processing are byproducts of mining for titanium ores,

since the ores for the specific processing are seldom found in economically mineable rock. The principal domestic areas for raw materials are Florida and Georgia, although mining has occurred in western and other southeastern states. Outside of the U.S., major deposits are located in Australia, Canada, Brazil, South Africa, Sri Lanka, India, and Mexico. Often ores with higher specific mineral content were imported, such as Nigerian zircon sand for hafnium processing and Australian zircon sand for zirconium processing.

The beach and fluvatile sand deposits from these areas are rich in marketable ilmenite, rutile, monazite, and zircon. Monazite commonly incorporates thorium and uranium as well as rare earths due to similarity in geochemistry and electronic structure. Ilmenite and rutile are ore materials for titanium processing, monazite is the principal ore for rare earth processing, and zircon the principal ore for zirconium and hafnium processing. Generally, these beach or placer sands are treated to produce heavy mineral concentrations containing the zircon, rutile, ilmenite, monazite, and other marketable minerals. The concentrates may then be treated by various combinations of gravity, electrostatic or electromagnetic methods to separate the individual minerals. Monazite being slightly magnetic can be separated from zircon by electromagnets. The purity of the zircon product (or conversely the degree of monazite contamination of the product) is obviously a function of the degree of separation effort. Initial treatment usually is provided at or near the mine site. As a rule of thumb, the sand deposits are usually but not always processed primarily for the titanium content in the form of rutile and ilmenite. The zircon and monazite fractions are then byproducts which are treated separately to extract zirconium and rare earths, respectively. Thorium is then a further byproduct of the rare earth processing of the monazite portion. This has been the major source of thorium up to the present.

For zirconium metal production, zircon sand is usually processed to minimize the monazite content since the phosphate content of the monazite has a deleterious effect on the metallurgical process. This in turn should mean a lower thorium and uranium content in the metallic zirconium wastes than in foundry wastes where the monazite content of the zircon sands should be of less importance to the process. However, Wagstaff has reported levels of radium-226 from the uranium decay chain to be about 100 pCi/g in incoming zircon sands at both foundries and metallic zirconium production facilities (Wa 78). As Table 1 shows, the uranium and thorium content of monazite concentrates varies depending on where the ore is mined. The amount of monazite in the zircon sand also depends on how well the separation facility removed the monazite before shipping to the use point. At the use point (such as a zirconium metal manufacturing plant), the manufacturer may find it necessary to further separate monazite from the sand. Low-level radioactive wastes may be generated at each separation point. The natural concentration of uranium and thorium decay series products in the sands are low but the industrial processing to obtain the specific minerals concentrates in the waste residues of these normally occurring radioactive materials. The disposition of these waste residues is the subject of this paper.

Case Studies of Three Facilities

The zirconium and hafnium processing method was developed by W. J. Kroll for the U.S. Bureau of Mines. The bureau established a pilot plant in 1947 at Albany, Oregon, to extract zirconium and hafnium using the Kroll process and a purification plant in 1951 at Oak Ridge, Tennessee, to produce high puri-

ty, low hafnium, reactor grade zirconium. The zircon sand is mixed with graphite or coke and is fused in an electric furnace to produce a mixture of carbonitrides of zirconium and hafnium. The carbonitrides are chlorinated in a vertical shaft furnace and the gaseous chlorides of zirconium and hafnium are collected in a nickel condenser. The zirconium and hafnium chlorides are reduced in the Kroll process to the metals by reaction under an inert atmosphere with magnesium. The end product, commercial grade zirconium sponge, will contain about 2% hafnium suitable for non-nuclear use. Current industrial practice uses zirconium tetrachloride produced by chlorinating zircon directly instead of the carbonitride (MF 75). In order to produce reactor grade zirconium, i.e., that containing about 0.3% hafnium, the commercial grade zirconium sponge is dissolved and the hafnium is solvent extracted to hafnium thiocyanate using methyl isobutyl ketone. The hafnium is precipitated as a hydroxide, calcined to about 99% hafnium oxide. The resulting zirconium sponge is crushed, compacted into consumable electrodes, and vacuum melted in an inert atmosphere to ingot. Further product purity is achieved by applying the deBoer-vanArke refining process. A similar procedure is applied to the hafnium solvent extraction in order to obtain high purity hafnium metal. The residues generated by the extraction processes contain graphite, coke, unreacted silicates, and non-volatile silicates.

Wah Chang Corp. began operating the Bureau of Mines' Albany, Oregon, facility in 1955. Today it is one of the major producers of reactor grade zirconium and hafnium metals. Concern over the environmental and health safeguards at the facility grew when explosions were encountered during digging operations near the facility's industrial waste piles. Apparently the explosions were caused by rapid combustion of the zirconium in the waste piles. At the same time the Radiation Control Section of the Oregon Department of Environmental Quality (DEQ) became concerned that the large chlorinated residue piles may be a radiological problem. A gamma radiation survey showed maximum reading of 1200 uR/h. When the Oregon DEQ checked the radium concentration of the piles, it found that the Ra-226 ranged from the original zircon sand concentration of about 60 pCi/g to over 1300 pCi/g. One water sample taken within the residue pile showed Ra-226 concentration of 45,000 pCi/L, hence the concern of a potential ground water contamination. These radiological parameters for the rare metals chlorinated residues can be compared with those for uranium mill tailings.

Most uranium mills in the U.S. typically processed an average uranium ore grade of about 0.15-0.35% uranium which would give expected radium-226 concentrations in the mill tailings ranging from 420-980 pCi/g. Individual tailings samples at a given mill may have concentrations that are more or less than these values by as much as a factor of five or so.

Due to Oregon DEQ's work at the Wah Chang facility, Oregon limited the volume and radium content of chlorinated residue which the facility may accumulate onsite before disposal in an out-of-state facility is required and mandated all users of zircon sand to file an application for a radioactive materials license. The criteria for release to an unrestricted area are 57 uR/h, 30 pCi/L of Ra-226 in effluent, and 0.03 WL of radon* (Wa 78). Of the twenty-

* One Working Level (1 WL) is a unit describing any concentration of short-lived decay products of radon-222 in one liter of air which results in the release of 1.3×10^5 MeV of potential alpha energy.

six potential users of zircon sand, the state estimates only four will require specific licensing. Similarly, Utah has restricted onsite accumulations to no more than 100 tons of chlorinated residue and no more than 3 curies of Ra-226.

Efforts to determine the extent of possible radiological problems are more difficult for sites which have ceased rare metals processing activities for several years either due to changes in site ownership or unfavorable economic climate. Locating residue piles and sludge ponds, estimating amount and origin of ores processed, and determining processing and waste disposal activities must rely on historical records which are vague or nonexistent.

In the mid-1950's the responsibility for zirconium production was shifted to private enterprise when the civilian nuclear power program was established. In order to meet the increased demands for reactor grade zirconium, Carborundum Corp. which operated a facility in Akron, New York, expanded its production capacity by building a facility in Parkersburg, West Virginia. The plant's designed capacity was 600 tons annually; it began operations in 1957. In the mid-1960's, Amax Specialty Metals Co., Inc., became a partner and in 1967 obtained full ownership of the company. The Parkersburg site was sold in 1977 to L. B. Foster Company, a steel pipe fabrication plant. As a result of Foster's plan to expand its buildings, highly flammable waste materials were encountered during backhoe operations. In investigating the causes of the explosion, it was discovered that zirconium and thorium may have been buried onsite, and that Amax Specialty Metals had not adequately terminated its license with the U.S. Nuclear Regulatory Commission (NRC) for possession of radioactive materials. The NRC estimates that two million pounds of zircon ore, mainly from Nigeria, were processed at the Parkersburg plant since 1957. A radiological survey of the site shows gamma radiation to range from a background level of 10 uR/h to 150 uR/h. Soil samples show concentrations of thorium-232 and its decay products to range from background level of 1 pCi/g to 10,000 pCi/g. The thorium contaminated area is limited to a few acres of the 100 acre site. The NRC's tentative clean-up goal of 5 pCi/g above background of thorium-232 with a three to four foot overburden and deed restrictions on excavation was developed based on an assessment of the long-term hazard due to thoron (radium-220). A radiological survey of Parkersburg by a contractor to Amax Specialty Metals estimates 50,000 cubic yards of soil may need to be removed. Some disposal alternatives being considered are burial at a disposal facility, burial onsite in a clay lined cavity with land use restrictions provided for the burial area, and ocean disposal. Whether there are other locations onsite where zirconium and/or thorium are buried may never be known since records on waste disposal and processing activities are incomplete.

The Akron, New York, zirconium and hafnium processing facility was the pilot plant for the Parkersburg, West Virginia, facility and presently is owned by Amax Specialty Metals Co., Inc. Processing activities by Carborundum Metals Co., Inc., began in 1953 at the Akron site to produce hafnium free zirconium under an Atomic Energy Commission (AEC) contract.

The plant's designed processing capacity was 162 tons of zirconium annually. Although the plant had a contract with the AEC to produce zirconium metal, there does not appear to be any AEC, NRC, or NYS (an agreement state) license for byproduct material other than for research and development purposes. An industrial license with the NYS Department of Labor (DOL) for x-ray

and gamma sources was in effect from 1960 through 1978.

During the summer of 1978, the EPA Region II radiation office queried the NYS DOL and Department of Environmental Conservation about the status of the Akron, New York, plant. The EPA was concerned that a situation similar to the Parkersburg, West Virginia, plant may exist at the New York plant due to their operating history. EPA was informed that Amax Specialty Metals Co., Inc., had contracted Atcor, Inc., to perform a radiological survey of the Akron site as the initial step in terminating its industrial license with the NYS DOL. The June, 1978, survey showed radiation levels ranging from a background level of 7 uR/h to 1500 uR/h outside, with some building measurements up to 40 uR/h (Le 78a). The extent of processing activities at the site is not well known due to incomplete records. There were areas where magnesium and zirconium residues were found but no pyrophoric incidents occurred. A single soil sample from an area with an external radiation reading of 1500 uR/h was analyzed and showed the soluble portion contained the radioactive material, but no further radiochemical analyses were performed. The elemental composition of the sample indicates it may be Nigerian ore, the principal ore processed at the Parkersburg plant. Surface soil samples were taken at locations with above background gamma radiation and were spectroscopically analyzed. The range of concentrations of Ra-226, Pb-214, Bi-214, Ac-212, Tl-208, and K-40 are shown in Table 2 (Le 78b). For these isotopes, the background concentrations are less than 1 pCi/g except for K-40 with concentration of 12 pCi/g. The limited results in Table 2 indicate levels of thorium and uranium chain nuclides elevated above expected background levels.

The monazite fraction of zircon ores typically runs about 3-10% thorium dioxide (ThO_2) content with a tri-uranium octoxide (U_3O_8) content up to 0.41%. Zircon sands of 96.7% pure zircon are currently imported and quoted on a minimum basis of 65% zirconium oxide (ZiO_2). Hence the maximum monazite content of incoming zircon sands should be less than 3.3%. Assuming a 10% ThO_2 content in the 3.3% monazite fraction of the zircon sand, the overall ThO_2 content of the zircon sand should be less than 0.33% or less than 300 pCi/g. Nigerian sands are reported to range from 0.4 to 7% ThO_2. Based on this and on the assumption that the monazite is some lesser fraction than 3.3% of the non-zircon portion of the sands, then the values of 120-150 pCi/g for the thorium chain nuclides do not seem unreasonable. Similarily, using a 0.41% U_3O_8 content in the monazite fraction and a maximum 3.3% monazite content, one would estimate a maximum uranium or radium-226 concentration, assuming equilibrium, of about 38 pCi/g, which seems to be in probably fortuitous agreement with the maximum measured values of 35 pCi/g for Ra-226. The higher K-40 values are certainly higher than the expected background values of about 12 pCi/g. However, one stage of the hafnium purifying process uses a potassium chloride molten mixture. If this plant used this process and if some of the molten mixture were spilled, it could account for the higher K-40 values since the potassium chloride probably contains about 400 pCi/g.

Between June and September, 1978, Amax Specialty Metals removed soil from areas with high gamma radiation levels. About 25 cubic yards of soil were shipped to the commercial low level burial site in Barnwell, South Carolina. Two of the three lagoons or infiltration ponds were excavated to a three feet depth and the material was disposed in a nearby hazardous chemical landfill. The radioactivity of the excavated material is not known since no analyses were performed prior to disposal.

In September, 1978, Atcor resurveyed the site after clean-up efforts and made recommendations for additional decontamination to reduce levels to "as low as reasonably achievable." However, it is not known to what extent these recommendations were pursued by the site owner. It is not known whether any attempts were made to identify the source of the slightly elevated levels of radioactivity in the buildings.

In December, 1978, the NRC performed a survey of the Akron site (St 78). The survey identified one area near a ridge with gamma radiation of twenty times background. Areas near the lagoons and the tube mill building had levels ranging from background to ten times. Analyses of the soil samples from these three locations indicate thorium concentrations in the range 6.0-19 pCi/g with background concentration of 1.2 pCi/g. The one air sample showed no Rn-220 daughters above background levels. Gamma radiation levels in the buildings were within twice background, indicating little contamination after removal of the gamma gauges.

The site could have been released for industrial use with little clean-up necessary in order to meet the NYS DOL Industrial Code Rule 38 that no gamma radiation levels exceed 250 uR/h at the surface and that source material in soil be less than 0.05% by weight, which is 5,000 pCi/g of Th-232. Due to experience at rare metal processing facilities in Albany, Oregon, and Parkersburg, West Virginia, Amax considered a more stringent clean-up program to meet the goal of "as low as reasonably achievable." In general, the clean-up program has been successful, although EPA Region II would have liked to have seen the levels reduced to twice background and to 5 pCi/g for Ra-226 and Th-232. A record of processing and disposal activities at the site would have greatly assisted in answering questions concerning the possibility of any buried radioactive materials.

Discussion

It appears from Figure 2 that the amount of zircon ore imported into the United States has been increasing steadily since 1930. The imports account for approximately 50% of the annual U.S. consumption of zircon concentrates; the remainder is attributed to domestic production and stock piles. Australia supplied about 60% of the imports before 1950, and over 95% after 1950. Brazil contributed about 20% during the period 1930 to 1950. In total, the U.S. consumed about 300,000 short tons of zircon before 1950 and 1.3 million short tons thereafter.

The potential radiological problem can be likewise divided into two periods. Prior to 1950, most of the Australian zircon was imported as a black sand mixture containing zircon (40-75%), ilmenite (14-43%), rutile (7-18%), and monazite (2-8%) ores (MY 36). No attempts were made to separate the ores until the sand mixture reached the processing facility. In 1948, the Commonwealth Government declared its intent to purchase and stockpile monazite ore. As a result, future shipments of sand had most of the monazite ore separated in Australia before export.

In order to provide a conservative estimate of the radiological content in the sands imported before 1950, the sand mixture is assumed to be composed of 8% monazite ore. Assuming the ThO_2 content in the monazite fraction to be as high as 10%, then the ThO_2 content in the beach sands could reach 0.8% or about 800 pCi/g. Similarily, assuming the U_3O_8 content in the monazite frac-

tion to be as high as 1%, then the U_3O_8 content in the beach sand could reach 0.08% or about 200 pCi/g. After 1950, the Australian ore had the monazite fraction separated to some extent, hence the sands contain a minimum of 96.7% zircon. Assuming the ThO_2 content in the monazite fraction remains 10%, then the ThO_2 content in the beneficiated sands could be as high as 0.33% or about 300 pCi/g, the U_3O_8 content could reach 0.03% or about 100 pCi/g. Subsequently, the rare metals processing facilities which operated prior to 1950 may have greater radiological problems with their chlorinated residues than those which use ore obtained after 1950.

We would expect that any user of zircon sands would receive some monazite in the zircon sands, since the separation of ilmenite, rutile, zircon, and monazite, as indicated in Figure 1, is often incomplete. Hence some small fraction of monazite, containing thorium and uranium will be present in any industry which extracts titanium, chromium, tantalum, etc., from beach sands. The monazite and hence the amount of thorium and uranium decay chain products will vary with the sand origin and the degree of ore beneficiation. In fact, facilities which need only the zircon, ilmenite, or rutile fraction of the beach sands and insist on high purity ores may not have as great a radiological problem as facilities which need only the monazite fraction or use sands with little ore separation.

Producers of reactor grade zirconium have rigid specifications for thorium and uranium content and generally require high purity zircon which implies low radioactivity content. To achieve this, the zircon is either purchased as high purity material or further processed at the rare metal producing plant to achieve purity. For metal production, then, the radioactivity remains in the wastes while very little goes with the metal product. On the other hand, the purity of zircon sands consumed at foundries is not critical, hence these sands may have the highest radioactivity content. Manufacturers of refractory materials, producers of milled or ground zircon, and ceramics manufacturers will most likely have some portion of the radioactive content incorporated into the products due to the manufacturing process.

In reviewing information from the annual Minerals Yearbook for 1929 through 1975, over twenty states were identified to have some facility which processed beach sands in zirconium, hafnium, and rare earths production or used beach sands in foundry processes. Table 3 provides a breakdown of the states according to the type of processing or use activity. Facilities presently operating in these areas of activity can be fairly easily identified and evaluated to determine where these facilities dispose their chlorinated wastes and whether the sands or concentrates used by the facilities have any appreciable monazite fraction. For facilities which have ceased operating or changed their ownership or their products, such an evaluation is more difficult.

Since only limited data and in some cases no data are available for radioactivity and exposure levels associated with industries such as discussed above, it seems apparent that considerable work needs to be done to assess the environmental and health impact of such industries.

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TABLE 1: Thorium And Uranium Composition In Monazite Concentrates (Weight Percent)

	ThO_2	U_3O_8
Australia	7-8	1
Brazil	6-7	0.2
India	9-10	0.3
Madagascar	9	0.4
South Africa	6	0.1
United States	4-5	0.4

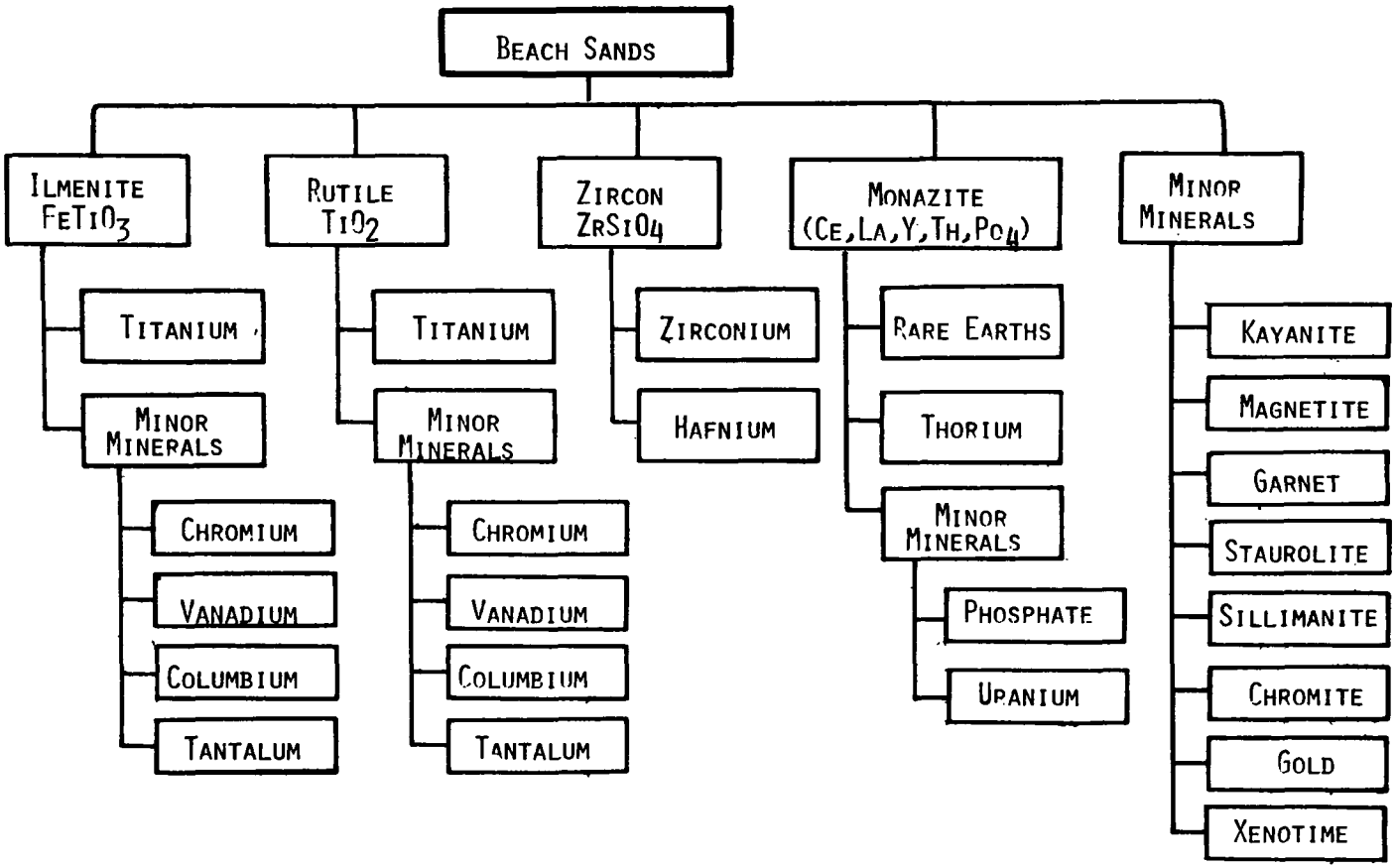
TABLE 2: Radioisotopic Concentrations In Soils From The Akron, New York Rare Metals Processing Facility (Le 78b)

Radioisotope	Range of Concentrations (pCi/g Dry)
Ra-226	2.1 - 35
Pb-214	0.66 - 7.0
Bi-214	0.47 - 2.7
Ac-228	1.3 - 140
Pb-212	1.1 - 150
Tl-208	1.1 - 120
K-40	8.8 - 120

TABLE 3: STATES WITH RARE METAL AND RARE EARTH PROCESSING ACTIVITIES

Producers of zirconium oxide, zirconium and hafnium sponge metal, ingot, and alloy	NJ, MA, AL, MI, OR, WV, NY, OH, PA CA, NH
Refractory firms using zircon in products	KY, NY, PA, MO, OH, WV, MI
Producers of zirconium compounds and chemicals	NJ, NY, MA
Producers of zirconium oxide for other than metal production	NY, AL, OH, SC, NJ, WV
Milled and sold ground zircon	NJ, NY, OH, SC, DE, CA, PA
Producers of rare earth compounds and chemicals	NY, PA, CA, CO
Producers of high purity rare earth metals	NJ, PA
Processed rare earth concentrates	IL, TN, NJ
Processed Canadian uranium mill solutions for rare earths	MI

FIGURE 1: POTENTIAL BEACH SAND COMPOSITION



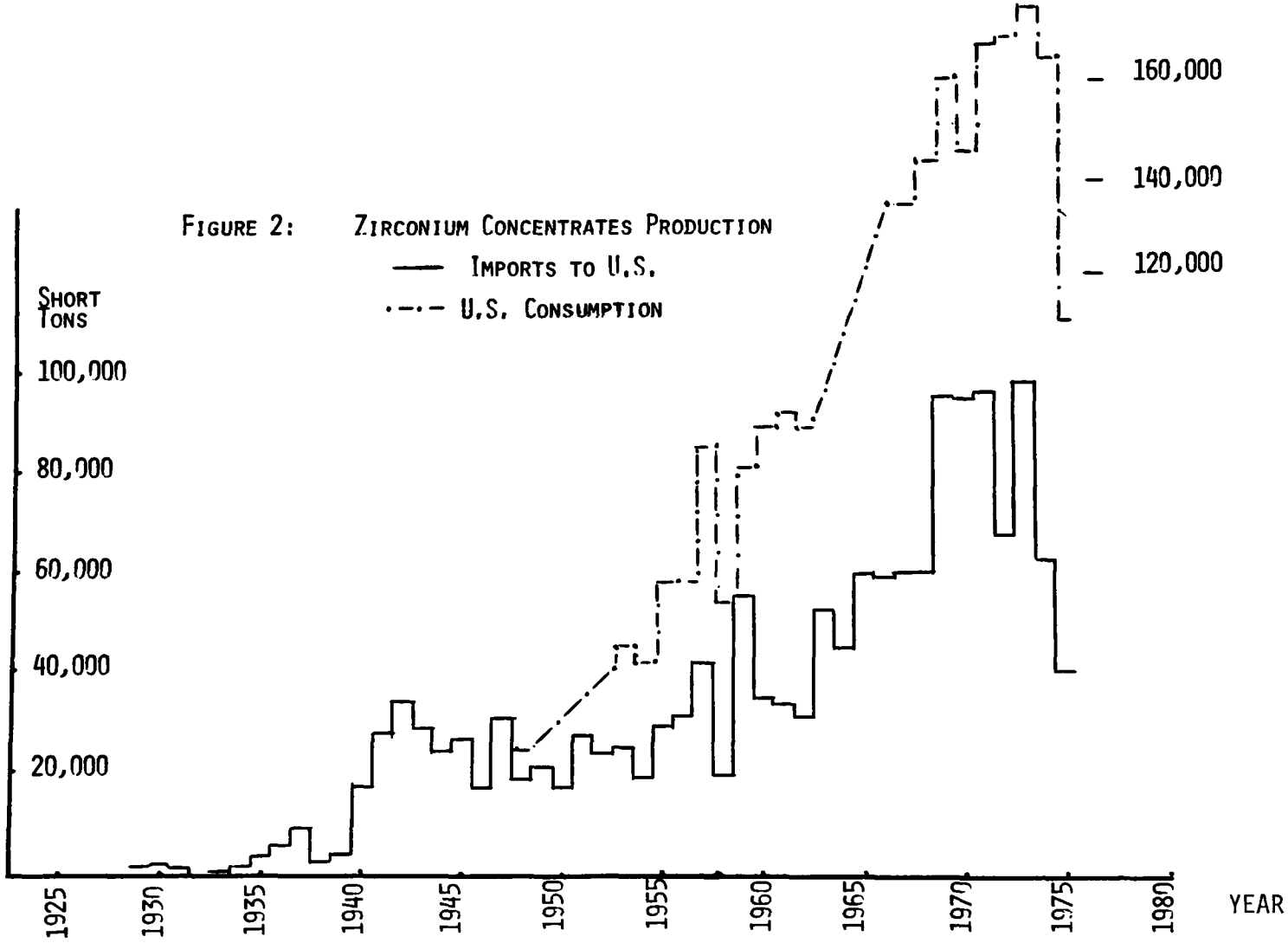


FIGURE 2: ZIRCONIUM CONCENTRATES PRODUCTION
IMPORTS TO U.S.
U.S. CONSUMPTION
SHORT TONS
100,000
80,000
60,000
40,000
20,000
160,000
140,000
120,000
1925
1930
1935
1940
1945
1950
1955
1960
1965
1970
1975
1980
YEAR

SESSION B

WASTE HANDLING AND TRANSPORTATION

Session Chairperson

W. C. McArthur

Hittman Nuclear & Development Corp.

DOE'S LOW-LEVEL WASTE MANAGEMENT PROGRAM

Sheldon Meyers
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Introduction

I wish to thank the Health Physics Society and its members for this opportunity to discuss with you the DOE's less publicized low-level waste program. I realize that the definition of radioactive waste by category or type has proven troublesome. But since I have been preceded by several speakers who have addressed low-level waste from differing perspectives, I will settle for a simple definition of the term. In describing our low-level waste program, I will be talking about solid radioactive waste which is currently being disposed of by shallow land burial at carefully selected locations.

In May 1978, Secretary Schlesinger approved a reorganization within the DOE which established an Office of Nuclear Waste Management which I now head--reporting directly to the Assistant Secretary for Energy Technology. Within that office there are three programmatic divisions. The Division of Spent Fuel and Transportation deals with spent fuel storage and all waste-related transportation requirements. The Division of Waste Isolation is responsible for all efforts related to deep geologic repository disposal of defense and commercial waste. The Division of Waste Products is responsible for technology development for both commercial and defense wastes, and also handling and treatment requirements leading to disposal of defense wastes. This includes disposal of low-level wastes at DOE sites.

The management of low-level waste is a part of our defense waste program, since the only low-level waste that DOE is directly responsible for is that generated at DOE sites. We do not have responsibility for the commercial radioactive waste burial grounds licensed by the Nuclear Regulatory Commission (NRC). However, DOE R&D activities related to disposal of its own low-level waste are also applicable to disposal at the licensed sites.

Before describing our low-level waste program, I would like to say a few words about the past low-level waste situation. Past reports and Congressional hearings have indicated that performance of low-level waste disposal operations "is not uniformly good" and that radionuclide migration in ground water has occurred at some sites. While releases into the environment have thus far not been a public health hazard, they have raised questions concerning the adequacy of the concept of shallow land disposal of low-level radioactive waste. In view of these questions, and because of the predicted future need for this disposal concept, DOE has underway a program to develop an improved technology for shallow land burial, including improved environmental monitoring needs at DOE disposal sites. Also, since some wastes now categorized as low level may not be acceptable for shallow land burial in the future, DOE plans an expanded program for investigating alternative disposal methods.

As a further preface, I wish to call attention to a recent Government initiative which will impact our low-level waste program. With an earlier DOE

task force report as a starting point, the President on March 13, 1978, established an Interagency Review Group (IRG) on nuclear waste management with representatives from 14 Government entities. The IRG was given the mandate to undertake a comprehensive review of nuclear waste management in its broadest sense, and to make policy recommendations to the President, as another step towards the establishment of a National waste management policy.

Key IRG Low-Level Waste Findings

To review briefly, the IRG had as its objective the formulation of recommendations for dealing comprehensively with the Nation's nuclear waste. They have summarized the state-of-knowledge for managing the various waste types including low-level waste. The results of their deliberations on low-level waste can be summarized as follows.

As to technical findings, they consider that:

- Technologies exist for management and disposal of low-level waste. Existing practice must be improved considerably. Siting must be improved and research and development accelerated, including R&D on alternative disposal methods.

In terms of policy, they recommend that:

- DOE assume responsibility for developing a National plan for low-level waste, with input and involvement of other Federal agencies, states, industry, and the public. The plan is to include establishment of an adequate number of regionally located disposal sites.

- States be provided the option to retain management control of existing commercial low-level land burial sites or to transfer them to the Federal Government.

If public laws are modified to extend Federal management over commercially operated disposal facilities, a program will be developed to implement such legislation.

Following its review of public comment on the draft report, the IRG reconvened to prepare a Decision Paper on Nuclear Waste Management, in fulfillment of its directive from the President. This paper is scheduled to be prepared for the President early this month (February).

We have begun implementation of some of these recommendations but, as you can appreciate, overall implementation of the IRG's recommendations awaits Presidential review.

The DOE Low-Level Waste Program

I plan to discuss today our current approach to the development of technology for an overall low-level waste program rather than operational detail--other relatively recent symposia presentations and various reports have provided considerable detail on operation of DOE's burial sites and quantities of low-level waste being disposed of by shallow land burial.

One major recent innovation affects responsibilities for management of our program and manpower resources devoted to structuring and implementing the program. In a decentralization move, assignments of broad responsibility for technology development in specific waste areas have been made to DOE Field Offices. Our Idaho Operations Office has this lead management responsibility for low-level waste, with their on-site contractor, EG&G, in a support role. Idaho has selected as an associate lead center the Oak Ridge Operations Office, supported by the Oak Ridge National Laboratory. Under this arrangement, Idaho has the responsibility for developing a detailed low-level waste management program and for coordinating implementation of the program among all DOE sites.

This decentralization effort has been underway for only a few months. Formal planning documents generated in the next few months will be given broad distribution for review and commend as one mechanism for greater involvement of interested parties outside DOE.

Perhaps the best way to discuss program content is to discuss briefly each of 10 major program elements. These elements are the top level of a detailed work breakdown structure to be followed in the development of a National low-level waste program.

1. Program Management: preparation of detailed program and technical management plans; establishment and coordination of Review Committees to include non-DOE personnel; establishment of system-wide quality control and quality assurance procedures; coordinate program interaction among all participants and with other DOE waste management programs.

2. Systems Analysis: program data acquisition, development of a consolidated data base and application of uniform methodologies; information and program analysis; information exchange and dissemination between participants and with other interested agencies and groups.

3. Criteria and Standards Development: DOE interface for participation with others in formulation of generic waste disposal/management criteria and standards; coordinate development of DOE criteria and standards; reconcile differences between DOE and generic criteria and standards.

4. Public and Regulatory Interface: formulate and implement programs which will include federal, state and local agencies, industrial and professional groups, and the public in the DOE low-level waste management program; establish appropriate interfaces with and between regulatory agencies, and coordinate environmental studies/activities required by NEPA regulations.

5. Technology Development: coordination, development, refinement, and/or implementation of environmental monitoring methods and programs, waste handling and treatment techniques, and shallow land burial technology.

6. Waste Generation Reduction: characterize wastes at sources of generation; forecast types and amounts of wastes generated; assess

means of reducing waste generation at existing facilities; establish waste reduction goals; coordinate improvements in waste reduction methodologies; coordinate implementation of waste segregation and volume-reduction policies.

7. Current System Operations: maintain cognizance of ongoing routine operation--direct management and implementation of site-specific operations remains the responsibility of individual DOE field offices.

8. Future System Development and Acquisition: locate and develop new waste processing and disposal facilities and sites as required to provide a nationwide system for low-level waste management.

9. Commercial: establish interfaces with commercial waste generators and commerical waste burial sites as required to exercise any DOE responsibility for handling and disposing of commercial low-level wastes.

10. Alternatives to Shallow Land Burial: provide new emphasis on study/research efforts on likely alternatives to shallow land burial; for example, intermediate depth disposal, deep disposal in either existing or new structures, and disposal in mined caverns.

A major document for public review now in preparation will delineate a National Strategy for low-level waste. It will contain considerably more detail on planned efforts under the program elements discussed above. Since milestones for program accomplishments will be contained in this document, it will require reconciling differences in Federal agency timing for the major milestones reflecting current agency commitments. These milestones are primarily aimed at establishing criteria and standards required for the Federal agencies to fulfill their responsibilities. Any duplication of efforts between Federal agencies will also become more apparent, and resources can be redirected for better efficiency.

Another ongoing effort is development by DOE of a contingency plan for acceptance of commercially generated low-level wastes in the event licensed disposal capacity becomes inadequate. This plan was requested by the NRC. It is still in the formative stages, but will address possible changes in commercial disposal capacity and a variety of options for DOE acceptance of the wastes. I do want to stress that, at this point, DOE considers this only as a planning exercise which it is prudent to have available. Hopefully, circumstances requiring its implementation will not occur.

Conclusion

In summary, low-level solid wastes are currently disposed of by shallow land burial. Continued reliance on this disposal method depends on development of an improved technology base, generation of acceptable and comprehensive criteria, and development of stablilization techniques for sites no longer needed to ensure that disposal areas will remain safe over the long term with minimal reliance on continuing maintenance and surveillance. We envision large potential increases in low-level waste quantities, particularly rubble and soil from decommissioning actions and other high volume wastes containing very low concentrations of radioactivity. This, plus the possibility that

certain "low-level" wastes may not be acceptable for shallow land burial, have led to an expanded DOE program to develop and assess alternative disposal technologies.

Successful development and implementation of the planned DOE program will to a large degree depend on assistance from organizations you represent here at this meeting--the fact that a major meeting would focus exclusively on low-level waste management highlights the importance of this subject. One of our major aims is greatly increased direct involvement of interested individuals such as yourselves. Several of the DOE and DOE contractor personnel directly available in our low-level waste program will be available throughout this week's meeting. I hope you will find time to discuss the DOE program with them.

I would like to conclude by stating that we are making every reasonable effort to assure that DOE generated low-level waste is managed in a safe and environmentally acceptable manner. Any additional low-level waste assigned to our custody will be treated in the same manner.

A NUCLEAR POWER PLANT APPROACH TO TEMPORARY ON-SITE STORAGE OF RADIOACTIVE WASTE

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Abstract

Designed storage space for solidified radioactive waste and spent filters at nuclear power plants has long been an area of concern. Increased personnel exposures arise when waste cannot be shipped for offsite disposal on a timely basis. In addition, significant cost savings can be realized in both transportation and burial costs if the short-lived isotopes in the solidified waste are permitted to decay. In order to increase flexibility in plant operations a large storage area for radioactive waste was designed. The design utilized existing structures as much as possible. A large concrete roof over a heat exchanger vault was connected by a dock to the radwaste processing area. The maximum anticipated radiation levels from the solidified waste were utilized in designing the shield wall around the storage area. Salient features of the storage area include four spent filter storage wells, a dual track overhead crane for moving 50 ft^3 liners and an enclosed truck pad to facilitate loading. The 6000 ft^2 storage area can conveniently handle 99 50 ft^3 liners on 5 ft x 5 ft pallets or 396 55 gal drums on similar 5 ft x 5 ft pallets. This is approximately one half of one unit's (PWR) average annual output of solidified evaporator bottoms. The spent filter storage wells will permit approximately two years decay with an estimated annual savings in transportation and burial charges of $12,000. Provisions for maintaining radiation exposures ALARA were included in the design of the facility. It was felt that conservatively estimated savings in radiation exposures and shipping and burial costs justified the project.

Introduction

Storage of solidified radwaste is frequently a problem at operating nuclear power plants. Since Three Mile Island Nuclear Station (TMINS) Unit 1 went commercial in 1974, several radwaste handling and storage problems have been identified. In 1976 plans were initiated on a Radwaste Storage area to alleviate several of these problems. Any proposed modifications were to satisfy the following criteria:

1. Storage space for normal volumes of solidified or compacted waste between processing and shipping.

2. Storage space for spent filters to allow decay for reduced shipping and burial cost.

3. Mechanized movement of processed radwaste to reduce personnel exposures.

4. Storage of processed waste in an area where personnel access is not required to reduce personnel exposures.

5. Increased speed and efficiency in loading of radwaste shipments to reduce personnel exposures.

6. Storage space for new empty radwaste containers.

Existing Conditions

The volume of waste processed and shipped to burial sites has varied over the years. In 1975, the first complete year of commercial operation, the volume of waste solidified for shipment was 16,180 ft^3. In 1976, the volume was decreased to 12,260 ft^3 and in 1977 to 8,000 ft^3 due to an incident with a leaky liner which curtailed shipments for several months. The 1978 volume was 13,100 ft^3. When Unit 2 reaches full commercial operation, (currently expected in January 1979), it is anticipated that the total annual volume will be 30,000 ft^3/yr.

The radwaste is currently stored in two locations: in a room for which volume reduction of radwaste is planned and below the Spent Fuel Storage Area, one level below the radwaste processing facility. Neither of these locations has proven to be satisfactory. The proposed volume reduction room is located between Units 1 and 2 and is in a main traffic pattern between the units. This area, therefore, is unsatisfactory for a storage area due to radiation exposure considerations. Storage area on another elevation is not acceptable in that it requires a large expenditure of manhours and manrems due to the excessive handling of the drums. Furthermore, these areas do not provide the necessary space for flexibility in storage to allow adequate time for decay and variations in shipping schedules.

Over the past years, further difficulties have arisen regarding the shipping of solidified radwaste. TMINS Unit 1 has utilized the Maxey Flats burial site in Morehead, Kentucky, which is now closed. More recently, the Sheffield, Illinois, facility, also in the Midwest, has closed. TMINS is now forced to ship waste to Barnwell, South Carolina. This facility has recently imposed a maximum volume per month limit. In addition, burial charges are proportional to radiation levels. Furthermore, with Unit 2 going commercial in 1979, the radwaste problems will be compounded. These problems and others have necessitated that Met-Ed provide additional radwaste storage area for both Unit 1 and Unit 2.

Design Features

Initial Criteria

Having identified the existing conditions several general design features or guidelines could be identified. The guidelines are as follows:

1. Locate storage area as close to and on the same elevation as the existing processing facility.

2. Provide storage area for spent filters.

3. Provide sufficient storage areas to be able to store waste six months at current or anticipated production rates for the two units.

4. Provide truck loading capabilities.

5. Provide adequate storage for empty 50 ft^3 liners.

6. Satisfy radiation dose limits surrounding the storage area.

7. Maintain radiation doses to plant personnel ALARA.

Using the above criteria and utilizing existing facilities as much as possible, a storage area was designed. An area outside the Auxiliary Building, a heat exchanger vault roof, was identified as "satisfying" many of the criteria listed above. This large flat area could be relatively easily connected to an existing dock. After considering several other alternatives (see Evaluation of Alternatives below), it was decided that this area over the heat exchangers could best be modified to meet the radwaste requirements.

The design was divided into two phases. Phase 1 involved modifications deemed necessary for better utilization of the existing facilities. Phase 2 was directly associated with the new radwaste storage facility. Phase 2 consisted of the following:

1. A covered dock from existing auxiliary building door to the heat exchange vault roof.

2. Construction of recessed spent filter storage wells in this dock.

3. An overhead crane for filter cask and shield handling.

4. A hydraulic dock board for matching of truck height with dock during loading operations.

5. A covered truck pad for transport vehicles.

6. Enclosure of dock for all-weather waste handling.

7. Shield wall between the storage area and the access road.

Radiological Criteria

(1) Spent Filter Wells. TMINS Unit 1 generates an annual average of 60 small and 10 large spent filters with a total of about 50 curies. The isotopic breakdown of the activity on the filters was assumed to be the same isotopic distribution as for the effluent discharges (see Table 1 below).

Table 1
Nuclide Composition of Effluent
Releases for 1976 and 1977

1976

Isotopes	% of Total	Initial Curies	T 1/2	Curies After 2 Year Decay
Mn-54	5.0	2.5	312.0	0.49
Co-58	49.0	24.5	71.3	0.02
Co-60	4.5	2.25	5.3y	1.73
I-131	3.0	1.5	8.d	0.0
Cs-134	10.5	5.25	2.06y	2.68
Cs-137	22.0	11.0	30.1y	10.5
All Others	<1.0	<0.5	-	0.0
Totals	100%	50.0	N/A	15.5

1977

Isotopes	% of Total	Initial Curies	T 1/2	Curies After 2 Year Decay
K-40	0.217	0.11	1.3E+9y	0.1
Mn-54	1.08	0.54	312.0d	0.10
Co-58	23.8	11.9	71.3d	.0098
Co-60	2.14	1.07	5.3y	0.82
Cs-134	30.9	15.45	2.06y	7.88
Cs-137	40.0	20.0	30.1y	19.1
I-135	1.86	0.93	6.6hr	0.0
All Others	<1.0	<0.5	-	
Totals	100%	50.0	-	28.0

It is currently planned to set regular 50 ft^3 liners into the spent filter storage wells so that spent filters can be dropped dry from the small spent filter transport cask into the liner. Each 50 ft^3 liner will accommodate a year's supply of spent filters. At the end of the first year, the second well will be used, etc. After two year's decay, the first year's accumulation will be shipped for disposal. The two year decay time (700 days) (10 half-lives for the predominant isotope Cobalt 58) will reduce the estimated initial activity of 50 curies by an estimated 44 to 70%. The longer half-lived cesiums then become the predominant isotopes; however, their lower energy radiations are easier to shield. The shielding for the storage wells was sized assuming each liner had 50 curies of activity on spent filters with the above-mentioned isotopic distribution. The plugs for the wells were designed so that radiation levels on the dock will be less than 5 mr/hr.

(2) Shield Wall Considerations. The shield wall separating the new radwaste storage area from a vehicle access road was sized utilizing actual radiation levels from 50 ft^3 liners of waste. Waste liners with contact readings of 1 R/hr and 5 R/hr were analyzed at specific locations in the proposed storage area. The activity in the drums was assumed to be Co-60. The shielding evaluation indicated that an 18 in. concrete shield wall would be required to satisfy the radiation zone criteria outside the storage area.

(3) Other. Numerous other areas of the design were influenced by radiological conditions. All surfaces which potentially could be contaminated are specified to be painted with an epoxy paint which will facilitate decontamination. Radiation monitors were placed in the storage area with a remote readout to inform operations personnel of the radiological conditions. Loading waste has been simplified by providing a dock board to allow driving a fork lift directly onto the transport trailer. The dual track crane over the spent filter storage wells can also be utilized for truck loading. The crane has a rated 15T capacity and, therefore, can handle a 50 ft^3 liner inside of a 3-inch lead shield. Since the storage area did not lend itself to any partitioning due to aircraft impact criteria imposed on the concrete walls of the existing building, radwaste containers will be stored in the area segregating the waste in accordance with the radiation levels. Higher level waste will be placed in the far end of the facility to minimize the exposures to personnel who enter the area with additional waste or who are in the process of loading waste for shipment to a burial site.

Evaluation of Alternatives

Conveyors

Consideration was given to a conveyor system for storage of the waste on the heat exchanger vault roof. The idea was to have seven parallel tracks. Waste would be placed on the conveyor at one end and shipped from the other. Due to the high cost of this system, potential maintenance problems, and reduced storage area this idea was dropped.

Open (Uncovered) Storage Site

Stored waste exposed to the elements can potentially contaminate run-off water. The roof over the proposed radwaste storage area was justified by assuming that all run-off would have to be monitored and possibly processed through the radwaste tystem. Calculations indicated that processing of normal annual rainfall collected on the heat exchanger vault roof as contaminated liquid waste would require about 400 hours per year of evaporator operation. The normal per gallon cost for processing contaminated liquid is $.25/gal. Solidification and disposal costs are approximately $3.60/gal. The estimated 200,000 gallons of rainwater/year more than justify the cost of the roof at $130,000.

Remote Storage (Away from Plant Proper)

In addition to the increased expense, the major objection to this option was that a storage location away from the processing area would not be as convenient as one next to the processing area. Furthermore, a remote storage area would necessitate longer periods of handling by the operators in order to get to the storage area and, therefore, would likely increase radiation doses.

Overhead Remote Operation Crane System

An overhead crane indexing system would have been the ultimate in a radwaste handling facility. However, the cost of such a system made its serious consideration short-lived. In addition to the higher cost, some difficulty would have been experienced in supporting such a system in light of the rigid aircraft impact criteria imposed on outside walls housing safety-related equipment.

Costs

Manrems

A total of 30.214 manrems were expended in 1977 in Radwaste Processing Operations and Maintenance. Twenty-two of these were absorbed by Maintenance Personnel, and the remainder by Operations, Supervisory, and Radiation Safety. Utility workers handling radwaste are included in the maintenance personnel exposure. 1978 saw a(n) crease to manrems. Annual manrems savings by mechanized handling are estimated at 50%.

Shipping and Burial Costs

Savings in shipping and burial costs assuming a decay time of 210 days (3 half-lives of CO-58 a predominant isotope) and assuming 1978 shipping and burial costs:

The cost difference between shipping a shielded truck of 50 ft^3 liners and an unshielded truck is $1,000/shipment. Met-Ed currently ships approximately 15 such shielded shipments/year and Unit 2 is expected to double this. The dividing line between shielded and unshielded shipments is approximately 200 mR/hr.

Provision of 210 day decay time could reduce radiation levels from 1 R/hr (requiring a shielded truck) to 0.125 R/hr, with savings of $1,000/shipment.

The Barnwell South Carolina burial ground currently imposes a $1.40 per cubic foot radiation surcharge for each container which exceeds 0.200 R/hr over those at less than 0.200 R/hr. A three half-life storage for decay could reduce 90% of all TMI radwaste to <0.200 R/hr. Annual radwaste volumes are currently estimated at 15,000 ft^3/year/unit and, therefore, savings of $21,000 per year/unit or $1.2 million over the plant life could be realized, again assuming 1978 shipping and burial costs.

These figures do not include the liner surcharge which is currently $1.50/ft^3$.

Having to ship filters without the benefit of decay also imposes a financial penalty. The radiation surcharge currently imposed at the Barnwell, South Carolina burial ground per ft^3 is $13.18 in the 20-40 R/hr range, and $4.67 in the 1-5 R/hr range. This difference of $8.50/ft^3$ for 100 ft^3/yr is a direct savings of $850. One trip per year for filter disposal will cost approximately $2,500 with decay. Without decay, three trips per year will be required due to radiation levels for direct savings of $5,000/yr plus an estimated $850/yr radiation surcharge savings. Manrem savings estimates due to remote handling and handling of decayed rather than fresh filters are not available at this time, but are assured.

Manhours

TMI currently expends approximately 6,000 manhours per year in radwaste handling operations. Mechanization by use of the forklift and use of the new storage area, versus the present temporary storage area on the lower level, could reduce this manpower requirement by 20%. Currently 50 ft^3 liners are transported from the operation level to the next lower level by three persons in the elevator for temporary storage. These liners must then be returned from the lower level by reversed process for loading on the shipping vehicle. Labor savings for forklift movement are estimated at approximately $15,000/year based on $12/manhour or $600,000 over the life of the plant. Addition of the Unit 2 waste volumes would make lower level storage of radwaste an intolerable situation.

Estimated Cost of the Radwaste Storage Facility

	Cost Estimate
Engineering	Sub Total $122,000
Construction	
Phase I	41,000
Phase II	
Covered Dock	242,000
Filter Storage Wells	20,000
Monorail	53,000
Dock Board	9,000
Dock and Storage Area Roofing	130,000
Shield Wall	27,000
	Sub Total $522,000
	Grand Total $644,000

Summary

This utility feels that the savings in manrems, shipping and burial costs, and manhours justify the expenditure of this sum to convert a currently unused space to a radwaste storage area.

RADIOACTIVE SLUDGE TREATMENT BY THE CENTRIFUGAL DEHYDRATION METHOD

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ABSTRACT

The radioactive sludge produced after chemical treatment of radioactive liquid waste usually contains 0.3-3% of solid by weight. The gross β activity of sludge produced at the Institute of Nuclear Energy Research (INER) varies 10^{-1}-10^{-3} μCi/g, and the use of centrifugal method for dehydration is justified. The effluent from the decanter goes through the precipitation and concentration stage and then the supernatant is pumped so that a stable concentration of liquid waste can be maintained during separation stage. The decanter used at INER for treating low level radioactive waste is a Westfalia SDB230 decanter which is of solid-bowl type. The liquid waste from a 40MW(th) Taiwan Research Reactor (TRR) and its associated hot laboratories is described in this report. The treatment capacity is about $2m^3/h$ and the characteristics of the separator and the associated systems are discussed. As a result of improvement the solid in concentrated sludge is about 20% by weight, and the production rate is about 240-250 kg/h. Furthermore, no water is added to the concentrated sludge. The solidification can be done in a closed type sludge-cement premixing system, and the homogeneous concrete can be obtained.

INTRODUCTION

The radioactive liquid waste produces radioactive radioactive sludge after chemical treatment. The radioactive sludge so treated contains solid generally at a few percents. The radioactive sludge produced at INER has 3-6 wt% of solid content depending on the period of gravity thickening. It is not econo-

mical to treat such great amount of sludge by storage or solidification. Therefore, it is necessary that dehydration be firstly made to minimize the volume of the radioactive sludge prior to solidification. The current treatments of dehydration can be classified as:

(1) Gravity,
(2) Air drying,
(3) Gravity filtration,
(4) Vacuum filtration,
(5) Pressure filtration,
(6) Centrifugal dehydration.

The merits and shortcomings of treatment mentioned above are presented in Table 1 where the volume reduction ratio is defined as:

$$\text{Radio of volume reduction} = \frac{\text{Volume of feeding sludge}}{\text{Volume of concentrated sludge}}$$

Table 1. The Comparisons of Different Dehydration Treatments for Radioactive Sludge

No.	Treatment	Merits	Shortcomings
1	Gravity thickening	simple operation, low cost	low volume reduction ratio
2	Air drying	simple operation, low cost	easy to become contaminated, vast space required
3	Gravity filtration	simple operation, low cost	laborious to remove concentrated sludge
4	Vacuum filtration	more adoptive to various sludges	filter frequently replaced, complicated and expensive facilities
5	Pressure filtration	high filtration speed	operation complicated, easy to become contaminated
6	Centrifugal dehydration	low contamination, high volume reduction ratio	not applicable to particulate sludge

The sludge produced at INER has approximately 10^{-1}- 10^{-2} Ci/g of gross β activity suitable to centrifugal dehydration. The effluent from such dehydration has less solid content. As long as it meets the application of gravity thickening, the sludge is setting and the clarified liquid can be pumped out.

In this way the solid content of the sludge may be applied to batch cycle treatment in a steady condition.

The separator used at INER is called the solid bowl type decanter. The inner diameter of the separators bowl is 22.5 cm while that of the surface of revolution liquid is 13.5 cm. The revolution speed of the bowl is 2850 rpm and its G force is approximately 820 (Gr76). Ten years ago, the G force of the separator was about 800-2000 and has increased to 3000-5000 in the recent years.

EXPERIMENTAL

The separator of the solid bowl type decanter is often used in a BWR type nuclear power plant. The type of decanter used at INER is a continuous centrifuge with a horizontal solid shell bowl. A conveyor screw is enclosed for the separation of solids from suspensions. By changing the regulating ring dam, altering the difference in speed of the bowl shell, and using the particular type of suspension, the maximum clarification and dryness can be obtained regardless of the specific gravity of solids.

The working principle of the bowl is as follows:
The bowl operates in two zones, i.e., separating zone and drying zone. In order to see that the separating function meets the requirements, it is necessary to make an appropriate selection between separating zone and drying zone. In other words, the following two principles can be followed to decide the proper inner diameter of revolution.

1. The solid output is required to be as dry as possible. The degree of purity of the clarified liquid is of secondary improtance. In this case, it is necessary to work with a long drying zone. This is done by selecting the largest inside diameter regulating ring dam of the bowl.
2. The effluent is required to be of maximum purity. The degree of dryness of the separated solid mater is of secondary importance. In contrast to principle 1, it is necessary to work with a long separating zone, and the smallest inside diameter regulating ring dam should be used.

In the majority of cases, however, it is necessary to compromise. The most satisfactory regulating ring dam and speed difference for the suspension to be processed are found by making several tests.

RESULTS AND DISCUSSION

The results of treatment under various operation condition will be described separately as below.

1. The feeding rate remains constant. By changing the regulating ring dam, the results are shown in Table 2 where n indicates the difference in speed between the bowl shell and the conveyor screw and ø is the diameter of regulating ring dam.

Table 2. The Efficiency of Concentration as a Function of Ring Diameter

Feeding rate (m^3/h)	n (rpm)	Regulating ring ø (mm)	Solid content of feeding (wt%)	Solid content of effluent (wt%)	Solid content of concentrated sludge (wt%)	Concentrated products (kg/h)
2	18	160	6.70	6.70	-	0
2	18	145	6.70	5.48	20.63	4.8
2	18	135	6.07	5.67	23.83	31.8
2	18	127.5	6.07	-	-	0

2. The diameter of the regulating ring dam is 135 mm and the feeding rate is $2m^3/h$. The results of treatment under various operation conditions are shown is Table 3.

Table 3. The Efficiency of Concentration as a Function of n

Feeding rate (m^3/h)	n (rpm)	Regulating ring ø (mm)	Solid content of feeding (wt%)	Solid content of effluent (wt%)	Solid content of concentrated sludge (wt%)	concentrated products (kg/h)
2	6	135	5.60	5.14	16.25	18.0
2	12	135	5.16	4.76	16.68	25.1
2	18	135	6.07	5.67	23.83	31.8
2	30	135	6.35	5.60	18.12	11.8

3. The diameter of the regulating ring is 135 mm and n is 18. The results of treatment under various feeding rates are shown in Table 4.

Table 4. The Efficiency of Concentration as a Function of Feeding Rate.

Feeding rate (m^3/h)	n (rpm)	Regulating ring ø (mm)	Solid content of feeding (wt%)	Solid content of effluent (wt%)	Solid content of concentrated sludge (wt%)	concentrated products (kg/h)
1	18	135	5.16	4.79	23.78	26.2
2	18	135	5.16	4.76	21.47	25.1
3	18	135	5.16	4.29	19.77	16.6
4	18	135	6.48	5.91	16.63	13.0
5	18	135	6.48	6.21	14.38	7.7

Based on the data given in Tables 2-4, the improvement of the dehydration system should be made.

1. The feeding and effluent receiving tank.
   It is known from the above operation tests that the concentration sludge in the dehydrated effluent remains about 90% of the feeding. As shown in Fig.1, the effluent cannot flow directly into the storage tank of radioactive liquid waste. As shown in Fig.2, after improvement the operation system consists of two tanks for feeding and effluent receiving and one receiving tank of sludge. The dehydrated effluent, as a result of the gravity thickening in the settling tank (K_O76), can remain the solid content of the sludge of feeding about 5-6 wt% and raise the operation efficiency of the decanter.

2. The use of flocculant agent(Ec70).
   In order to increase the products of the concentrated sludge, the flocculant agent was used to enlarge the particle size of the solid in the sludge so as to accelerate the setting speed of the solid. The dosage of the commercial flocculant agent is 40-60 g/m^3. When the flocculant agent is used, the production rates of the concentrated sludge are increased to 240-250 kg/h.

In conclusion as mentioned above, concentration of the radioactive sludge by centrifugal dehydration method features that the sludge can be treated in closed transportation system, and the concentrated sludge can further be applied to cement solidification by means of pre-mixing cement mixer. Upon solidification, the cement and the concentrated sludge can mix homogeneous and then fill in the 53 gallon drums. The operation is simple enough yet with less contamination and weather resistant.

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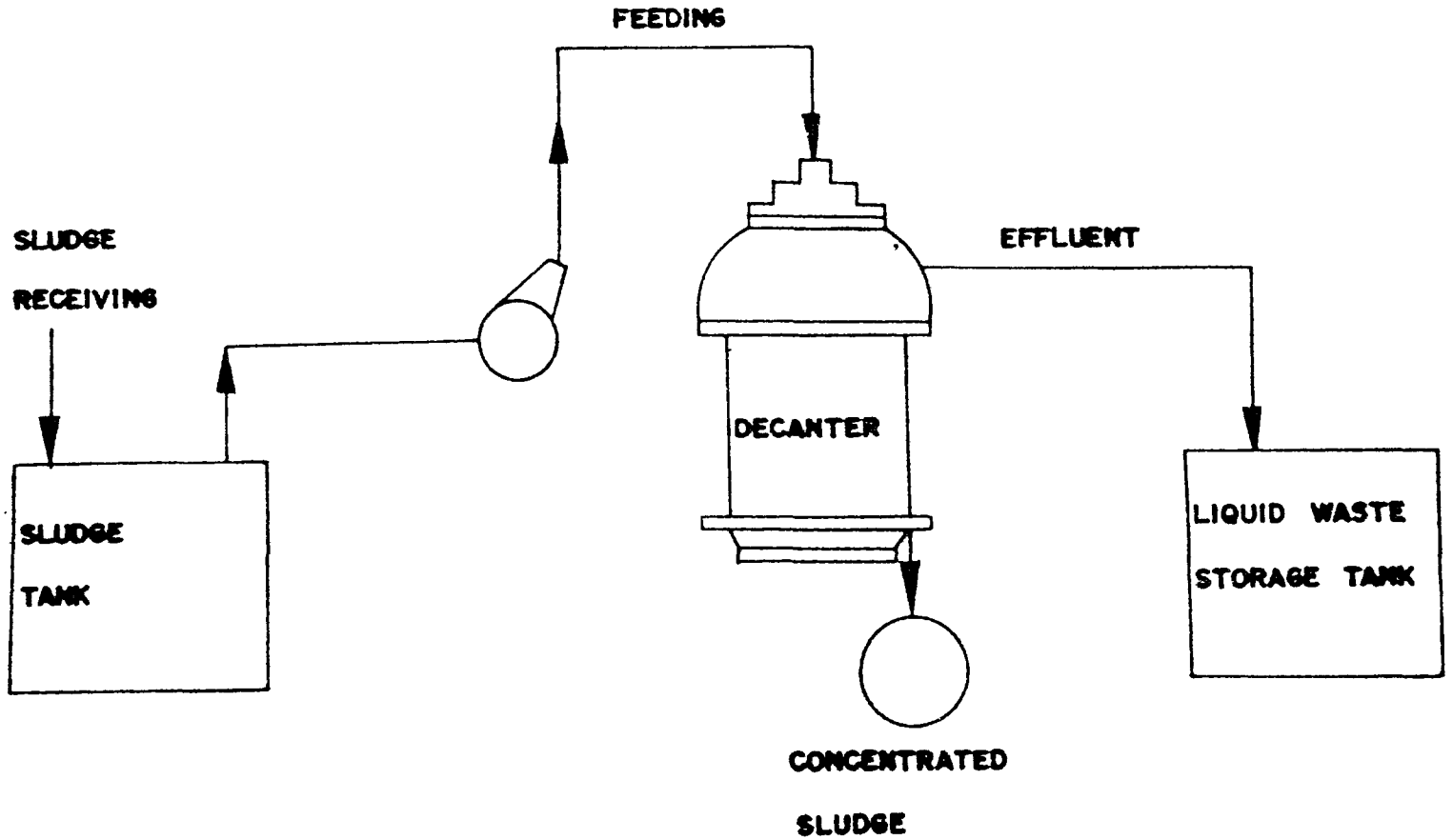

Fig.1 Before the Improvement of the Dehydration System.

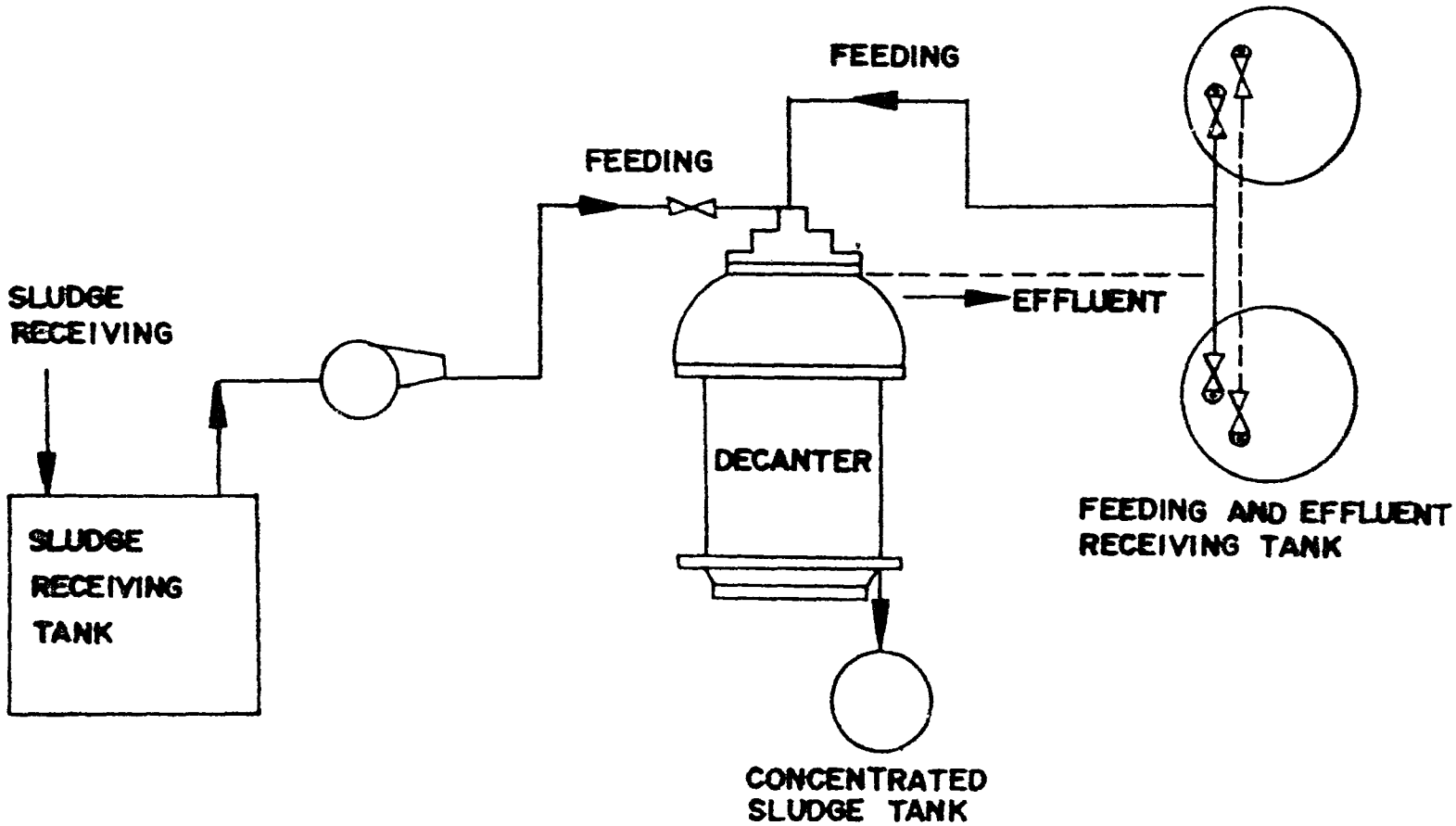

Fig.2 The Improvement Operation System

REDUCTION IN WASTE HANDLING AND TRANSPORTATION

by John E. Stewart
Werner & Pfleiderer Corporation

INTRODUCTION

The reduction in volume of low level wastes and associated handling and transportation is especially important to health physicists. In most cases the result is less exposure to operating personnel and less risk of exposure to the public during transportation and subsequent burial or other disposal.

Credit for initially exploring the benefits of volume reduction must go to the Europeans who had to face the radwaste problems earlier due to their limited burial and disposal facilities. [1] [2] They performed research on volume reduction and solidification processes using various binders in the early 1960's. This paper not only draws upon European experience, but also that of the U.S. and other countries for improving the radwaste process we know as the Volume Reduction and Solidification (VRS) system.

The radioactivity in liquid wastes is primarily in the solids they contain. It then follows that separating out the solids and solidifying them without the water is not only a logical technological advance in itself, but one with many attendant health physics benefits.

There are several low level waste solidification systems on the market, [3] plus two systems that reduce volume but must interface with a solidification system, and one that both reduces volume and solidifies at the same time. Other systems are under development. Each of the different systems has different health physics problems and it is impossible to discuss them all in one paper. Therefore, this paper will discuss only the Volume Reduction & Solidification (VRS) system of the Werner & Pfleiderer Corporation, which has been accepted for installation at several U.S. nuclear power plants and has a wealth of data accumulated as a result of over 50 system years of operating experience in Europe and elsewhere. The WPC - VRS system Topical Report has been evaluated and accepted by the NRC for referencing by utilities in their SAR's. [4]

To get a better idea of what volume reduction means to the U.S. nuclear power industry, refer to Figure 1. This graph appeared originally [5] with only the left-hand ordinate and we have converted MWt to MWe on the right-hand ordinate to enable you to quickly determine the waste quantities per 1000 MWe. From this graph, it is readily apparent that volume reduction is essential. Solidification, of course, is a regulatory requirement.

OPERATOR EXPOSURE REDUCTION

An integral part of every VRS system is a shielded control room for the operator. From this room the operator can observe the major processing operations through lead glass windows. Closed circuit TV systems monitor all other important phases of the process.

To illustrate this point, refer to Figure 2, which represents a typical approach to a VRS system design. Note that the process control panel and the crane control panel are both in a shielded area and that there are three lead glass windows. One window allows the operator to directly observe the filling chamber, a second permits observation of the drum capping and monitoring station, and the third looks directly into the filled drum storage area. Although not shown, several closed circuit TV cameras are located inside the processing area with the viewing screen in the control room. The net result is that the operator works in an area that is at or close to normal background radiation levels for a nuclear power plant.

All of the important phases of the process are interlocked to prevent operation under serious abnormal conditions and both audio and visual alarms are provided in the control room.

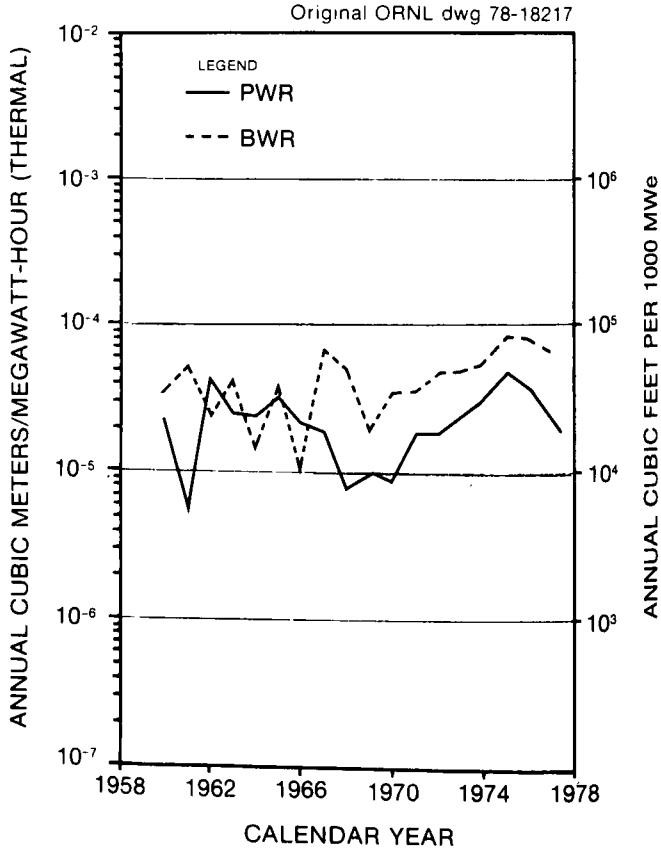

Figure 1. Annual solid waste volume shipped from U.S. nuclear power plants. Reference 5, ORNL/NUREG-43.

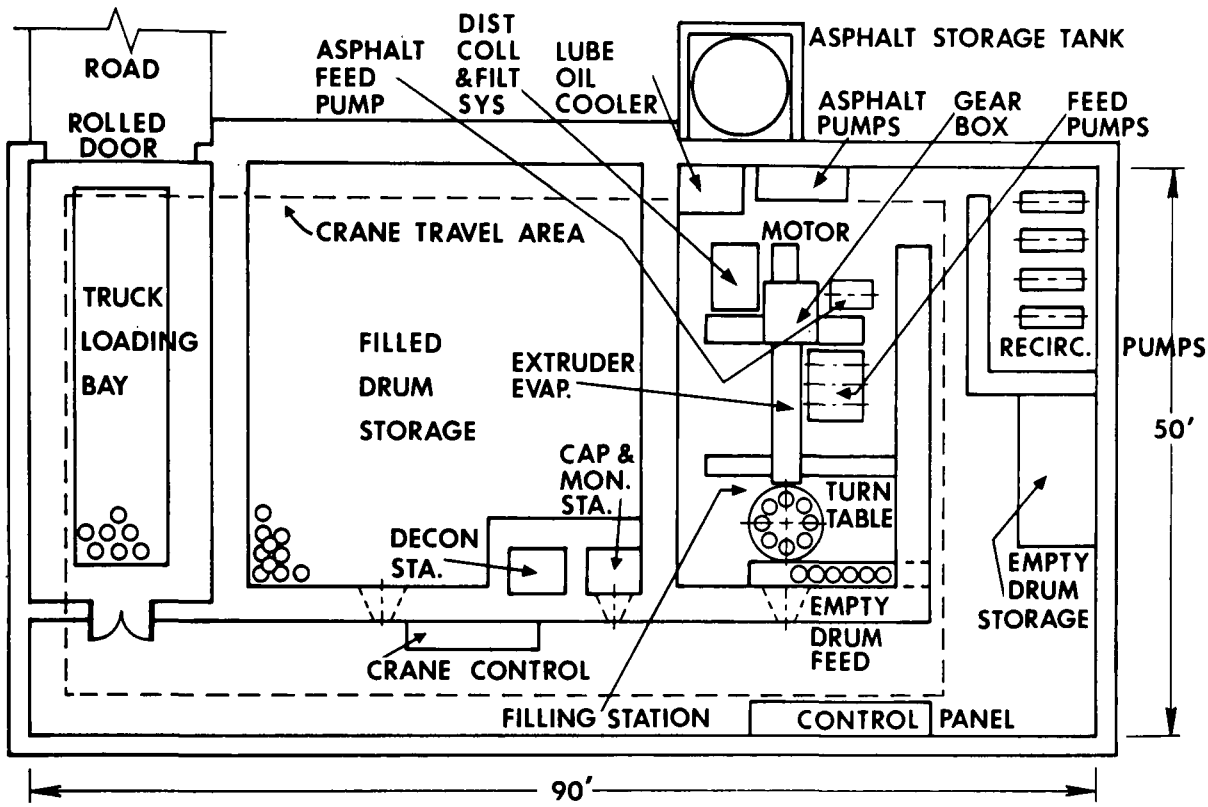

Figure 2. Typical design approach to VRS layout.

EQUIPMENT SEPARATION

VRS system design follows the philosophy of separating the hardware that contains radioactive material from other pieces of equipment. Note in Figure 2 that the asphalt feed tank is outside of the processing area and that the radwaste hardware is separated by shielding walls which effectively control the radiation levels in the various areas according to their function and anticipated occupancy times

The functional areas and their anticipated dose rate are given in Table I below. For location within the total system, refer to Figure 2.

TABLE I

FUNCTIONAL LOCATION	DOSE RATE R/HR	OCCUPANCY HRS/YR	EXPOSURE MAN-REMS/YR
CONTROL ROOM	0.00025	500-1000	0.125 to 0.25
FEED TANK & PUMP AREA	0.1	10	1
EXTRUDER-EVAPORATOR (PROCESSING AREA)	0.1*	10	1
DRIVE & AUXILIARY AREA	0.01	10	0.1
EMPTY DRUM STORAGE	0.00025	100	0.025
FILLED DRUM STORAGE	1.0 to 10.0	0	0
DRUM FILLING AREA	1.0 to 5.0**	10**	1.0
TRUCK LOADING BAY	0.1	10	1

*Less than 0.010 R/hr during maintenance periods because of self-cleaning action of the rotating screws in the extruder-evaporator.

**When maintenance is required, all filled containers will be removed and dose rate will be 0.01 R/hr.

OPERATING RELIABILITY

Note in Table I that the areas of highest dose rate have a minimun occupancy time per year. This is directly related to equipment reliability which determines the frequency and amount of time that personnel must be in a particular area. The VRS process has over 50 system years of operation. The two oldest installations are at Marcoule, France, and Karlsruhe, West Germany. Both of these are nuclear research centers and both keep records of the system operation. A third installation at Borssele, Holland (power plant) also kept track and their results are presented in Table II below.

TABLE II
OPERATIONAL RELIABILITY

BORSSELE & KARLSRUHE	7,500 HOURS WITHOUT MAINTENANCE
MARCOULE	13,000 HOURS BEFORE DISASSEMBLY FOR ROUTINE MAINTENANCE

It is anticipated that the normal operation of the VRS system in the U.S. will equal 2,000 hours per year. Thus, the reliability record established in Europe indicates that the VRS system would operate in the U.S. for years before maintenance personnel would be required to enter the areas of highest radiation. The exposures listed in Table I for the processing area and other areas where equipment containing radioactive materials are located is based on the maximum frequency of one annual inspection regardless of operating time.

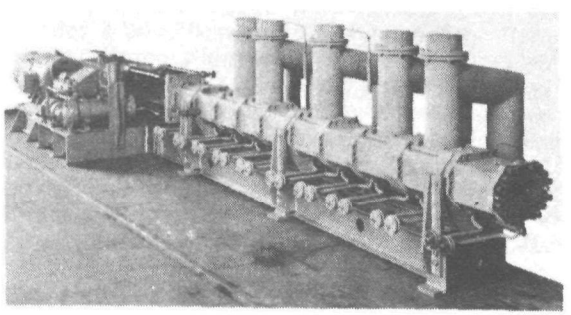

Figure 3. Model T-120 VRS extruder-evaporator as used in European installations.

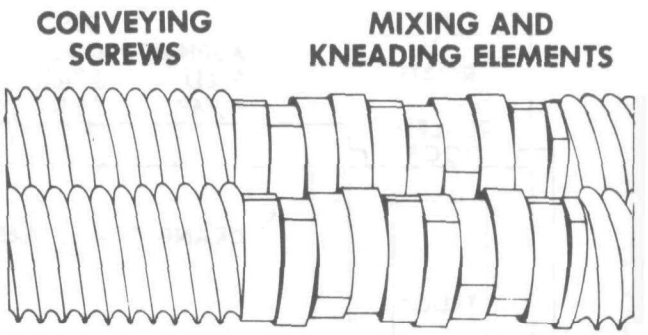

Figure 4. Co-rotating screws which operate with 1 mm of clearance to provide self-cleaning action inside extruder-evaporator.

COMPONENT DESIGN

The major piece of hardware is the extruder-evaporator shown in Figures 3 and 4. Basically the extruder-evaporator consists of two co-rotating screws installed inside of heavy metal sections called barrels. The barrels are constructed of thick nitrided steel and act as a shield while the wastes are being processed. In addition, at any given time during processing there is only about two liters of waste being mixed with two liters of asphalt binder. Thus, the low inventory in combination with the thick steel walls of the extruder effectively keep the radiation levels at the surface of the extruder-evaporator at low levels.

For example, in Figure 5, the external dose rates after processing medium level wastes of 15 and 90 Ci per cubic meter in the feed stream are shown for Karlsruhe[6], West Germany. While the wastes are 10 to 100 times higher than those expected at power plants, the relationship of shutdown dose rates to input activity is illustrated. It is, therefore, reasonable to expect service dose rates of less than 10 mR/hr when shut down for maintenance.

EFFECT OF LESS END PRODUCT

Volume reduction and solidification in the same single-step process provide the greatest health physics benefits. There is a reduction in the amount of handling on-site as well as in the area required for on-site storage prior to shipping off-site. The number of shipments to the burial site is reduced and along with that a significant reduction in the accident probability, which not only involves the health physicist but the general public as well.

Table III compares the shipments from a VRS system with those from a non-volume reducing cement system.

TABLE III
SHIPMENT COMPARISONS

WASTE TYPE	DRUMS/YR		DRUM	SHIPMENTS/YR		SHIPMENT
	VRS	CEMENT	REDUCTION	VRS	CEMENT	REDUCTION
BWR FILTER/DEMIN.	910	5,470	4,560	29	144	115
BWR DEEP BED RESINS	545	3,270	2,725	17	87	70
PWR WITH CPS *	175	1,225	1,050	7	34	27
PWR WITHOUT CPS *	415	2,900	2,485	14	78	64

*CPS — Condensate Polishing System

According to a recently published report (ORNL/NUREG-43, Oct. 1978, pg. 57),[5] "At this time, the main problem areas in radwaste solidification at the power plant appear to be drum capping, monitoring, and decontamination, which remains largely a manual operation. To cope with drumming problems, many plants have replaced or modified their original radwaste processing equipment in an effort to perform more of the operations remotely and automatically... The indication levels of some spent resins and filter cartridges and/or precoat filter sludges can be as high as 100 R/hr or more, and unless adequate shielding is provided during storage, transfer, and packaging, there is risk of high exposure for operating personnel." All of the above operations are done remotely in the VRS system, thus eliminating the risk of increased personnel exposures as reported by the nuclear plants to ORNL. It also follows that the fewer drums or other containers there are, the less is the handling that is required to move the containers from the filling area through capping, monitoring, decontamination, into on-site storage, and finally to the transport vehicle. The reduction in transport requirements and the resultant benefits have already been mentioned. At the burial site, the same benefits that apply at the plant site are in effect. In addition to the health physics benefits obtained due to VRS processing, there are other operational benefits dealing mostly with economics.

If there is a long time storage at the power plant or at the burial site, there is an additional benefit of the VRS system with regards to drum corrosion. The VRS system uses asphalt as a binder, which inhibits internal drum corrosion, thus reducing the risk of exposure from the source.

SYSTEM DESIGN PHILOSOPHY

The VRS design philosophy is to separate all components containing radioactive materials, to isolate the system operator from all of the potential sources of radiation, and to reduce to a minimum the occupancy time of any maintenance personnel when entry into a radiation area is required. Many of the design faults reported by ORNL and others as an operating deficiency in nuclear power plant radwaste systems are not present in the VRS system.

DISTILLATE COLLECTION SYSTEM. The distillate collection system of the VRS process warrants discussion. Water in the feed stream is heated to above the boiling point in the extruder-evaporator, causing it to be driven out through the steam domes mounted on the top of the extruder. This distillate is condensed, passed through filters and either returned to the plant system or stored until suitable for release to the environment. The

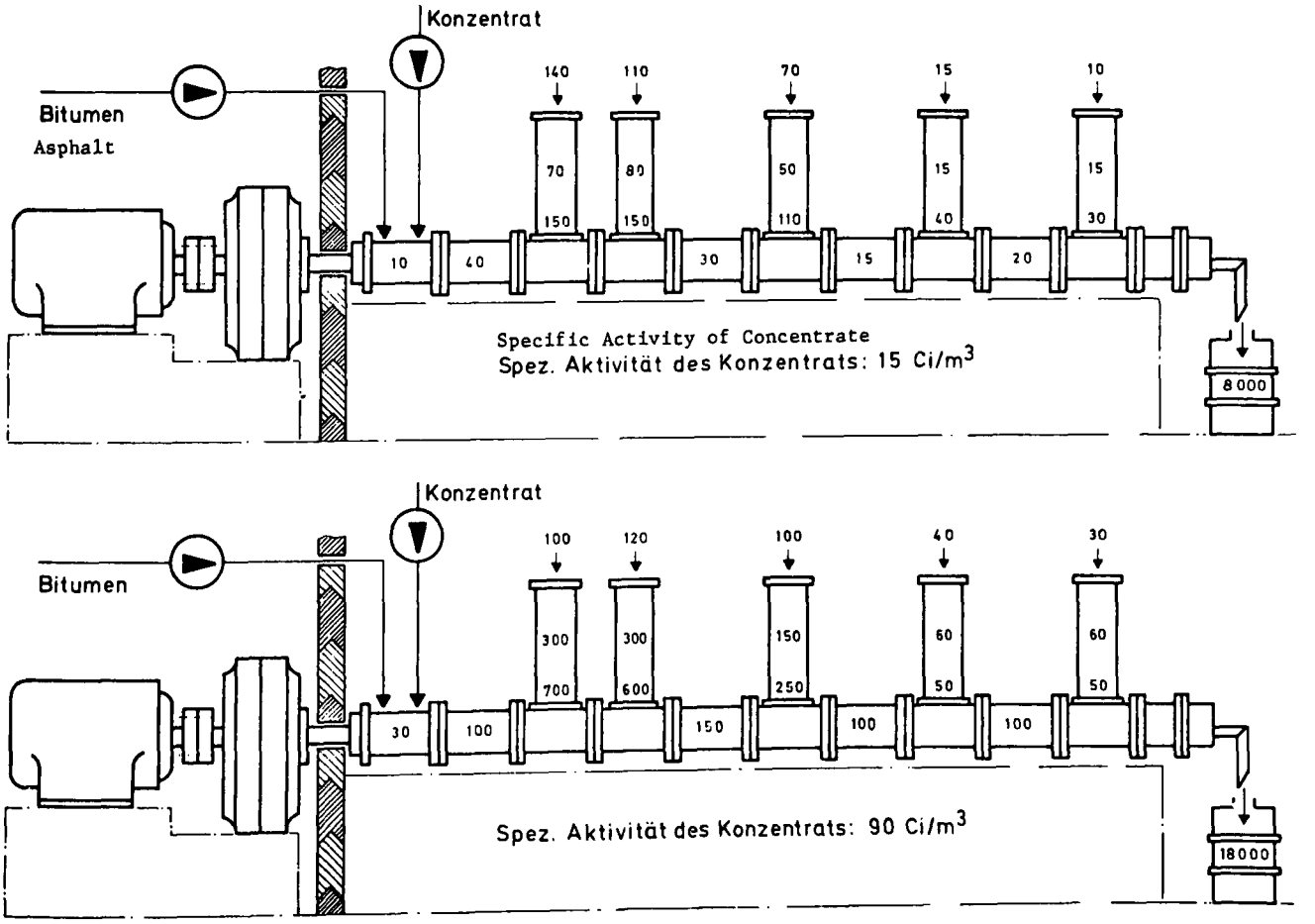

Figure 5. Operating dose rates (surface mR/hr) at Karlsruhe Nuclear Research Center where feed stream includes medium level wastes from fuel reprocessing center.

decontamination factor, measured from waste inlet to distillate, is 6,000. The radiation level of the distillate filters following filtration is only a few mR/hr. during normal changeout.

FILLING CHAMBER. The filling chamber is a shielded area that is virtually isolated from all other areas in the system. As the drums are filled with the mixture of radsalts and asphalt, some residual vapor may be present. Directly above the drum being filled is a hood having a negative draft. Thus, all vapors are drawn through the hood and pass through a prefilter before exhausting to the plant HVAC system.

MONITORING. Swipe samples related to monitoring and possible decontamination are done remotely by the operator. The container handling system is designed to prevent spills, but should one occur it can be cleaned easily in the VRS system. The asphalt binder is a slow flowing thermoplastic and it will harden quickly outside of the container. After it solidifies, it can be scraped up and put in any of the drums in the chamber. Any film or residue after scraping can be cleaned with steam which is returned to the extruder-evaporator. With the asphalt there is no dust or free liquid, nor any solids that have to be mechanically chipped away, all of which complicate the health physics problem related to cleaning up such a spill. Also, each drum is monitored and if contaminated it can be cleaned with high-pressure heated water in the decontamination station.

CATALYSTS AND SOLVENTS. No chemical catalysts or promoters are needed and steam (or heated water) is the only solvent recommended in the VRS system. This eliminates the risks normally associated with solvents and chemicals considered to be volatile and undesirable in a radwaste system.

PROCESS UPSETS

LOSS OF POWER AND HEAT. Rarely would the operator need to enter the radiation areas with the VRS system. Even if there is a complete loss of system power and steam heat, the operator does not have to enter the processing area or filling area. What would actually be required is that the operator must shut the system controls off, then simply wait for the power or steam service to be restored, even if the asphalt/radsalts mix in the exturder-evaporator hardens. When power and/or steam is restored, the operator waits until the proper temperature profile in the extruder-evaporator is achieved, then resumes normal operation as if nothing had happened. This is in direct contrast to what happens when an upset occurs with thermosetting binders, which often solidify in place.

CHANGE IN pH VALUE OF THE FEED. The pH value of the VRS radwaste feed stream is recommended to be in the 7-11 range as shown in Figure 6. However, if the pH should drop into the acid range, or rise to high alkaline levels, the processing will continue without disruption. The radwaste solids that go into the container will be immobilized normally as the thermoplastic asphalt hardens.

OIL IN THE FEED. Although oil in the feed may prevent solidification in other systems, thus creating health physics risks, it will not prevent solidification in the VRS system. Oil and asphalt are compatible, both being a petroleum derivative. The net effect is that oil will reduce the viscosity of the asphalt in the end product in a direct ratio according to the amount of oil. The amount of oil normally found in radwaste feed streams, providing it does not exceed one or two percent of the end product volume, will not noticeably affect the VRS end product.

CHANGES IN MIX PROPORTIONS. The VRS will produce a solidified end product even if the proportions of feed stream wastes to asphalt are upset. These are normally controlled remotely from the control panel. Should upset processing conditions become normally uncorrectable by the operator, both visual and audio alarms will be given and the system will shut itself down because of the interlocks designed and built into the system.

REAGENTS. No reagents are used in the VRS system. Therefore, reagent age and temperature effects, which in turn could upset solidification, are eliminated. Other possible effects of using chemical reagents are also eliminated.

END PRODUCT BENEFITS

GENERAL. Again quoting the ORNL/NUREG-43 report (pgs. 58 & 59),[5] "The free-liquid problem took most of the discussion time in the Solidification Workshop held in New Orleans, January 12-14, 1977. Although free liquid may appear in all the solidification systems (with the exception of asphalt which boils the water away), nearly all the complaints came from users of UF resins. Water is an end product in the polymerization of UF resins, and the amount varies according to the proportion of urea and formaldehyde. The age of the reagent and the

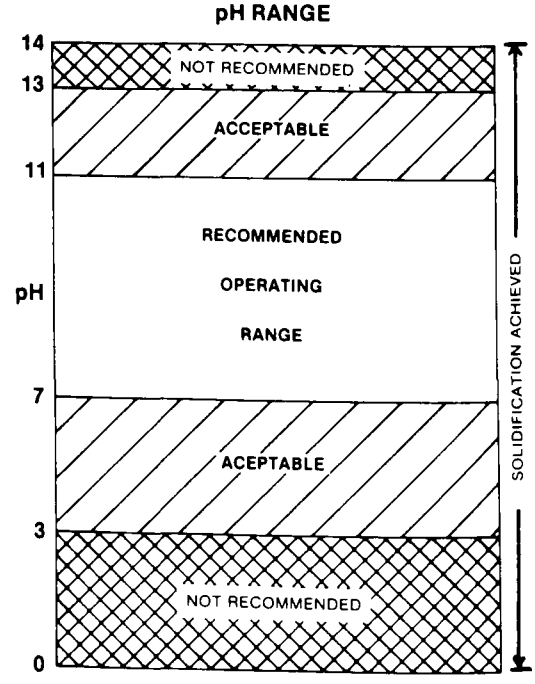

Figure 6. Recommended pH values for VRS system operation.

temperature also have an effect . The broomstick method used in several plants to detect free water was admittedly inadequate, but no one has yet found a more reliable method . . . The incorporation of spent resins into cement requires careful control of the proportions of solid and liquid in the mix to ensure adequate mechanical strength . . . the setting of cement may be retarded or prevented by the presence of organics such as oils and surface tension depresants."

The selection of the above excerpts is not meant to imply that asphalt in the VRS process is perfect, but rather to provide a basis for comparison regarding the most common complaints with respect to use of non-VRS systems. None of the complaints expressed in the above excerpts are present in the VRS system or its end product. The health physics relationship is discussed below.

FREE WATER. There is no end product free water with the VRS system. Water in the feed stream is boiled off simultaneously with the mixing of the radsalt particles in asphalt. Thus, the use of a "broomstick", or any other method to detect free water, is completely eliminated in the VRS system. The net result is no exposure to the operator who otherwise would have to be at the container to detect and, if found, drain free-water.

HOMOGENEOUS END PRODUCT. The mechanical, no free-water mixing process of the extruder-evaporator assures a homogeneous end product. This means no "hot spots" due to clumps of waste or pockets of water.

LOW GAS EVOLUTION. Asphalt resists decomposition due to internal radiation from the contained wastes up to 10^9 rads integrated dose. This is above the 10^7 rads integrated dose normally experienced for nuclear power plants. However, over the time period of storage and burial, gas (hydrogen and/or methane) is generated within all wastes, regardless of the binder used. Cement and other thermosetting binders form a rigid, nonflexible solid that may contain water inside of a hermetically sealed container. Too great a build-up of gases or freezing of the water inside the sealed container could cause the container to rupture and result in health physics problems.

Asphalt, however, has no free-water and the lid on the drum is not hermetically sealed, but rather crimped in place. Also, asphalt is not a rigid, inflexible material. These factors allow any gas generated inside the container to migrate out of the asphalt and out of the container with no internal pressure build-up.

NON-EXPLOSIVE MIX. Asphalt is an inert material. Therefore, it is non-explosive by itself and because of its inherent plasticity, resists external explosive forces. In the weight percentage mixes used in the VRS process, the end product has a negative oxygen balance.[7]

REDUCED DRUM CORROSION. Asphalt is an inert material widely used for its water-proofing and insulating qualities. Thus, the process of internal drum corrosion is reduced significantly, and along with it the health physics problems associated with exposure of the contents to the environment.

ON-SITE STORAGE. The Atomic Industrial Forum has recently published "On-site Low-Level Radwaste Management Alternatives," [8] a report prepared by NUS Corporation. The generation rate of waste for a 1000 MWe nuclear plant was 26,300 cubic feet as solidified with cement, and 5,200 cubic feet when volume reduced and solidified. This report concludes that "Volume reduction of trash and liquids result in savings in all major cost areas: (a) capital expenditures for storage structures, (b) annual operating costs, (c) decommission cost, and (d) waste disposal cost." The present-worth cost for an on-site engineered radwaste storage (containerized storage with VR and solidification) is $24,604,000, as compared with $121,230,000 for shipment of solidified radwastes to commercial shallow land burial facilities 500 miles away and assuming 20% annual escalation in burial rates.

While the economic attraction of saving up to $96,626,000 is outstanding, there are other benefits of using a VRS system with on-site storage. These include less exposure to plant personnel and the public, more favorable public relations, and elimination of a crisis for commercial burial facilities. VRS processing and above ground storage on site is the concept now in use at Eurochemic, Mol, Belgium.[9]

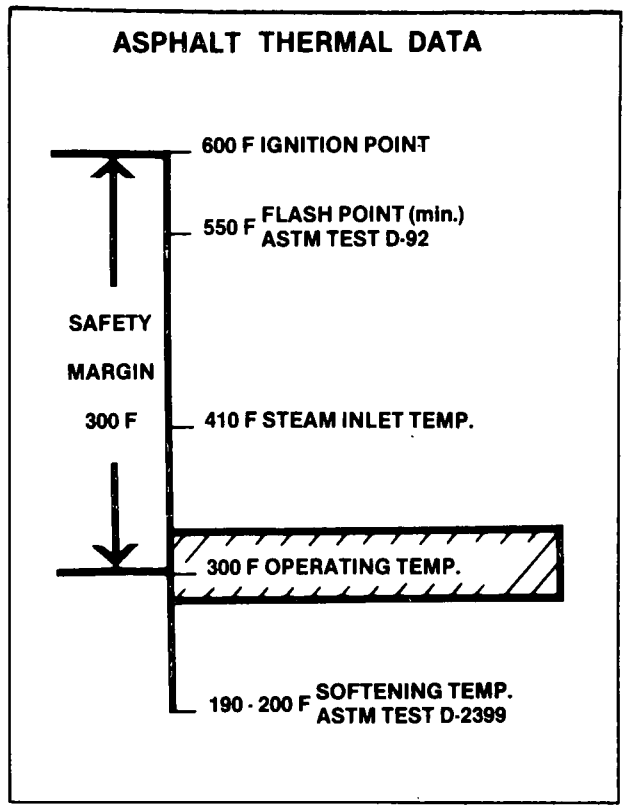

Figure 7. Thermal data of asphalt as used in the VRS process.

PROCESSING AND TRANSPORTATION SAFETY

Safety is a paramount consideration to utilities and radwaste system operators. Safety encompasses several factors such as flammability, explosivesness, and radiation effects. All of these are discussed below.

Thermal data for asphalt as used in the extruder-evaporator process is given in Figure 7. Note that the flammability safety margin during processing is 300 F

Because steam is used to provide process heat, the chance for a hot spot to develop, which is possible with electric heat, is zero.

The National Fire Code, published by the National Fire Protection Association, rates asphalt as follows.

TABLE IV

ITEM	RATING
HEALTH	Zero (Safe and non-hazardous)
FLAMMABILITY	One (Requires significant preheating before ignition can occur)
REACTIVITY	Zero (Safe and non-hazardous)
UNUSUAL REACTIVITY	None

Combined with radwastes, both in the extruder-evaporator and in the end product, the safety characteristics are generally enhanced. For example, the presence of borates, sulfates and other solids raises the ignition temperature (see Table V).

The only place where ignition and sustained burning is of real concern is in the event of a serious accident during transportation to the burial site. The potential hazard of that event has been evaluated by the technical staff of the Nuclear Regulatory Commission and published in their acceptance of the Topical Report [4] on the use of asphalt in the extruder-evaporator process. To quote their evaluation, "On the basis of our Conservative evaluation of the radiological consequences of a severe accident to the proposed system, we conclude that the radwaste volume reduction and solidification (asphalt) system is acceptable."

Another aspect of safety is the effect of an explosive force, which was mentioned briefly earlier in this paper. Asphalt is not explosive either by itself or when mixed with radsalts, nor will an external explosive force cause any damage of any consequence. That fact has been proven in Europe by actual testing sponsored by Eurochemic and conducted by the Royal Military School in Brussels, Belgium. Brittle materials such as cement, urea formaldehyde and polyester, will shatter under explosive forces. However, asphalt normally has sufficient plasticity that it will not shatter, though some deformity may occur.

NET RESULT AT THE BURIAL SITE

The reduction in radwaste volume and the corresponding reduction in the number of containers shipped to the burial site means that the site can accommodate more curies of waste. The lifetime of the burial site is increased five fold or more if all buried wastes are processed in a VRS system.

The container with asphalt encapsulated wastes means that the external drum corrosion is the limiting factor for container integrity. Previously the wastes were acidic and the drums also corroded internally. [10] [11] While no credit is given to drum integrity at this time, the corrosion inhibiting factor of asphalt in the VRS process is an added safety feature. Drums buried with asphalt encapsulated wastes may last up to 15 years.

However, in the event that the drum does corrode, the leach resistance of the non-hydroscopic binder will retain the radwaste in place for an increased amount of time. Both Brookhaven National Laboratory and the Hanford Engineering Development Laboratory [12] [13] have shown the benefits of leach resistance on the retention of the various radionuclides.

TABLE V

ASPHALT

IN PROCESS

CONDITION	ADVANTAGE
WATER IN FEED	WATER IS A FIRE EXTINGUISHER, THUS INHIBITS IGNITION
AS WATER EVAPORATES, SOLIDS TAKE ITS PLACE	SOLIDS GENERALLY RAISE IGNITION TEMPERATURE
VRS OPERATING TEMP. 300F STEAM INLET TEMP. 410F IGNITION TEMP. 600F	300F SAFETY MARGIN IS MAINTAINED DURING PROCESSING

IN THE END PRODUCT

PRESENCE OF BORATES SULFATES & OTHER SOLIDS	IGNITION TEMPERATURE RAISED
PRODUCT DISCHARGE TEMP. IS BELOW PROCESS TEMP.	MORE THAN 300F SAFETY MARGIN
SOLIDIFIED END PRODUCT STORED AT AMBIENT TEMP.	MORE THAN 500F SAFETY MARGIN
ASPHALT IS INERT	SIGNIFICANTLY REDUCED ENVIRONMENTAL IMPACT & CONTAINER CORROSION

THE HUMAN FACTOR

The TV camera system is used by the operator to observe the processing area and the container handling operation. However, one or more lead glass viewing windows are recommended to enable the operator to more naturally observe the operations. The window(s) permit the operator to have a 3-D plus a near natural color view of his operations. Hopefully this combination will enhance his interest and lessen the chances for error.

Many features that are an integral part of the system, such as spray nozzles inside the tanks, flush water connections in transfer and metering lines, steam dome clean-out provisions, etc., are designed to meet ALARA requirements of NRC Regulatory Guide 8.8.

SUMMARY

The use of VRS and the resultant reduction in waste handling and transportation produces many benefits that are of prime importance to the health physicist. The operators still handle the same number of curies but performs significantly fewer operations. This reduction carries over to less exposure time and fewer man-rems.

The reduction in waste handling and transportation using the volume reduction and solidification system with asphalt has the following advantages.

- Removes unwanted water to reduce volume.
- Uses proven binder in a reliable process.
- Elimination of free-water inhibits drum corrosion, permits longer on-site storage.
- Reduces transport trips and lessens accident probability.
- End product is retrievable and/or reprocessible.
- Process and end product accepted by NRC, ANI, EPA, and burial sites.
- Uses system/equipment isolation approach in design to reduce man-rems.
- Economically most attractive system.

All in all the Europeans have researched, developed and proven this system over the past two decades. We have further improved the VRS system to meet the stringent requirements so vital to health physicists.

At the present time no credit is given to leach resistance, however, the industry is investigating methods to make solidified end products better. Several

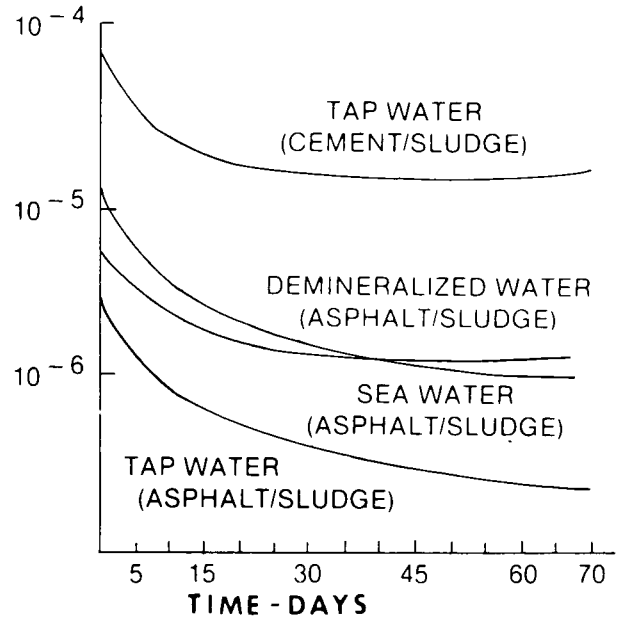

Figure 8. Leach rate test results from AERE, Harwell, England.

establishments in Europe have extensively investigated the leach characteristics of cement and asphalt, some of which are shown in Figure 8 and Table V. This is one of the reasons why asphalt encapsulated wastes were adopted so early in Europe and are gaining in popularity here in the U.S.

A general observation is that the leach resistance of asphalt bound wastes is 100 times better than that of cement in the end product.[14] Whenever the leach resistance needs to be improved, there are methods available. Hydroscopic salts, e.g., sodium sulfate, are the worst for leaching in either cement or asphalt. A thin layer (5 mm) of pure asphalt on the outside of the end product or the inside of the container will virtually eliminate leaching. Another method of improving leach resistance is to add small quantities of calcium hydroxide in place of or in addition to sodium hydroxide for pH adjustment of the radwaste feed.

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SOLIDIFICATION OF LOW-LEVEL RADIOACTIVE LIQUID WASTE USING A CEMENT-SILICATE PROCESS

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Abstract

Extensive use has been made of silicate and Portland cement for the solidification of industrial waste and recently this method has been successfully used to solidify a variety of low level radioactive wastes. The types of wastes processed to date include fuel fabrication sludges, power reactor waste, decontamination solution, and university laboratory waste. The cement-silicate process produces a stable solid with a minimal increase in volume and the chemicals are relatively inexpensive and readily available. The method is adaptable to either batch or continuous processing and the equipment is simple. The solid has leaching characteristics similar to or better than plain Portland cement mixtures and the leaching can be further reduced by the use of ion-exchange additives. The cement-silicate process has been used to solidify waste containing high levels of boric acid, oils, and organic solvents. The experience of handling the various types of liquid waste with a cement-silicate system is described.

Introduction

The cement-silicate solidification process involves incorporation of waste liquids and sludges in a silicate matrix using Portland cement as a setting agent. This paper describes the use of the Chemfix Process developed by Conner (Co74a) and used by the Chemfix Corporation to solidify many millions of gallons of industrial waste liquids and sludges. The process was tested with several types of nuclear waste, including fission product reactor waste, plutonium fabrication waste, and radium contaminated feed plant raffinates, in the period 1969-1973 but it was not used until 1975, when the patent was licensed to Delaware Custom Materiel, Inc. Since then the process has been used to treat over a million gallons of low-level liquid radioactive waste generated in nuclear power plants, fuel fabrication plants, research institutions and hot cell operations. Cement-silicate systems are planned for installation in a number of nuclear reactors now under construction or being planned (Nu76), but there are no in-plant systems now in operation. The cement-silicate process produces a stable solid at low cost with simple equipment and shows great promise for the treatment of low-level radioactive waste.

The Cement-Silicate Process

The cement-silicate process might be better described as a silicate-cement or simply a silicate process. According to Conner (Co74b) the basic

reaction is between soluble silicates and polyvalent metal ions. The compound formed is thought to be based on tetrahedrally coordinated silicon atoms alternating with oxygen atoms in a linear chain, with the metallic ions producing crosslinking between the oxygen side groups. The structure is similar to the naturally occurring pyroxene minerals. The silicate is usually added to the waste as an aqeous solution of sodium silicate, but other alkali silicates in either liquid or dry form can be used. The polyvalent metal ions come from the waste solution, an added setting agent, or both. The setting agent should have a low solubility, but a large reserve capacity of metallic ions, so that it controls the reaction rate. Portland cement is usually chosen as the setting agent because of the ready availability and the additional solidification reactions that it provides as a cement. Lime, gypsum, calcium carbonate and other compounds containing aluminum, iron, magnesium, nickel, copper, chromium or maganese are also reported to be suitable setting agents.

The gel-like structure which is rapidly formed in the reaction entrains large amounts of water or other liquids and prevents the settling of solids during the final setting and hardening process. Reactions such as hydration, hydrolysis, and neutralization also occur between the setting agent and the waste to produce the final product. The process occurs at ambient temperature and pressure and can be used in either a batch or continuous mode.

The amounts of silicate and setting agent necessary to solidify a given volume of liquid waste vary according to the composition of the waste, the desired reaction time, and the preferred characteristics of the solid. Neutral aqeous wastes with high concentrations of metal ions are easily solified with small amounts of silicate and setting agent. Highly acid or alkaline wastes and those containing organic materials may require 2-4 times as much material to produce an acceptable solid. Larger amounts of silicate and setting agent form the gel at a faster rate and produce a harder solid, although too much of either reagent can degrade the quality of the product. The proportions may usually be varied for a given waste to produce a material that will gel in a minute or less for batch processing in drums or a mixture that will flow as a viscous liquid for an hour or more for addition to large lagoons or diked areas with continuous processing equipment. A typical mixture for one l. of neutral pH, aqeous waste would include 80 ml. of sodium silicate solution(density 1.4 g/ml.) and 240 g of Portland cement and would produce 1160 ml. of solid. About 165 l. of this waste could be solidified in the ordinary 200 l. steel drum and still leave about 10 cm of free space above the solid. The weight of the solid would be about 223 kg.

The solid which is formed in the cement-silicate process varies from a moist, clay-like material to a hard, dry solid similar in appearance to concrete. The density varies from about 1.2 to 1.4 g/ml. The large amount of water contained in the solid is not all chemically bound. In open air the solid will dry out and lose water, with some shrinkage, but in a sealed container the solid does not change. When buried without a container it comes to some equilibrium moisture content with the surrounding soil. The moisture loss is not an important factor for sealed containers of most waste, but could be a problem with waste containing tritiated water.

The solid is similar to concrete in that it is not very corrosive to steel. A drum of solidified water was opened after being stored at room temperature for 20 months. The solid still had the same appearance and hardness as when the drum was sealed. The drum, which had a gross weight of 675 pounds, was dropped from a height of 122 cm onto both ends to determine if it would still meet the requirements for a 7A container. There were no visible openings in the drum after the test and there was no loss of contents. The drum was then cut away from the solid and examined. The interior of the drum was corrosion free, except for minor rusting at several points where moisture had probably condensed on the lid immediately after solidification.

The increase in volume of the solid, as compared to the waste liquid, is a very important consideration in any solidification process. As the weight and volume of additives to the waste are decreased, the cost for containers, transportation, and burial is reduced and less burial space is required. In the example illustrated above for water the solid has a volume increase of 16% and a weight increase of 35% over the liquid waste. Using only Portland cement to solidify the water (2 kg/1.) the volume increase would be 66% and the weight increase 300%.

Leaching Studies

One of the important characteristics of a solidified waste material is the susceptibility to loss of the incorporated radioactive material in ground leachate. A standard method has been proposed by the IAEA (He71) for leach testing solidified radioactive waste. However, this method requires a long curing time and periodic sampling over a leach time of many months. It is a good test for the intercomparison of standard samples, but it is not practical for testing a large number of samples to determine the optimum reagent proportions and operating conditions for solidifying a given type of waste.

The following sensitive, yet relatively rapid, leach test was used to compare various wastes solidified with the cement-silicate process. A 25-100 ml. sample of the waste was solidified and stored in a sealed container for several days. The solid was then chopped or ground into small pieces with maximum dimensions of 1-3 mm. A volume of 20-25 ml of the material was weighed and then lightly packed into the barrel of a 30 ml syringe (inside diameter 21.4mm) between glass wool plugs. The leaching solution, which was introduced into the syringe barrel through a tube in a one-hole stopper, passed through the sample and out through a hypodermic needle fastened to the tipe of the syringe. The leaching solution was fed from an aspirator bottle so that the sample was immersed at all times. The flow rate was limited to about 1 ml/min. by crimping the hypodermic needle. Some of the samples had a clay-like consistency and the maximum flow rate achievable was less than 1 ml/min, but the slower rate did not appear to effect the amount of material leached from the sample. Unless otherwise indicated the leaching solution was deionized water.

Approximately one 1. of leachate was collected from each sample. Tests with a ^{137}Cs tracer showed that essentially all of the readily leached material would be removed by this volume of leachate. The leach rate of more tightly bound materials was found to be a relatively constant value after collection of less than one 1. of leachate. The findings agree with previous studies by Connor (Co74b) on the leaching of polyvalent metal ions from solidified industrial wastes.

Table 1 shows the results of leach tests conducted on samples of several fuel fabrication plant sludges solidified with the cement-silicate process. The uranium analyses were done using the delayed neutron activation analysis technique or the etched fission track method. The other constituents were determined by conventional water analysis techniques at commercial laboratories. The concentration of uranium and other metal ions in the leachates was quite low. Some nitrate ion is leached from the solid, as expected, because it is not chemically bound. The low value for chemical oxygen demand (C.O.D.) in the leachate shows that the oils and other organic materials are not readily leached from the cement-silicate solid. This result is somewhat surprising, but was also reported by Conner (Co74b) for industrial wastes.

Table 1

		Concentration in ppm.	
Waste Description	Analysis	Waste	Leachate
Fuel fabrication plant sludge, 34% O.D.S.	U	9.4	$1x10^{-5}$
	Zr	12,000	< 5
	F	340	2.4
	NO_3^- as N	13,660	95
Fabrication plant sludge contaminated with organic solvents, 21% O.D.S.	^{235}U	78	0.005
	C.O.D.	36,480	95
	Fe	1,640	< 0.01
	Zn	158	< 0.01
Fuel fabrication plant sludge, 32% O.D.S.	U	7.1	< 0.003
	Ni	51	< .03
	Pb	23	.04

Unlike polyvalent metal ions, monovalent ions are not chemically bound in the silicate reaction and are readily leached from the solid. This has been demonstrated using the leaching test described above with a ^{137}Cs tracer. In an effort to improve the cesium retention a number of additives to the cement-silicate process were tested. The addition of shale to the mixture greatly decreased the amount of cesium leached from the solid. Excellent results were obtained with Conasauga shale from Oak Ridge, Tennessee and from unidentified shale samples obtained in the vicinity of Port Matilda, Huntington, Johnsonburg, and Bradford in Pennsylvania. Tests were also made with sodium bentonite (Wyoming or western bentonite), calcium bentonite (southern bentonite), and three different illite clays. The shales and clays were ground fine enough to pass a U.S. #40 sieve and added to the waste mixture with the setting agent. The samples were made by solidifying

a 5% Na_2SO4 solution tagged with ^{137}Cs, using the equivalent of 400g of Portland cement, 200 ml. of sodium silicate solution, and 160 g of shale or clay per 1. of solution. Samples were also prepared using only Portland cement for solidification in the proportion of 2 kg per 1. of solution. The relative leachability is expressed as the specific leach fraction, which is the concentration of ^{137}Cs in the leachate (μCi/g) divided by the concentration of ^{137}Cs in the solid (μCi/g).

The results of the leach tests with the shale and clay additives are shown in figure 1. The Conasauga shale additive produced a marked reduction in cesium leaching in both the cement-silicate and the cement only solids. However, it was much more effective in the cement-silicate process than with cement only. Both bentonite clays produced about the same reduction, but the illite clays showed a wide variation. The Beavers Bend, Oklahoma sample showed the greatest reduction, the Goose Lake, Illinois sample the least reduction, and the Fithian illite from Illinois was intermediate. Samples in which ground glass replaced the shale or clay, to determine the effect of the bulk and surface area of the additive, did not show any reduction in cesium leaching. Tests were also made with the Conasauga shale additive in solids produced with plain water and with a simulated waste, W-7, used in some leach studies of grouts at the Oak Ridge National Laboratory (Mo76). The W-7 waste is highly alkaline and contains large concentrations of sodium, nitrate, carbonate, and sulfate ions. The specific leach fraction for the solid made with water was 2.6×10^{-5} and for the solid from the W-7 solution 1.9×10^{-5}, or almost the same as the average value of 1.5×10^{-5} obtained with the 5% Na_2SO_4 solution. The amount of cement and silicate used for the water solidification was about half that for the 5% Na_2SO_4 and for the W-7 solution about 5% more than for the 5% Na_2SO_4 solution.

The solid produced by the cement-silicate process has been suggested as a liner and covering for sanitary landfills to remove metal ions and other materials from any leachate passing through it (Co74c). This technique could also be used at radioactive waste burial grounds. A sample of cement-silicate solid prepared using water and shale additive was used to filter ^{137}Cs tagged water and ^{137}Cs tagged leachate from a cement-silicate solid without the shale. In both cases essentially all (99.98%) of the ^{137}Cs was removed by the cement-silicate filter with the shale additive. Thus the presence of waste solidified by the cement-silicate process could help prevent the release of materials leached from other wastes.

Field Experience

The cement-silicate process has been used to solidify many types of nuclear waste. One application involved about 330,000 1. of evaporator concentrates containing fission products in the 1-10 mCi/1.range from decontamination and hot cell operations. The mixing was done in concrete disposal containers of up to 6000 1. capacity. Processing was completed in less than one week, using field-assembled equipment and working outdoors. Even though working conditions were adverse and the equipment less than optimum the project was completed without significant radiation exposure or contamination problems. A number of the concrete containers were not water tight

and liquid started to seep through them after filling. However, the seepage stopped almost immediately after the waste was solidified. The open top containers were allowed to stand uncovered for many months before moving to the disposal site. During this period there was some drying and shrinking of the solid allowing precipitation to get between the container wall and the solid. Some of this contaminated liquid leaked through the walls and caused minor contamination.

Continuous processing at rates of 700-1100 1./min. has been used in the treatment of sludges contaminated with very low concentrations of enriched uranium. The treated sludges were pumped to a diked area for solidification and storage on-site. The solid was firm enough to be walked upon in about twenty four hours and was later covered with earth to prevent infiltration of rain water. Such sludges are usually processed so as to produce a solid that can be handled with convential earth moving equipment. The material can then be easily moved at some future date, if necessary.

The nuclear reactor waste that has been processed to date has been mixed in open 200 1. steel drums using a portable mixer. The volume of waste which can be mixed in a drum depends upon the amount of silicate and setting agent required, but is usually 145-165 1. A large amount of the reactor waste has contained oils from the turbine and other sources. The oil concentration is adjusted to 30-50% of the liquid volume, using other aqeous waste when possible, and the oil is emulsified with 4-6 1. of detergent prior to solidification. Boric acid waste is neutralized with lime or sodium hydroxide before solidification. Lime is preferred because it also acts as a setting agent. Low concentrations of acid do not require pretreatment because of the excess alkalinity in the cement. Dewatered or slurried reactor resins are easily solidified with the cement-silicate process. The small amount of solidification chemicals used results in a volume increase of 10% or less. However, a relatively powerful mixer is required to adequately mix the resins with the solidification agents. Most of the reactor waste has been material which could not be processed with the station waste treatment system. Processing has been done in whatever space available, even outdoors, using portable equipment. A two man crew can solidify about 80 drums of waste per day.

Batch processing in 200 1. drums has also been used to solidify a large quantity of process waste at a high-enrichment uranium fabrication facility. This waste contained a large proportion of organic solvents but was successfully solidified using high ratios of silicate and setting agents. Research laboratory waste containing 20% or more of liquid scintillation fluid and various other organic solvents has been routinely solidified. A volume of 150-170 1. of waste is usually processed in each drum. The waste collected from the various laboratories is combined in a 200 1. drum and test mixes are tried on 25 ml samples. Because of the varying composition of wastes received from the research laboratories, each drum is tested to find the optimum proportions of silicate and setting agent. The processing is easily done by one or two persons using simple, inexpensive equipment and the direct cost is less than that of previous methods using only absorbents, which are no longer acceptable at licensed landfills.

When processing waste that is difficult to solidify, it is not uncommon to have a small volume of liquid that is not incorporated in the solid. If this occurs it is easily observed, because the liquid collects at the top of the container. The solid formed under the liquid is of good quality and can be used. Small amounts of excess liquid (1-2% of the waste volume) are usually taken up by the solid as the hydration and setting of the cement progresses. Larger volumes may be decanted to another container for further processing or dry cement can be added to the container to incorporate the liquid residue.

Equipment Requirements

Mobile processing units on semi-trailer chassis have been used by the Chemfix Corporation to solidify industrial waste in a continuous mode at rates of about 1100 l./min. The units contain storage tanks for silicate solution and cement plus a hopper with an agitator for mixing the liquid waste with cement. The waste is pumped to the van, mixed with cement in the hopper then pumped to a receiving lagoon. The silicate is injected into the suction of the discharge pump so that it is mixed with the waste just before discharge. These high flow rate units have been used to solidify large volumes of sludge contaminated with uranium, but the units are not designed to handle waste with a high concentration of radioactive material. There are no provisions for shielding or complete containment of the waste stream. However, the cement-silicate process involves only mixing and transfer of materials at ambient temperature and pressure. Conventional off-the-shelf hardware could be used to build a continuous process system for use in a confined shielded area.

Batch processing is readily accomplished within a 200 l. drum. The drum is filled with the liquid waste and mixed while the cement is added. Silicate solution is then added quickly and the mixer is operated for a few seconds more, before removing it from the drum. A 1/3-3/4 hp. mixer is sufficient for liquid waste, but heavy sludges or resin beads require a mixer of 2-3 hp. Air-powered, gear-reduction mixer motors are ideal for this purpose because of the light weight, variable speed, and the ability to be stalled without damage. A small air mixer mounted on the lid of a 200 l. drum along with a funnel for cement and a feed pipe for silicate solution addition has operated very well for processing research laboratory waste. A large amount of waste has also been processed in open drums, adding the cement and silicate by hand and mixing with a clamp-on drum mixer.

A solidification kit for processing waste in a 200 l. drum has been developed by Delaware Custom Materiel, Inc. The kit consists of a drum containing a disposable mixer blade, with the shaft held by bearings welded to the inside of the lid and bottom of the drum. The upper end of the shaft is accessible through a bung in the lid for turning with an external motor. The cement can be added to the drum before it is capped. The liquid waste and silicate are added through the bungs in the lid. This technique reduces the possibility of spreading radioactive contamination during the filling and mixing operation. An air-driven motor is clamped to the drum lid to turn the

mixer.

Batch processing can be done in disposal containers much larger or smaller than the 200 l. drum. In a previously mentioned project the drum mixer was scaled up and used to mix single batches in 3800-6600 l. concrete tanks. At the other end of the scale, prepared kits containing sufficient silicate and cement to solidify 500 ml. of waste are being used by the radioisotope users at a university. The materials are packaged in a one l. steel can, which is the mixing vessel and the disposal container. The waste is added to the cement in the can and diluted to 500 ml. Then the silicate is added and the can is capped and shaken for a few seconds. The waste is solidified in less than a minute and discarded with the other solid radioactive waste. This procedure is used only for small volumes of waste containing relatively large amounts of radioactive material. The more hazardous liquid waste is immediately fixed in solid form. This keeps the concentration in the large volumes of low-activity liquid waste at the level where handling it is not difficult.

Summary

The cement-silicate process involves the interaction of soluble silicates and a setting agent with liquid waste to produce a solid material suitable for disposal. The silicate reaction chemically binds polyvalent metal ions so that the ions are not readily leached from the solid and shale additives can be used to reduce the leachability of radioactive cesium.

The chemicals used in the process are inexpensive and readily available. The volume increase of the waste in the solidification process is small resulting in further savings of container, transportation, and burial costs, as compared to other solidification processes. The reaction rate and the chemical and physical properties of the solid can be controlled to allow processing by continuous or batch methods and to suit the disposal site or container.

The process has been used to solidify many types of low-level radioactive waste including those which contain oils and other organic materials. Processing equipment is simple and can be built with readily available hardware, making the process suitable for low-volume and high-volume applications.

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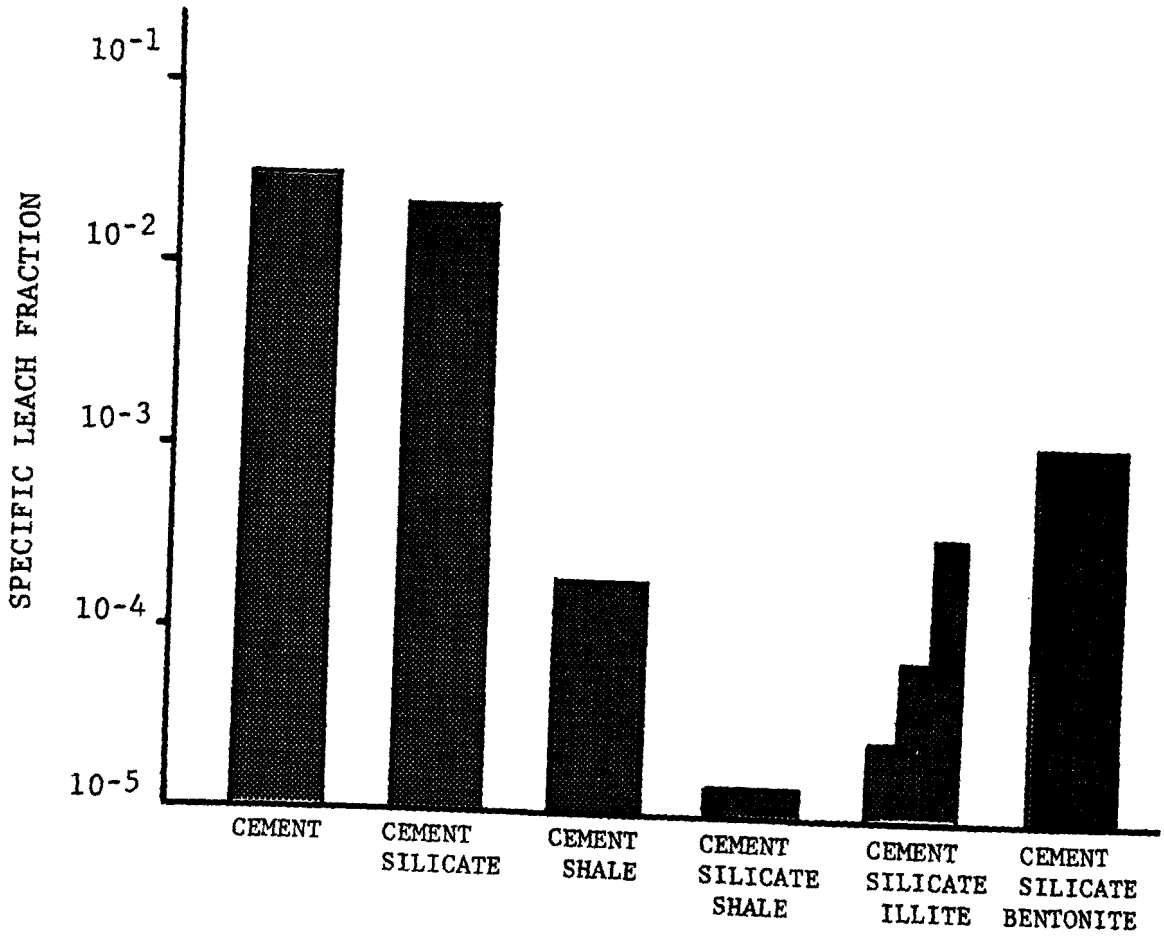

FIGURE 1
CESIUM-137 LEACH TESTS FOR
5% Na_2SO_4 SOLUTION SOLIDIFIED
WITH CEMENT AND CEMENT SILICATE

COLLECTION AND DISPOSAL OF LOW LEVEL WASTE AT AN EDUCATIONAL INSTITUTION

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Abstract: Low level radioactive wastes are generated by a number of different laboratories and departments at the University of Virginia. Radioactive materials are utilized in a variety of research applications including medical and basic sciences, as well as for diagnostic and therapeutic uses at the University Hospital. Radioisotopes are purchased from commercial sources and are produced locally for use in research and medical diagnosis and treatment by the University of Virginia Reactor. In 1974, the University Radiation Safety Committee adopted rules for discharging radioisotopes to the environment which are more restrictive than the Nuclear Regulatory Commission regulations. The committee's philosophy is that no radioactive substances should be discharged to the environment which can be reasonably avoided, including those used in medical diagnosis and therapy. This policy has caused a significant increase in the accumulation of low-level radioactive wastes. The volume of low-level wastes at the University has increased from about 1.5 M^3 in 1969 to over 68 M^3 in 1977. Disposal costs have increased proportionately. Currently the University employs a full-time technician to collect and package radioactive wastes under the supervision of the health physics staff of the Radiation Safety Office. In 1976, the Radioactive Waste Management Facility (RWMF) was completed. This facility houses the Radiation Safety Office staff and has modern facilities for collecting and packaging all types of radioactive wastes. The facility is being used to limit the total cost of radioactive waste disposal, while fulfilling the objectives of the Radiation Safety Committee. Methods used to limit waste disposal volumes and costs are compaction, storage and decay of short half-life isotopes, solidification of liquid wastes, and education and training of radioactive material users throughout the University in reducing waste volume.

Introduction

The University of Virginia holds a Nuclear Regulatory Commission (NRC), Type "A" Broad By-Product Material License, an NRC "Institutional" Reactor Operations License, and a Virginia State License for ionizing radiation producing equipment and radioactive material not regulated by the NRC. The Radiation Safety Committee has administrative control over all uses of radiation and radioactive materials at the University including medical therapy and diagnosis. The Radiation Safety Committee is so structured that the Radioactive Drug Research Committee, required by Food and Drug Administration regulations also functions as the Medical Isotope Subcommittee, an advisory body on medical uses of radiation in diagnosis and therapy.

The University Radiation Safety Officer is a member of the general faculty and is appointed by the Chairman of The Radiation Safety Committee. The Radiation Safety Officer directs the operational unit involved in radioactive waste management, and implements the rules and regulations promulgated by the Radiation Safety Committee for the safe use of all ionizing radiation sources at the University.

The Radiation Safety Office staff consists of professional health physicists, technicians and clerical personnel for the routine operation of the Radioactive Waste Management Facility and the University's radiation safety program.

Facilities Generating Radioactive Wastes

The University is arbitrarily divided into three areas for waste management purposes. These are the reactor facility, which houses a 2 Megawatt swimming pool reactor, an 80 watt training reactor and experimental and research laboratories; the academic departments, which include radioactive materials and radiation use in the Van de Graff accelerator, the Physics Department and the Departments of Biology, Chemistry, Materials Science and Environmental Science; and, the Medical Center, including a medical school, basic science research laboratories, clinical research laboratories and the hospital which has active nuclear medicine and radiation therapy departments.

Of the above, the medical center generates by far the greatest quantity of radioactive waste. Approximately 90% of all radioactive wastes at the University originate within the medical center.

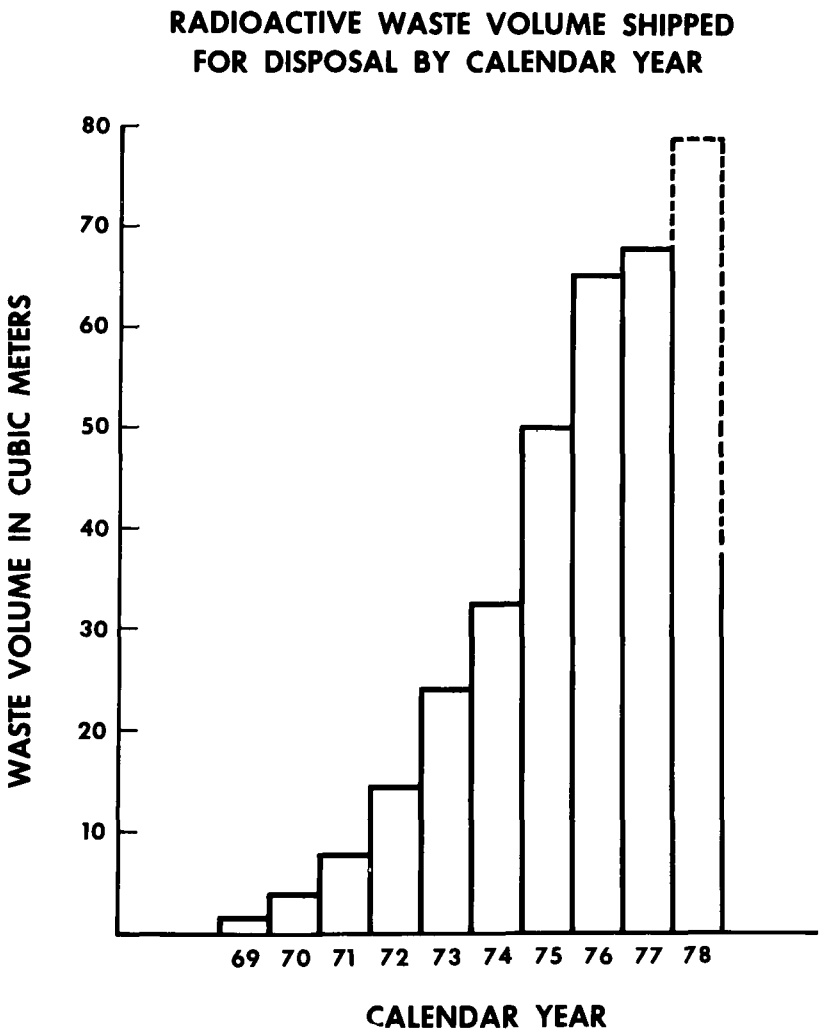
Development of Radioactive Waste Management

Prior to 1972, the Reactor Facility staff collected and disposed of their own radioactive wastes and those generated by the academic departments, while the Medical Center maintained a completely separate radioactive waste collection, storage and disposal operation. My mid-1972, the Radiation Safety Office was established as a service department and a University-wide program of waste collection, storage and disposal was begun. Initially, the coordination was only administrative and the dual collection and storage operations continued to function under the broad supervision of the Radiation Safety Office. When a new Medical Education Building was put into service in late 1972, a third collection and storage operation was organized to accomodate more than 60 laboratories using radioactive materials in that building.

In 1974, the Radiation Safety Committee revised the rules and regulations concerning discharge of radioactive materials. The revision stated that no radioactive materials would be discharged to the sanitary sewer system if it could be reasonably avoided. This regulation had a significant impact on the quantity of radioactive wastes collected (figure 1). By the end of 1974, it was apparent that the waste collection and storage operation was costly and inefficient with the increased volumes of waste being handled, due to both the increasing use of radioactive materials and to prohibition of low level radioactive waste discharge to the sewer system. A subcommittee of the Radiation Safety Committee was appointed to study the waste disposal operation and make recommendations for improving the cost effectiveness of waste handling.

Based on the recommendations of this Subcommittee, planning was begun in late 1974 for a centralized radioactive waste management facility. Funds were obtained through the University Research Policy Council and the facility

Figure 1.


RADIOACTIVE WASTE VOLUME SHIPPED
FOR DISPOSAL BY CALENDAR YEAR
WASTE VOLUME IN CUBIC METERS
80
70
60
50
40
30
20
10
69 70 71 72 73 74 75 76 77 78
CALENDAR YEAR

was completed in June 1976. A technician was added to the Radiation Safety Office staff to assume primary responsibility for radioactive waste management under the direction of the health physics staff. An additional secretary was also added to help staff the new facility. The Radiation Safety Office staff currently consists of three health physicists, one technician and one full-time and one half-time secretary plus a paid student position which is normally filled by a graduate student in the Nuclear Engineering Department.

Radioactive Waste Management Facility

The 2500 square foot waste management building contains a small laboratory used for environmental and bioassay sampling, a large waste handling room, office space for two health physicists and clerical staff, and a small emergency decontamination facility consisting of a shower, washer and dryer and clothing supply lockers. The shower, washing machine and floor drains in the waste handling room and decontamination room are connected to a hold up tank which is designed to hold contaminated water until it can be sampled. A recirculation pump and demineralizer system is provided to remove radioactivity from the decontamination water prior to discharge to the sewer system. An outside concrete pad and storage cabinet provide a storage area for flammable liquid.

The large waste handling room contains a compactor to compact solid wastes in 55 gallon barrels to obtain a more cost-effective package. The compaction ratio for most waste collected is 4 to 1. A barrel handling crane is provided so that one man can easily manipulate compacted drums in the storage area. The room also has a fume hood which is used for opening packages of radioactive materials as they arrive at the University. All packages containing radioactive materials entering or leaving the University are processed through the waste facility.

The floor of the waste handling room is covered with an epoxy resin for easy decontamination. An area monitoring system with local meters and remote indication at the secretary's deck provide constant monitoring of the radiation levels within the waste room.

Waste Packaging and Shipment Procedures

Radioactive wastes are picked up from the users' laboratory on request and are brought to the waste facility for packaging. Each radioactive material user is supplied with both solid and liquid waste containers. These containers are not currently standardized and users are permitted to use containers other than those provided if they are approved by the Radiation Safety Officer. The collected wastes are segreated according to half-life (Table 1) and are then packaged for shipment or set aside for decay. Short half-life isotopes are held for approximately 10 half-lives and then monitored. If the radioactivity has decreased to background levels, the waste is disposed of as routine non-radioactive waste material. Occasionally, when waste volumes become prohibitively large, some short half-life material is, by necessity, shipped for disposal.

Typical Radioisotopes in Collected Wastes
TABLE 1

Long Half-Life*	Short Half-Life*
^{3}H - 12.2 years	^{24}Na - 15 hours
^{14}C - 5600 years	^{32}P - 15 days
^{35}S - 87.2 days	^{51}Cr - 27.7 days
^{45}Ca- 163 days	^{99}Mo-^{99}Tc - 66 hours
^{54}Mn- 312 days	^{127}Xe- 36.4 days
^{55}Fe- 2.7 years	^{131}I - 8 days
^{57}Co- 271 days	^{133}Xe- 5 days
^{60}Co- 5.2 years	^{141}Ce- 32.2 days
^{75}Se- 120 days	^{201}Tl- 73 hours
^{90}Sr- 30 years	
^{125}I- 60 days	

* Classified for Disposal Purposes

Solid wastes are placed directly into 55 gallon drums and compacted. Liquid wastes are poured into 30 gallon drums filled with vermiculite, which are then packed in 55 gallon overpack drums with the additional space filled with vermiculite. Animals and biological tissues are packed in 30 gallon drums and stored in a large walk-in freezer until shipment. Liquid scintillation vials are left intact and are packed in 55 gallon drums with alternate layers of vermiculite. All containers are currently being supplied by Teledyne Isotopes of Westwood, New Jersey. The containers are prepainted with the appropriate labeling and meet all regulations for shipment. Of course, detailed records are kept of all waste collection and disposal.

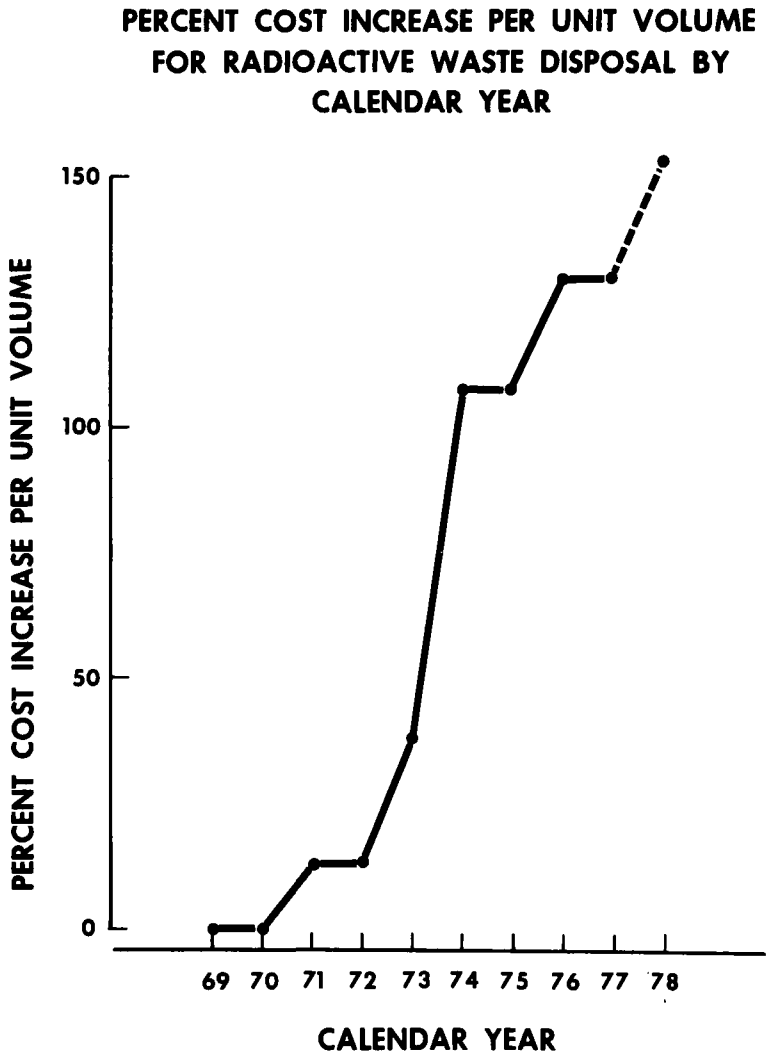
The use of radioactive materials continues to expand within the University and the cost of waste disposal continues to rise (figure 2). The current method of waste collection and disposal seems to be the best approach to the problem within the restrictions placed on radioactive material use by the licensing agencies and the University's Radiation Safety Committee.

Objectives of the Waste Management Program

The objectives of the current waste management program are to provide the most cost-effective and efficient radioactive waste disposal while maintaining a good relationship between the University and the surrounding community. By restricting the discharge of radioactive materials to a greater extent than allowed by the NRC or State regulations, the University is demonstrating its' commitment to the safe use of radioactive materials within the community.

These objectives are being met by insuring that all radioactive wastes are properly handled and contained, and by a continuing education and information program for radioactive materials users, employees and staff of the University, and members of the general public.

Figure 2.


PERCENT COST INCREASE PER UNIT VOLUME
FOR RADIOACTIVE WASTE DISPOSAL BY
CALENDAR YEAR
PERCENT COST INCREASE PER UNIT VOLUME
150
100
50
0
69 70 71 72 73 74 75 76 77 78
CALENDAR YEAR

COLLECTION AND HANDLING OF RADIOACTIVE WASTES FROM A LARGE UNIVERSITY

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Abstract

Various types, levels, and amounts of radioactive wastes are generated by the University of Missouri-Columbia research, teaching, and clinical programs. Health Physics Services is responsible for collecting these wastes from the more than 200 laboratories spread over the sprawling campus and disposing of the wastes in an economical and safe manner. Most of the radioactive wastes are laboratory paper trash and liquids containing low levels of activity but some high level wastes and contaminated large animal carcasses also require disposal. The wastes are stored at one of four interim storage locations until ultimately disposed of by incineration, sewer release, local burial, shipment for commercial waste disposal or decay. The economics, safety, and handling aspects of the various disposal methods must be considered.

Discussion

Health Physics Services at the University of Missouri-Columbia (UMC) is responsible for the disposal of all radioactive wastes produced by the Columbia Campus. The radioactive wastes are generated by the teaching, research, and clinical efforts of the basic sciences departments, the School of Medicine, the University Hospital, the College of Veterinary Medicine, and the College of Agriculture located on the sprawling campus. The waste handling responsibilities of the group also extend to the UMC medical research laboratories in Saint Louis which is 125 miles from the main campus.

The radioactive wastes generally are the normal solid and liquid radioactive wastes from a basic science laboratory such as paper, gloves, scintillation fluids, vials, and other disposables. In addition, wastes also range from technetium milk from molybdeum cows to radioactive milk and manure from dairy cows and even at times to the cows that produce it.

The types of waste generated by such diverse sources as those found on the Columbia Campus require a flexible system of waste handling. The system must be able to cope with wastes ranging from the low-level trace activities in liquid scintillation fluids to multicurie activities produced from high specific activity labeling procedures. In addition, the waste handling system must be capable of contending with waste containers ranging from one milliliter shipping vials through bags of trash and bottles of liquids to pony and cattle carcasses.

The system must be able to absorb the expanding volumes of wastes resulting from increasing numbers of authorized users and their radioisotope laboratories (table 1). Growth of the radioisotope program of the campus is indicated by the 22% increase in the number of authorized radioisotope users over the past five year period and even more directly by the 38% increase in the number of radioisotope laboratories. As would be assumed the number of waste pickups and waste volumes correlate with the increasing number of authorized users (table 2).

The annual number of waste pickups has increased about 43% over the past five years while the number of containers or items collected increased by about 30%. These increasing volumes are more dramatically indicated in the disposal figures (table 3). Solid waste volume handled by the waste disposal system increased by 104% over the last five years while liquid waste disposals increased even more showing a 143% increase.

The increasing volumes of waste entering and leaving the radioactive waste disposal system are indications of several subtle influences. The function of a university is to train and educate students in the modern techniques needed for the sophisticated research now prevalent. This training produces young faculty members and research investigators trained in the use of radioactive materials and in the methods of applying them to advance research interests. These younger faculty members tend to rely more on research methods utilizing radioactive materials than did their predecessors. Coupled with the researchers increasing use of radioactive materials is their ability, aided by the improved instrumentation, to use the radioactive tracers more efficiently. For example, we have noticed that investigators performing iodination of proteins for radioimmunoassay procedures require decreasing activities of iodine as their techniques improve. Iodinations which initially took four or five millicuries to complete may only require one-half to one millicurie to provide the stock material for several months of assays.

The functions of the waste handling system can be separated into two areas of responsibility: 1) the authorized user and 2) Health Physics Services.

The Authorized User

The authorized user, as the starting point in the waste flow process, is the most critical in the waste disposal program. Only the user can predict with any degree of certainity how the radioactive material he uses fractionates among the types of waste that are generated. Solid waste is almost impossible to assay and liquid waste is difficult to assay with any degree of confidence, particularly for low energy beta emitters. Health Physics Services does not have the time available to expend in assaying all the waste collected. It is a requirement that the user be responsbile for recording the radionuclides and activities in the wastes before the waste is removed from the laboratory.

Radioisotope laboratories are supplied with either of two sizes of plastic-lined fiberboard containers lined with polyethylene bags for solid wastes and with one gallon polyethylene jugs for liquid collections (table 4). Other types of containers such as paint cans, small polyethylene containers and steel drums are supplied as needed for specific handling, containment, or volume problems. The user disposing of scintillation vials is instructed to repackage them in their original containers for disposal, however, in some cases the vials are collected in the fiberboard or steel drums.

It is the policy that Health Physics collects and processes all radioactive waste. Sewer discharges from laboratories are discouraged except for some specific instances involving large volumes of low-level wastes that are difficult to collect and transfer. For washing of contaminated glassware, the laboratory personnel are instructed to rinse the container into the liquid waste jug until the remaining activity approaches background levels. The

glassware then can be washed with no recorded activity to the sewer. These procedures have reduced the sewer discharge for the Columbia Campus users to less than ten millicuries per year excluding, of course, those radioactive materials administered to humans for diagnostic or therapeutic procedures.

Animal carcasses containing radioactive material are held for disposal by freezing or refrigeration. The carcasses are stored in freezers at the authorized users' locations or transferred to Health Physics for storage in the freezers in the waste storage facilities. The stored carcasses are wrapped in plastic and labeled according to date, isotope, activity, authorized user, or identification number.

Health Physics Services

Waste pickups are performed upon user request. The requests from the users are made by telephone or by interoffice mail correspondence. The form of the waste as to solid, liquid, or animal, isotopes and activites are identified at the time of the request. Health Physics personnel can then arrive at the laboratory with appropriate replacement containers. Health Physics has two vehicles which are used for the radioactive waste collection, a van and a station wagon. Pickups are usually made within one day and often within a few hours of the request. At the laboratory isotopes and activities in each containers are confirmed, the bag liners in the solid waste containers are taped closed, and the waste transferred to the vehicle with a two wheel cart. Upon reaching the radioactive waste storage building, waste information is recorded on radioactive waste record cards (figure 1) and a sequential identification number assigned to each container. The container is then tagged with identification number, isotopes, activities, and date. Waste containers are segregated according to form and half life in the radioactive waste storage building.

The radioactive waste storage building is located next to the Health Physics Services building in the Research Park (figure 2). The building is a 728 square foot brick veneered concrete block building on a concrete slab foundation. A flammable storage room occupies one corner. Scintillation vials and other liquids are stored on steel shelving in this room. Room air is exhausted at a rate of six air changes per hour by an explosion proof fan. Two 20 cubic feet chest type freezers for small animal carcass storage are in the large room. The remainder of the building can be used for storage of solid wastes, empty containers, and usually one of the vehicles. The building provides storage for about six months of waste accumulation at our current collection rate.

Additional waste holding facilities are located at the Medical Center where a 60 square feet storage room and two 20 cubic feet chest freezers are available and at the University waste disposal site about five miles southwest of the campus where a storage building with a waste compactor is available.

Ater the collected solid waste approaches a volume of about 200-500 cubic feet of low-level materials, a burial is made at the local radioactive land burial site. A backhoe is scheduled to dig an appropriately size burial trench, which is generally twelve feet deep, two to four feet wide, and ten to fifteen feet long. Burials are performed within the requirements of 10 CFR 20.304.

Containers are checked for isotopes and activities and loaded for transport to the waste disposal site. Identifying tags are removed from the containers as the liners are emptied from the containers into the trench. Upon return to the office the tags are cross checked with the waste disposal cards and activities summed by isotope to assure compliance with 10 CFR 20.304. Just before the trench is backfilled the frozen animal carcasses are transferred from the freezers to the site, identifying tags removed, and the carcasses placed in the trench. The burial is given an identifying number, located on the burial site map, containers are recorded as buried on the waste disposal cards, and the burial report completed.

An incineration amendment to the University NRC license allows open pit incinerations. We have restricted individual incinerations to less than 100 gallons of liquid and less than one millicurie per liter specific activities. No more than five incinerations per week and less than 52 per year are permitted, however, the frequency of incinerations has been much lower than this allowable rate. Records for the incineration are prepared and maintained in a manner similar to those for the shallow land burials.

Wastes with activities too large for disposal by local burial or incineration are held for shipment to a commercial disposal firm. Isotopes with half lives of less than 100 days are routinely held for decay to levels acceptable for shallow land burial or incineration. This policy has resulted in an accumulation of stored iodine-125 waste due to the recent increase in iodinations for radioimmunoassays.

Safety margins inherent in the system are: No container is assigned an activity of less than one microcurie, burials and incinerations tend to average from 40 to 60% of the allowable 100%, and isotope decay is estimated somewhat conservatively. Also all radioactive waste collected from laboratories is considered to be radioactive and accordingly disposed. No radioactive waste is disposed as normal waste even though the external radiation measurements may be at background levels.

The waste disposal program is a major part of the health physics program at the University of Missouri-Columbia. It is estimated that about one-fourth of the effort of the six person group is devoted to waste disposal. Also included are the costs for containers, local burial excavations, commercial disposal charges and one-half of the operational costs of the two vehicles. The waste disposal system is able to move radioactive wastes from a laboratory or use area to an ultimate disposal in a safe economical manner while producing a minimal environmental insult. It is felt that our simple and flexible waste disposal system can cope with the various wastes and volumes produced by a major research and teaching institution.

TABLE 1

Authorized Users and Radioisotope Rooms by Fiscal Year

	1973-74	1974-75	1975-76	1976-77	1977-78
Authorized Users	123	127	137	143	150
Radioisotope Rooms	167	174	215	218	230

TABLE 2

Radioactive Waste Totals by Fiscal Year

	1973-74	1974-75	1975-76	1976-77	1977-78
No. of Pickups	282	309	354	376	402
Containers Solid	231	443	288	309	298
Gallons Liquid	736	694	711	852	973
Scintillation Vials (Gallons)*	(-)	(-)	(299)	(355)	(333)
Animals (Bags)	-	-	56	105	40
Activity	-	-	-	12,681	44,966

*100 filled vials ≃ 0.25 gallon

TABLE 3

Radioactive Waste Disposed by Fiscal Year

	1973-74	1974-75	1975-76	1976-77	1977-78
Local Burial					
Burials	4	4	5	10	8
Volume (Ft^3)	707	678	1028	1311	1510
Activity (mCi)	1046	1080	559	176	208
Incineration					
Incinerations	10	9	11	12	14
Volume (gallons)	500	539	755	925	1213.5
Activity (mCi)	95	95	116	94	130
Sewer Disposal (mCi)	1.000	0.25	0.25	6.400	3.000
Commercial					
Shipments	1	1	2	1	1
Volume (Ft^3)	56.8	58	43	15.5	45
Volume (Gallons)			45		
Activity (mCi)	5,354	1,550	3,797	230	15,434
Cost	200.00	370.00	155.00	0	0

TABLE 4

Item	Source		Cost
Plastic jugs (1 gallon)	University Storeroom		$ 0.40 each
Plastic bags (6 mil)	Home Plastics, Inc. Des Moines, Iowa		0.42 each
Fiberboard drums	Continental Can Co.	28 gal.	13.50
Inside plastic	Overland, Missouri	18 gal.	7.50
Silk screened			

FIGURE 1

UNIVERSITY OF MISSOURI

RADIOACTIVE WASTE RECORD **CAMPUS** Columbia

Pick up No	Authorized User	Isotope & Form (S or L)	Quantity (mCi)	Disposition (Date)			
				Storage	Incinerate	Burial	Shipment
1753	Johnson, H.	H-3 S	0.200	8/12/78		8/22/78	
1754	Johnson, H.	H-3 Animal	0.050	8/12/78		8/22/78	
1755	Johnson, H.	C-14 S	5.000	8/12/78		8/22/78	
1756	Johnson, D.	H-3 Animal	0.110	8/14/78		8/22/78	
1757	Johnson, H.	H-3 L 1 gal	0.010	8/15/78	8/19/78		
1758	Johnson, H.	H-3 L 1 gal	0.010	8/15/78	8/19/78		
1759	Johnson, H.	H-3 S	0.100	8/18/78		8/22/78	
1760	Pickett, E.	Se-75	0.010	8/18/78			

Radsafe 23-5-78

CAUTION

1760

RADIOACTIVE MATERIAL

ISOTOPE 75Se

AMOUNT 10 μCi

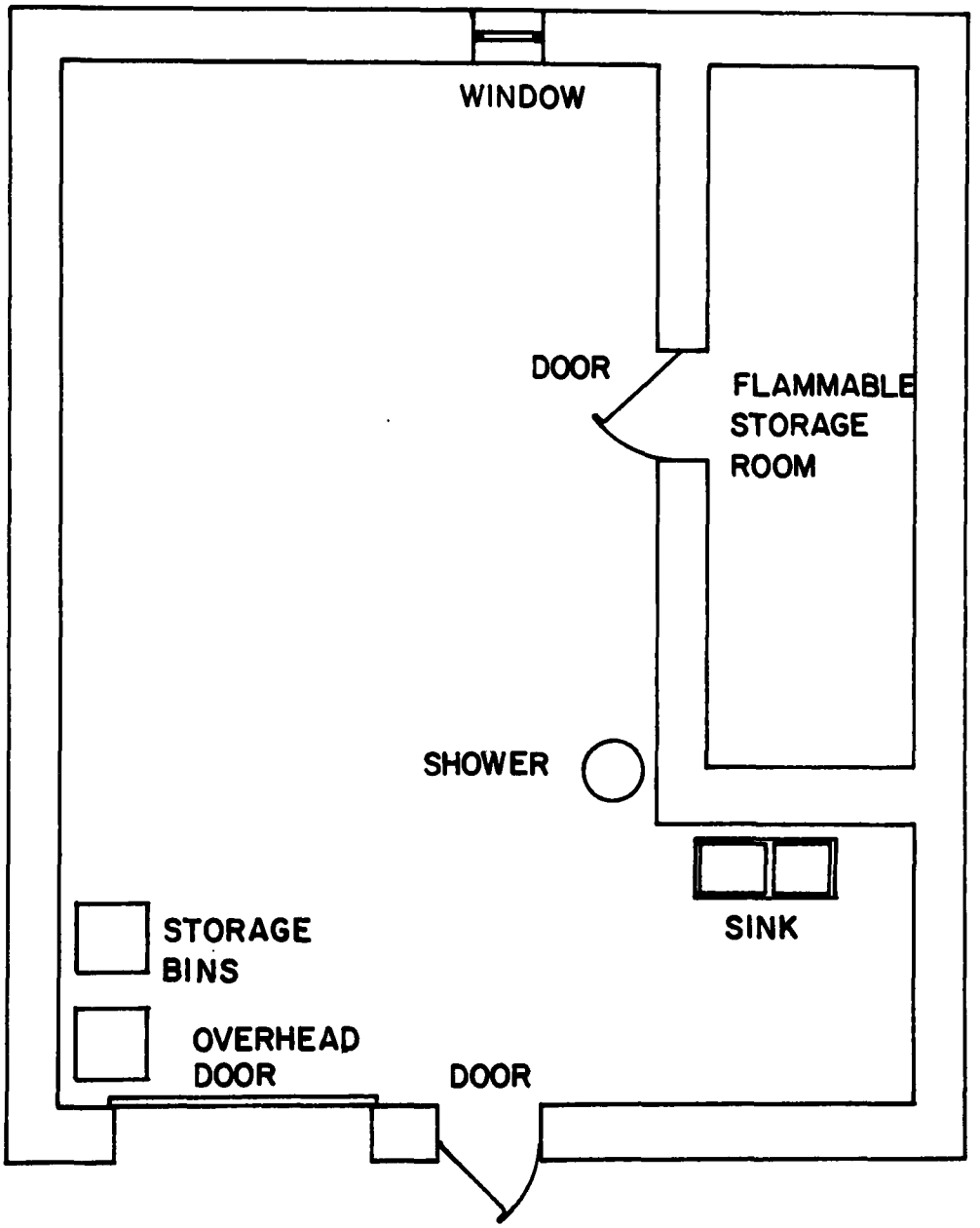
DATE 8/18/78 BY

DO NOT REMOVE THIS TAG WITHOUT AUTHORIZATION OF

09-874 Printed in U S A

Nuclear Associates Inc. ● Garle Place, N.Y

FIGURE 2



HEALTH PHYSICS

STORAGE BUILDING

AN ALTERNATIVE TO STOP THE PROLIFERATION OF LOW-LEVEL TRU WASTE CONTAINERS

J. Bruce Peterson

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Abstract

Of the many different container configurations now being utilized for interim storage of low-level TRU waste materials none is readily acceptable for direct shipment and isolation in the Waste Isolation Pilot Plant (WIPP). The proliferation of these waste containers is a direct result of efforts by the waste generators to package their unique TRU wastes into containers that meet DOE Manual Chapter 0511 twenty-year retrievability requirements under the differing environmental conditions of onsite storage. TRU wastes that continue to be packaged in non-standard containers will require repackaging to meet shipping regulations and WIPP acceptance criteria before storage in terminal isolation. Specifications for a standard container were developed. A prototype container has been built.

Background

As presently specified in DOE Manual Chapter 0511-044d(4), solid transuranic waste packaging and storage conditions shall be such that the packages can be readily retrieved in an intact, contamination-free condition for 20 yr.

The retrievable storage site for defense transuranic wastes at the Idaho National engineering Laboratory (INFL) has been accepting waste since November, 1970, and has stored this waste in an area designated the Transuranic Storage Area (TSA). The packaging and storage conditions for the waste stored at the TSA meet the requirements that the containers be readily retrievable in an intact, contamination-free condition for 20 yr.

Current DOE Division of Waste Management plans are to continue using the retrievable storage areas until the New Mexico Waste Isolation Pilot Plant (WIPP) facility attains full operational status in FY-1988. According to projections, WIPP will begin receiving transuranic wastes in FY-1983. This waste will be stored so that it can be monitored to evaluate the behavior of the waste types under the storage conditions. Projections indicate that the Pilot Plant phase will continue for 3 to 5 yr, after which, with retrieval demonstrated and experimentation successfully completed, the pilot plant will be converted to an operational repository for permanent disposal of wastes.

*Mound Facility is operated by Monsanto Research Corporation for the U. S. Department of Energy under Contract No. DE-AC04-76-DP00053.

During 1977, Monsanto Research Corporation (Mound Facility) participated in a study leading to the establishment of conceptual design criteria for defense transuranic waste packaging for Interim Storage and Terminal Isolation. A contractor questionnaire was used to gather pertinent data. Site visits were made to formulate an integrated contractor consensus; a packaging meeting was held to examine, discuss, and integrate packaging philosophies; and data collected from these activities and from Task Force meetings were consolidated to provide input to the conceptual design criteria.

Development of Design Criteria

An analysis of the information exchanges with the contractors dictate that both a drum configuration and a box geometry (preferably a modular concept) are needed. This analysis and mutual packaging consensus are based on the following contractor requirements and waste generation history:

1. Present material handling systems
2. Current and future waste processing systems
3. Present material assay systems
4. Available modes of transportation
5. 71% of the low-level TRU waste generated in 1976 was packaged in box geometry

In addition, it was found that the cost of any new packaging system is extremely important to the contractors. This cost conservation is not only based on future generation of low-level TRU wastes at the contractor sites, but also strongly influenced by known and planned decontamination and decommissioning projects where substantial increases in low-level TRU wastes are projected.

It was also concluded that the packaging acceptance criteria should be consistent for both DOE and commercial TRU wastes, since both types of generators produce essentially the same types of waste.

From these criteria, a set of conceptual design specifications was assembled for the waste container which is defined as a box or drum, including any associated liner and/or shielding material, that immediately surrounds (and is considered to be an integral, disposable part of) the waste material. These major considerations were also incorporated: public and generator safety, waste forms to be packaged, requirements for interim storage, transportation from interim storage to terminal storage, and cost effectiveness.

Structural Design

The structural design of all low-level TRU waste containers must meet the minimum requirements of a Type A package as outlined in 49 CFR 173.398b. Low-level TRU waste is any solid waste material, other than high-level waste, which is contaminated with long-lived alpha emitters to the extent that, under the provisions of DOE Manual Chapter 0511, it is not suitable for surface burial, but which exhibits sufficiently low radiation levels ($\leq$500 mrem/hr) that it is amenable to handling by "contact" methods. This minimum structural design requirement shall be required for all TRU waste packages to

assure safety to personnel during handling, loading, and unloading operations. During shipment, the Type A containers may be placed inside a reusable Type "B" overpack. The Type "B" container must meet more rigorous structural design requirements and tests than Type A containers to provide for maximum safety during shipment. Cost effective packaging and transportation of TRU waste materials will require the single use Type A packages to be relatively inexpensive but capable of meeting the requirements of contamination control from the time the containers are filled until they are backfilled inside the WIPP facility.

Design Life (Decomposition)

The design life of all TRU low-level waste containers for contamination-free retrieval shall be 10 yr minimum when stored in a noncorrosive atmosphere (pH 7-8), 60% relative humidity, and 100°F. The design life parameters may suggest a change in DOE Manual 0511 from 20-yr intact contamination-free retrievability to a 10-yr intact contamination-free retrievability concept. Life of the shipping container will start from the time the container is manufactured until backfilled in the WIPP. The 10-yr life is based upon the forecast that the WIPP will be fully operational for TRU waste containers in 1988. Life cycle of the container will include manufacturing, delivery, storage, transmittal into the WIPP, analysis, and backfilling. This life cycle should be approximately 5 yr; however, it could approach 10 yr because the backlog of interim stored wastes will be in direct competition with freshly packaged waste for isolation space in the Isolation Facility. All filled waste containers must be protected from environmental conditions that could significantly reduce the design life of the waste containers to less than 10 yr.

Materials of Construction

Materials of construction shall be based on design life and structural design requirements. Ferrous and nonferrous metals, plastics, reinforced plastics, fiberboard, corrugated fibers, wood, and concrete have been considered for container materials. All these materials can meet the requirements for hazardous materials transportation and are acceptable in the WIPP in limited quantities. Therefore, the choice of materials, or combinations thereof, can be made from the above group. However, choice will be influenced by the waste form, container design, economics, and, most important, final WIPP TRU Waste Acceptance Criteria.

Maximum Weight of Container and Contents

The weight of a single container filled to 98% capacity is limited to 25,000 lb (11,400 kg) based on a contents density of 125 lb/ft^3 (2000 kg/m^3). This design weight is based on the 25,000 lb (11,400 kg) maximum capacity of the WIPP low-level hoist cage.

Shape

The container family should be modular, having a shape which will provide maximum packing efficiency in storage. The cylindrical container has a

packing efficiency of ~0.69 and the void space will be 31 ft^3 for every 100 ft^3 of waste in terminal isolation. The cylindrical container, up to 8 ft^3, is readily mass produced and available in metal, plastic, and fiberboard. However, because of the underground location for isolation of TRU wastes, emphasis must be placed on container shape with higher packing efficiencies for the waste materials.

Dimensions

Waste container dimensions should be based on criteria to provide flexibility in mode of transportation.

Handling Appurtenances

All low-level TRU waste containers must be provided with cleats, offsets, or chimes which permit handling by fork lift.

Security Seal

The outside of each waste container must incorporate a feature such as a seal that is not readily breakable and that, while intact, will be evidence that the package has not been illicitly opened.

Cost

Current low-level waste packages which can meet the requirements of DOE Manual Chapter 0511, WIPP, and DOT Type A have costs ranging from \$3.57/ft^3 (4x4x7 ft fiberglass reinforced polyester resin box) to \$18.19/ft^3 (DOT 17H, 55-gal, stainless steel drum) for the packaging materials. Cost per cubic foot of storage volume for the standardized container family should be toward the lower end of this range to be cost effective.

Commercial Construction

Continuing efforts to develop an Acceptable TRU Waste Container System were enhanced by the results of a survey of container manufacturers completed in January 1978. The purpose of the survey was to determine if any commercially available containers were applicable to the shipment and storage of low-level TRU waste. The survey provided an overview of current packaging technology and availability. A marketing information center supplied a mailing list of 4191 National Manufacturers in seven major container categories who were currently engaged in the manufacture of containers used for packaging and shipment of various industrial commodities. The manufacturers were contacted by mail and invited to submit technical information on: container types; size, shape, internal volume; weight; closures; DOT certification (if applicable); performance data; unit and quantity cost.

One container manufacturer, Lanson Industries, submitted a proposal to work with Mound Facility to develop a standardized waste container system which could meet the above criteria.

Two container sizes were designed by Lanson. A basic 2x2x3 ft (12 ft^3) rectangular container which will overpack a 17C 55-gal drum and a 4x4x7 ft rectangular container to overpack the FRP plywood boxes were designed. These container dimensions are slightly larger than stated to provide the overpack feature. They are sized such that the small containers will fit inside the larger container with minimum void volume. Both container designs are top-loading; however, the large container is fitted with handling appurtenances which will accommodate rotation onto its side for loading large equipment or boxes with a forklift.

These containers will be manufactured from Corten A Type 4 weathering steel which has five to eight times the corrosion resistance of low carbon steel and, if painted, will hold paint two to three times longer than low carbon steel.

The 4x4x7 ft container designs included a container of 11 gauge reinforced metal and a 3/8 in. thick container designed to withstand the ½ atmosphere reduced pressure test without reinforcement. All the containers have clamped, gasketed closures with a feature for welding.

On January 10-12, 1979, the qualification testing of the top loading, 4x4x7 ft overpack prototype was completed.

The prototype passed the following tests with only minor damage and no loss of contents:

1. Reduced pressure - 7.5 psig
2. Vacuum - 7.5 psig
3. Puncture test - 13-lb pin at 40 in.
4. Drop test - 48 in. with 14,700-lb load
5. Compression test - 25,000 lb for 24 hr, also 85,000-lb structural engineering analysis on compression

The design features, quality of workmanship, testing results, and competitive costs contribute to the fact that this Mound/Lanson prototype is far superior to any of the contact handled TRU waste containers currently in use. However, for this container to be competitively priced ($700-800 each) at approximately $\$6/ft^3$, it must be manufactured in large quantities on a semi-automatic production line. The concept of central procurement would allow the procuring contractor to purchase in large lots and distribute in small quantities, as needed. The central procurement concept has been around for a while and might be one for GSA to consider.

VOLUME REDUCTION OF LOW-LEVEL RADIOACTIVE WASTE WITH A HAMMERMILL

by

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ABSTRACT

A Jacobson Model J-3 Hammermill was recently installed at the University of Rochester to process low-level radioactive wastes from hospital and research laboratories. The hammermill will handle both glass and plastic vials of all types. The waste is poured into a hopper located on the top of the seven foot high assembly. The material is gravity fed into the hammermill which is driven by a 15-horsepower motor at 3600 rpm. The waste is pounded by the rotating hammers into a metal screen perforated with one inch holes. The ground-up product is discharged from the bottom of the unit into a 55-gallon shipping drum. A volume reduction of 4:1 has been achieved on an equal mixture of glass and plastic vials.

DISUCSSION

A hammermill is an industrial type machine usually associated with farm products. A Model J-3 Hammermill was ordered from the Jacobson Machine Works, Inc., 2445 Nevada Avenue North, Minneapolis, MN 55427. The basic unit is shown in the mid portion of the picture. The hammermill was ordered with the custom made base, motor mount, and inlet hopper as shown. The total assembly weighs 1300 pounds, and costs $4,068.00 including transportation.

The hammermill was installed in an existing basement room which is used for receipt of all radioactive waste in the Medical Center. Installation was accomplished by positioning the unit in location and bolting on the inlet motor. A 6 inch vent pipe was installed from the hammermill fan to a nearby isotope hood. A drum lid was adapted to fit the 12x12 inch discharge outlet. A pallet truck is used to position an empty drum and elevate it to make a tight seal with the adapted drum lid.

After gaining some operating experience, two problems were evident. First, the inlet hopper was only 10 inches wide. A galvanized steel extension was made that was bolted in place. This extended the hopper to 22 inches wide by 36 inches long making it possible to empty the entire contents of a 13-gallon waste collection container with no spillage. A slide gate was also fabricated as a part of the hopper. The gate may be adjusted to limit the flow of material into the mill so that an overload will not occur. A controlled flow is important for processing plastic since the motor will stall or fuses will blow if too much is fed in at once.

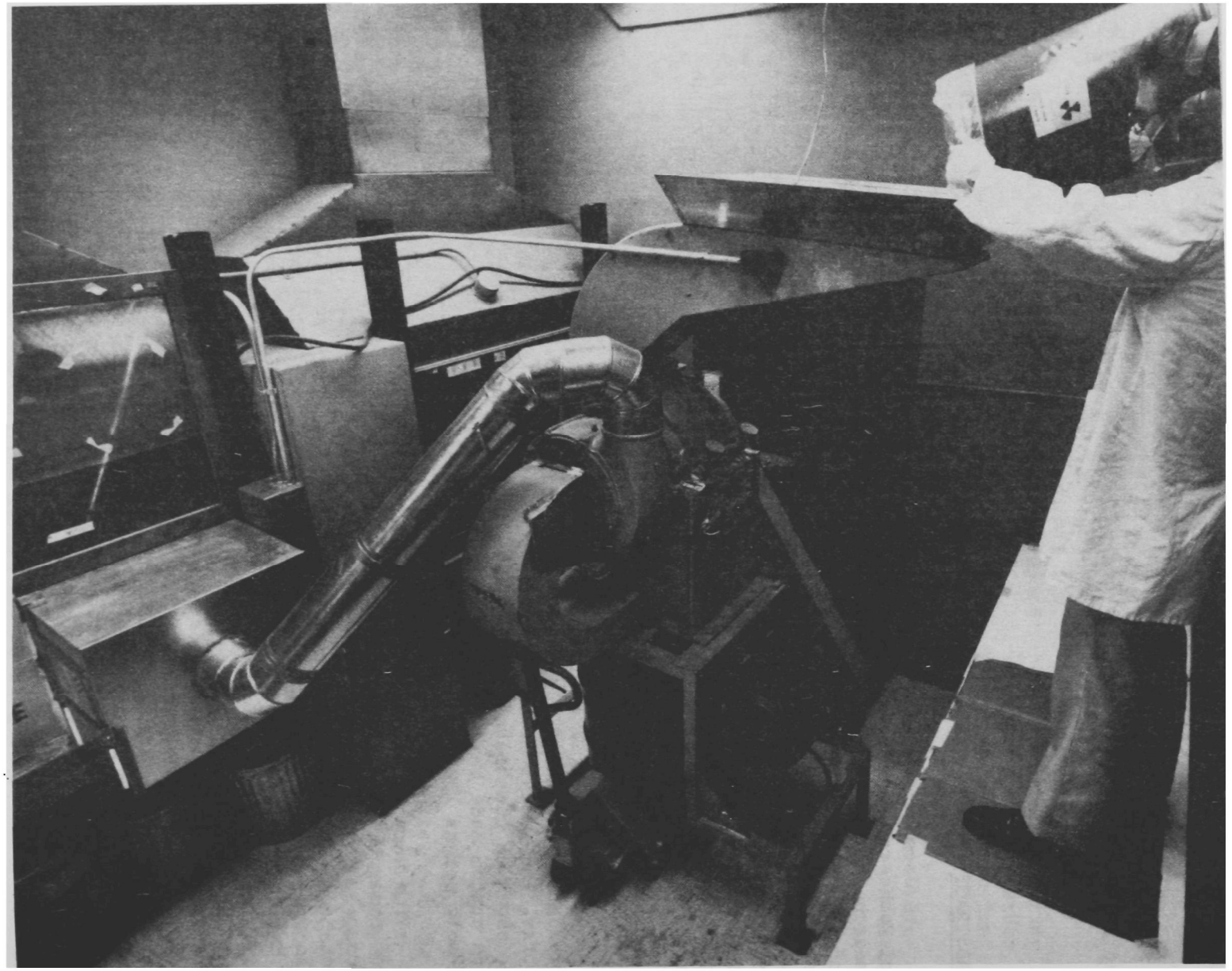

FIGURE 1. HAMMERMILL

The second problem was the dust in the discharge air from the fan. An attached fan draws air down the hopper through the mill and the waste barrel, and finally up from the barrel through the fan and out the discharge vent pipe. This airflow is necessary for cooling the hammer operation, and for pulling trash through the screen. The problem is that much of the finer dust and paper is not deposited into the drum, but is carried out the discharge vent. Several steps were taken to solve this problem. A small water supply was added to the inlet hopper so that a trickle of water would enter the mill to dampen the dust. The air flow was reduced by installing a damper in the vent pipe, and a hole was cut in the duct between the drum and the fan. This latter step allows some make-up air to enter the fan. A gate was installed so that the make-up air could be regulated.

Finally, a filter box was constructed to trap dust in the discharge. It consists of both a fine mesh screen and a furnace filter. A clean-out door is provided so that the filter can be changed and the box cleaned out with a small vacuum cleaner. This is usually necessary after filling two to three 55-gallon drums.

There are two items which require periodic inspection and replacement. They are the screen and the hammers. The screen is a curved piece of steel plate perforated with one inch holes Other hole sizes are available, but this size gave the best results for general purpose use. The screen lasted five months, and its replacement cost is only $20. Access to the screen is gained by the removal of two hand nuts and a hinged cover.

The other item requiring inspection is the hammers. There are four rows of hard faced steel hammers. These rotate around the center of the axis at 3600 rpm, and just barely clear the screen. This pounds the waste through the holes in the screen. The hammers must be rotated and reversed so that all four edges are used. Properly rotated, it is estimated that a set of hammers will last about a year depending on the amount of use the mill receives. A replacement set is about $60. It takes about an hour for two people to rotate or change a set of hammers.

In actual operation it is necessary to wear ear and eye protection, lab coat and gloves. Odor masks are also worn (3M Company) both for comfort around pungent waste, and for protection from Iodine-125 vapor. Glass vials feed themselves by gravity, but the lighter plastic vials need assistance by pushing open the hinged safety gates located in the inlet hopper. These gates prevent the flyback of material after it enters the mill.

In use, segregated quantities of glass and plastic are fed into the mill. For glass the discharge is a finely ground product. Plastic comes out in chunks no larger than one inch in diameter as determined by the screen size. An overall volume reduction of 4:1 is achieved when equal quantities of glass and plastic are processed. The resulting gross weight of a 55-gallon drum filled in this manner is about 400 pounds. For plastic alone the volume reduction is 2:1 with a gross weight of 265 pounds for a full drum. It requires about one hour for one person to fill a 55-gallon drum using the mill.

This is about the same amount of time required to handle waste using a compactor. An institutional type compactor had been used to reduce the volume of glass waste. The compactor was a Slugger-S model made by International Dynetics Corporation. It is of the screw drive type, and crushes waste directly into 55-gallon drums. A volume reduction of 3:1 was usually achieved when compacting glass alone. Plastic will not compact.

This compactor is still in use to reduce the volume of paper wastes and other solids that have large quantities of radioactivity associated with them.

In order to determine how the hammermill effected the overall waste volume, it is necessary to review the waste collections from previous years. Not including drums filled with animal carcasses, an average of 18 drums per month were filled in both 1976 and 1977. In 1978 through the end of August this had increased to an average of 20 drums per month. During those years the large compactor was in use. From September of 1978, when the hammermill was fully utilized, through January 1979, an average of only 14 drums per month were filled. This is a reduction of six drums per month. At a cost of $60/drum, this gives an annual savings of approximately $4320. The purchase cost of the mill with electrical hook-up, alterations and maintenance items is about $5000. This makes the payback period just over one year.

It is interesting to note that during the five months operating experience, six of the fourteen drums per month are still filled with compacted waste, two are filled with mini-vials and Nalge filmware containing liquids to which vermiculite has been added, and the remaining six per month are filled by the hammermill. It is expected that this ratio of 1:1 between compacted material and milled material will change since a new policy has been instituted of further segregating burnable waste into plastics only and paper/gloves only. This segregation is done at the user level and should provide additional waste that can be processed in the mill. The filtration system will also be improved so that higher activities of waste that are now compacted can be safely run through the mill without contaminating the laboratory and the environment. This should provide greater volume reduction in the overall waste picture, and increased savings.

The experience gained using this hammermill indicates that a hammermill is more effective in volume reduction than a compactor. The hammermill also lends itself to many adaptations which might include automatic feeding via a conveyor or hopper system. It could also be used to pulverize filled scintillation vials if a method were devised to extract the liquid and carry off the solid materials via an auger.

ACKNOWLEDGEMENT

The assistance of Robert J. Williams of the R. E. Williams Co., Inc. of Buffalo, NY is gratefully acknowledged. As the local representative of the Jacobson Machine Works, his operating experience and ideas were invaluable.

SESSION C

WASTE DISPOSAL OPERATIONS AND ALTERNATIVES

Session Chairperson

A. A. Moghissi

U.S. Environmental Protection Agency

REGULATORY CONSIDERATIONS FOR RADIOACTIVE WASTE DISPOSAL

Teledyne Isotopes
D.R. Fuhrman
S.A. Black
J.P. Pasinosky

Abstract

Packaging, transportation, and burial of radioactive wastes involves a complex, sometimes nebulous set of regulatory conditions, restrictions, and exceptions for proper handling. These regulations affect the shipper of radioactive wastes directly as in the case of the Department of Transportation (DOT), Nuclear Regulatory Commission (NRC), state highway permits or bridge and tunnel authorities, or indirectly by imposing restrictions on the burial site of which the shipper must be aware.

Due to the complexity of the regulations involving the various levels and types of radioactive wastes, this paper concentrates on "low level" (i.e., Type A and LSA quantities) wastes. When considering the shipment of Type A or LSA waste, the shipper must first consider the packaging, marking, labeling, etc. requirements as specified by the DOT and compatible with the respective burial site restrictions. The appropriate shipping papers must be completed certifying the packaging, marking, and labeling in accordance with DOT regulations.

In transporting radioactive wastes, the DOT imposes requirements and restrictions on the carrier. Driver's logs and examination certificates must be in the driver's possession. In addition, requirements for the safe operation of vehicles carrying hazardous materials are imposed by the DOT. In addition to the DOT regulations, certain states, localities, highway authorities or bridge and tunnel authorities impose restrictions or require permit authorization prior to transporting radioactive wastes through their jurisdiction. In many states, the respective environmental agencies have recently imposed permit requirements for transport of any hazardous materials.

The regulatory process governing radioactive waste disposal has become a maze of requirements, conditions, restrictions, and exemptions which are often redundant and ambiguous, which raises the costs to commercial waste disposal companies, carriers, and ultimately the generator.

Introduction

The question of which and what regulations are applicable must have gone through the mind of any person involved in disposing radioactive wastes. This length paper could not attempt to identify and describe all the regulatory processes for each type of waste, but it is hoped that it will serve as a guide to direct the generators of radioactive wastes to the correct sources or at least to initiate the right questions which will lead them to the correct sources.

The first part of the paper will give an overview of the regulations affecting all phases of radioactive waste disposal, i.e. generator to carrier to disposal site. In this section the applicable regulatory agencies at various stages and a list of some of the regulatory considerations each group must contend with, will be identified.

The second part will describe, in greater detail, the regulatory considerations for the disposal of low specific activity (LSA) material. The discussion will include which regulations apply to LSA material shipped by various methods, i.e. packaged and unpacked material in exclusive use vehicles, and materials shipped in non-exclusive vehicles.

Regulatory Overview

Regulations pertaining to the disposal of radioactive wastes are diverse and complex, requiring careful review of the appropriate regulations according to the particular wastes in question. Due to large potential liabilities from infractions or accidents, generators of radioactive wastes need to be cognizant of all aspects of disposal and be assured that all parties involved in the transportation and disposal of their wastes are reputable firms with extensive esperience and liability coverage.

Generally, radioactive wastes generated are disposed by two basic methods: (1) The generator acts as the shipper and coordinates all transportation and burial. In this case, the generator would perform all the necessary documentation, marking, labeling, packaging, and monitoring needed to comply with the regulations affecting the shipment. (2) The generator contracts a disposal service to act as a consultant and to remove and transfer the waste to an authorized burial site. In this case, the disposal service may supply the necessary packaging, labeling, shipping documents, and monitoring to insure that the generator will adequately comply with all the regualtions governing transportation and disposal. It is important that careful consideration is given in selecting the disposs1 service because the liability, at least in part, may ultimately reside with the generator.

In 1966, the Department of Transportation Act was passed giving the DOT regulatory responsibilities for the safe transport of radioactive materials by all modes of transportation in interstate or international shipments except for postal shipments. Postal shipments are the jurisdiction of the U.S. Postal Service, in 39CFR.

Economic considerations for transportation of radioactive materials are under the jurisdiction of the Interstate Commerce Commission (ICC) for land shipments and Civil Aeronautics Board (CAB) for air shipments through the issuance of operating authorizations and controlling tariff rates.

The Nuclear Regulatory Commission (NRC) regulates the possession and use, including transport, of byproduct, source and special nuclear materials through the licensing of these materials. The transfer of fissile material or quantities exceeding Type A to a carrier is subject to the conditions and requirements set forth in 10CFR71.

Various states have entered into agreement with the NRC which entitles them to acquire regulatory authority for the possession and use of byproduct, source and special nuclear material. The exception is for critical quantities of SNM. These states, called "Agreement States", have developed regulations for the safe use of radioactive materials including intrastate shipments.

In addition to federal regulations pertaining to the possession, use, and transportation of byproduct, source and special nuclear materials, states have

the authority to regulate radioactive materials which do not fall into one of these three categories (e.g. radium-226 and cobalt-57). These Materials are usually subject to the conditions set forth by radiological health groups in each respective state.

Recently a number of states have adopted regulations, through state environmental protection groups, to control the transportation of certain types of hazardous wastes, including non-byproduct material. These agencies have developed a manifest system to account for the waste at any stage during the transfer from the generator to the disposal site known as the "cradle to graveyard" system.

International shipments are subject to additional requirements as set forth by the DOT in 49CFR. Each shipment must be identified by its contents, mode of transport, and destination to determine which international authorities have jurisdiction over such shipments.

Regulatory Considerations of the Shipper

In considering the shipment of radioactive wastes, it is necessary to evaluate the proper regulatory requirements which are primarily subject to the type of isotopes, quantity, and form. The DOT details requirements for transportation based on the type of material in question(e.g. limited quantity, LSA, Type A, Type B, etc.) Once the material is properly identified and the quantity and the form determined, appropriate packaging can be selected. Generally, DOT specification packaging is required except in cases of limited quantities and low specific activity (LSA) materials shipped under certain conditions for transport. DOT specification packaging must meet certain test criteria to qualify for a Type A, Type B, or Large Quantity package.

In the case of Type A packaging the regulations now provide for a pure performance based DOT, Spec. 7A, Type A general package. A shipper must assure that his package will compare to tests and design specifications for the 7A package. The construction must be adequate to prevent the loss or dispersal of its contents and to maintain its radiation shielding properities if the package is subjected to "normal" conditions of transport.

Type B packaging, in addition to general packaging requirements and performance standards for "normal" conditions of transport, must also meet certain test conditions incident in an accident with a limited loss of shielding integrity and essentially no loss of containment.

Each package offered for shipment must be marked and labeled in accordance with requirements in 49CFR except for certain shipments of limited quantity or LSA material. Two labels must be placed on opposite sides of the package in accordance with the requirements in 49CFR.400. This symbol was recommended by the International Commission of Radiation Protection (ICRP) in 1956 and adopted by the American National Standards Institute (ANSI) as the standard radiation symbol.

The type of label is based on the external radiation levels at contact and three feet from the package, as defined in 49CFR173.399. The radiation level at three feet is termed the Transport Index, which is directly transcribed on "Radioactive Yellow II" and "Radioactive Yellow III" labels.

Contamination control, as described in 49CFR173.397, is required for any package offered for transportation and for the vehicle after being used for "exclusive use" shipments of radioactive materials.

Shipping documents, required with each shipment of radioactive material, must include shipping papers which identify the material and other required information. In addition, the shipper's certification is required as amended April 15, 1976 and must be instituted by July 1, 1979.

Certain "special" conditions require added precautions and packaging specifications as in the case of pyrophoric material and liquids. The specific requirements pertaining to these shipments must be carefully considered in accordance with 49CFR.

Fissile radioactive materials require additional labeling with Fissile Class I, II, or III labels. Special packaging and shipping requirements are imposed on these shipments to ensure against nuclear criticality. Packaging specifications, labeling, and special procedures are detailed in 49CFR173.396 of the DOT regulations and 10CFR71 of the NRC Regulations.

Placarding of the transport vehicle is normally the responsibility of the carrier for loads containing packages bearing a "Radioactive Yellow III" label. The exception is for "full-load" LSA shipments which require placarding of the vehicle and is under the responsibility of the shipper. For shipments with packages in excess of 200 mr/hr at contact and 10 mr/hr at three feet the shipper also has the responsibility to provide to the carrier specific instructions, included with the shipping papers, which detail the radiation levels at three feet from the package, external surface of the vehicle, two meters from the vehicle, and in any area in the vehicle normally occupied by personnel.

In addition to the DOT, the NRC imposes specific requirements in 10CFR71 which control the packaging and transportation of fissile material and quantities greater than Type A. Specific requirements must be carefully reviewed to determine the applicability of these standards. Shipments of less than Type A quantities, shipments for medical use, various quantities of fissile material, and certain Type B shipments, are exempt from the provisions of this part.

In considering the destination for burial, the shipper must be aware of requirements and conditions imposed by the burial site. Each burial site has a list of conditions which are readily available on request. Particular packaging specifications, in addition to DOT requirements, may be imposed on the shipper for certain types of materials such as bulk liquids, scintillation vials, or animal carcasses. Restrictions on total activity and transuranic elements are also a consideration. Careful determination of the type and method of packaging and proper documentation of the waste will avoid a possible rejection of the waste at the burial site.

Regulatory Considerations of the Carrier

When a disposal service is used, many of the above considerations are performed by the particular company. The disposal service acts as an intermediate agent supplying all the necessary packaging, labels, and shipping documents to meet the necessary regulatory requirements. The disposal service may transport the waste directly to the burial site or store the waste until aggregate quantities from multiple pickups total a sufficient amount to economically send to a burial site. Regardless of which method is employed, additional regulations are imposed on the disposal service or the common carrier.

During transport of the waste DOT regulations affect every aspect of the transportation. Basic requirements are imposed on a hazardous waste driver such as written examinations, medical examinations, traffic convictions and accidents, driving test, previous employment records, and an annual review of the driving record. When operating a vehicle, pre-inspection of the vehicle is required, driver's logs must be maintained, and certain restrictions for inclement weather are imposed.

Added restrictions have also been imposed on the transporters of hazardous wastes when traveling on certain roads, bridges, tunnels, or through certain communities. A definite routing plan must be determined before leaving a shipper' facility. Information regarding these restrictions may usually be obtained through bridge and tunnel authorities, the DOT, or other local agencies with authority over transportation on roadways.

If the disposal service uses intermediate storage, regulations imposed by local agencies may also be a consideration in addition to state and federal agencies. The disposal service is required to obtain a license for possession and transfer to a disposal site from the NRC and state in "non-Agreement States", or from the state in "Agreement States". Local fire or health codes may impose conditions on the storage facility such as the use of automatic fire systems.

Other regulatory agencies may be involved with certain shipments depending on the transportation mode and destination of the shipment. Agencies involved may include the International Air Transport Association (IATA) or International Atomic Energy Agency (IAEA) for international shipments, and the U.S. Postal Service, U.S. Coast Guard, Federal Aviation Administration, or Interstate Commerce Commission for domestic shipments.

Regulatory Considerations of the Disposal Site

Today, regulations governing disposal site operations are very complex and the ruling authority is not always well defined. The regulatory processes have become subject to many external forces such as environmental and public activist groups.

Generally, regulatory responsibility is maintained by the NRC for byproduct, source, and special nuclear materials for activities conducted in "non-Agreement States". In "Agreement States" regulatory responsibilities, except for special nuclear material, are subject to state control. Stringent requirements are imposed on the burial sites to minimize mishaps during burial operations and prevent releases to the environment. Details of trench configuration and parameters are well defined and incorporated in the operating licenses. The trenches must be filled and backfilled to certain specifications. To evaluate the containment, extensive environmental monitoring is required at numerous locations around the trenches and control zone perimeters. Burial sites also have restrictions on transuranic isotopes and possession limits. After a trench is completed, backfilling techniques are employed to insure proper drainage. A marker, called a tombstone, is placed on one end of the trench and is inscribed with the total activity, volume, and date of completion of burial operations.

Details Pertaining to a Shipment of LSA Material

In the following analysis, a shipment of low specific activity (LSA)

material will be evaluated according to various modes of transportation. LSA material is defined in 49CFR173.389 to include the following:

1. Uranium or thorium ores and physical or chemical concentrates of these ores;
2. Unirradiated natural or depleted uranium or unirradiated natural thorium;
3. Tritium oxide in aqueous solutions provided the concentration does not exceed 5 millicuries per milliliter;
4. Material in which the activity is essentially uniformly distributed and in which the estimated average concentration per gram of contents does not exceed:
   (i) 0.0001 millicuries of Group I radionuclides; or
   (ii) 0.005 millicuries of Group II radionuclides; or
   (iii) 0.3 millicuries of Groups III or IV radionuclides.
5. Objects of nonradioactive material externally contaminated with radioactive material, provided that the radioactive material is not readily dispersible and the surface contamination when averaged over an area of 1 square meter, does not exceed 220,000 dpm per square centimeter of Group I radionuclides or 2,200,000 dpm per square centimeter of other radionuclides.

Once the radioactive material has been determined to meet the criteria for LSA, packaging requirements are subject to the method of transportation (i.e. exclusive use vs. non-exclusive use; and closed transport vehicle vs. any vehicle).

For packaged shipments of LSA radioactive material transported in vehicles assigned as exclusive use, an exemption from specification packaging, marking, and labeling is provided if the shipment meets various additional conditions:

1. Materials must be packaged in strong, tight packages so that there will be no leakage of radioactive material under conditions normally incident to transportation.
2. Packages must not have any significant removable surface contamination as defined in 173.397.
3. External radiation levels must comply with 173.393.
4. Shipments must be loaded by the consignor and unloaded by the consignee from the transport vehicle in which originally loaded.
5. There must be no loose radioactive material in the vehicle.
6. Shipments must be braced to prevent leakage or a shift in the load under conditions normally incident to transportation.
7. Except for shipments of unconcentrated uranium or thorium ores, the transport vehicle must be placarded in accordance with 172.500.
8. The outside of each outside package must be marked "Radioactive-LSA".
9. Specific instructions for maintenance of exclusive use shipments must be provided by the shipper to the carrier.

Unpackaged shipments of LSA materials must be transported in closed transport vehicles and comply with the following conditions in addition to packaged LSA shipments.

1. Authorized materials are limited to the following:
   (i) Uranium or thorium ores and physical or chemical concentrates of those ores.
   (ii) Uranium metal or natural thorium metal, or alloys of these materials; or
   (iii) Materials of low radioactive concentrations, if the average estimated radioactive concentration does not exceed 0.001 millicuries per gram and the contribution from Group I material does

not exceed one percent of the total radioactivity.

(iv) Objects of nonradioactive material externally contaminated with radioactive material, if the radioactive material is not readily dispersible and the surface contamination, when averaged over one square meter, does not exceed 220,000 dpm per square centimeter of the other radionuclides.

2. Bulk liquids must be transported in certain specification type containers specified in 49CFR.173.

LSA shipments transported in non-exclusive use vehicles must be packaged in accordance with the requirements for Type A shipments and marked and labeled in accordance with 172.300 and 172.400.

Summary

The complexity involved with properly classifying radioactive wastes then deciding which set of regulations apply with all the conditions, requirements, exceptions and exemptions, lend itself to frequent infractions of the regulations for the average person who deals with occasional shipments. Instititutions generating large volumes of waste can afford to staff persons who will become familiar with the regulations and keep abreast of amendments. The smaller generators are subject to limited knowledge of the regulations pertaining to their shipments, or to rely on a disposal service.

Regardless of the types of wastes or quantities that are being disposed, a thorough review of the applicable regulations must be performed to ensure full compliance and a safe transfer.

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(9) 49CFR, Department of Transportation.

OPERATIONAL EXPERIENCE AT CHEM-NUCLEAR'S BARNWELL FACILITY

by

David G. Ebenhack

Chem-Nuclear Systems, Inc.

Abstract

Chem-Nuclear's low level burial ground located in Barnwell, S. C. with primary licensing by the State of South Carolina has seen a substantial increase in volume of waste and presently serves the majority of fuel and non-fuel cycle radwaste generators in the country. The waste, upon receipt, is monitored and disposed of in one of our engineered trenches. The packaging requirements, trench design and surface management minimize the possibility of release through the water pathway. The Health Physics practices and the environmental programs evaluate and monitor the personnel and population exposure pathways.

Discussion

Chem-Nuclear Systems, Inc. (CNSI) is a nuclear service orinetated corporation providing expertise, service and assistance in a number of areas for both fuel cycle and non-fuel cycle customers. One of Chem-Nuclear's major operations and the one for which we are the most widely known is the operation of a low-level radioactive waste burial site.

Chem-Nuclear's facilities are located outside of Barnwell, South Carolina adjacent to DOE's Savannah River Plant and Allied General's Reprocessing Plant. Within the approximate 300 acre tract operated by Chem-Nuclear is the burial site itself as well as transportation and maintenance facilities, support facilities for the mobile solidification, resin, technical and decontamination services offered by CNSI and administrative facilities.

Primary licensing is through South Carolina's Department of Health and Environmental Control, Bureau of Radiological Health. All radiologically controlled activities, burial packaging and burial site design, construction, monitoring and perpetual care considerations are regulated by the State. Transportation and cask compliance and regulatory activities are shared by the State, DOT and NRC. Chem-Nuclear is also licensed by the Nuclear Regulatory Commission to possess quantities of special nuclear material greater than can be authorized by the State of South Carolina under the Agreement State Program.

Burial activities have increased significantly during the last several years. The closing of the Sheffield, Illinois site left the Barnwell site the only remaining commercial low-level burial site east of Beatty, Nevada. The volume in cubic feet of waste buried each year since the initial licensing and operation in 1971, is depicted on the slide.

In 1978 we buried a total of 2,225,049 cubic feet of waste with a collective activity of 652,061 curies. The State of South Carolina has recently placed a ceiling on the annual volume of our burial operations which averages 200,000 cubic feet per month. With an average volume of 185,421 cubic feet per month in 1978, the importance of volume reduction at the point of origin becomes obvious.

Approximately 225 customers shipped waste to our site in 1978. Six of these customers, however, service a large number of additional customers, thus, bringing the total number of organizations utilizing the Barnwell site to an estimated five hundred (500). The nuclear industry which makes up the fuel cycle accounts for approximately 75% of the waste by volume. Medical, academic, industrial and research facilities, catagorized as non-fuel cycle, generate the remaining 25%. The percentage (by volume) of waste received from each state is shown here. Regionally, the largest volume comes from the Northeast with 47.8% of the total, followed by the Southeast with 32.5%, the North Central with 18.2% and the Mid-West with 1.4%. We received no waste in 1978 from the far West.

Waste shipped to the site arrives in a very large variety of package shapes, sizes, weights; cask and trailer types with radiation and contamination levels ranging from background to $\sim$ 20,000 R/hr and 500,000 dpm/100 cm^2 respectively. Special handling and offloading procedures have been developed and are continuously being improved upon to maintain and reduce personnel exposure as low as reasonably achievable. Contamination levels on the site are kept at background levels to further assure radiological control. The average monthly absorbed dose to the different catagories of site personnel shows that we have maintained exposures to about half or less of the established standards. We expect to be able to reduce this further through refinement of the handling and offloading techniques. One recent change in this area is the establishment of a set of site criteria. These criteria predominately specify special packaging and loading configuration which when fully impelmented, should reduce exposure and increase the site safety and efficiency.

All shipments, prior to acceptance on the site, are surveyed for contamination and radiation levels. The shipping papers are checked for accuracy and completeness, and the packaging is checked for compliance with our site criteria, S. C. and NRC license requirements and DOT specifications. The State makes spot inspection of shipments to monitor our performance as well as the shipper's compliance with the DOT and license requirements.

After shipments are received on site, they are directed to one of the trenches for offloading. Fuel cycle and non-fuel cycle waste is segregated into different trenches. After offloading, the truck is taken to the exit gate where it, or any other vehicle or piece of equipment which has been in the controlled area, is monitored for contamination prior to exit. If radiation or contamination levels on sole use vehicles are detected above 0.5 mr/hr or 2200 dpm/100 cm^2, then release is not permitted until sufficiently decontaminated. Unrestricted use vehicles are decontaminated to essentially background. Decontamination is accomplished by wiping, vacuuming, sandblasting and removal - disposal or a combination of these. The site Health Pnysics staff performs the site survey and decontamination activities and monitors all off-loading evolutions.

The engineered trenches consist of two basic designs - the regular and the slit trench. The slit trenches are 20 feet deep, 3 feet wide and 250 to 500 feet long. The slit trench design is utilized for high exposure rate - mostly component waste. We have two active slit trenches - one utilized when offloading horizontal casks and the other is a specially-engineered, TV-monitored pit for offloading vertical cask.

The regular trenches are nominally twenty-two feet deep with dimensions varying from 50 to 100 feet wide and 500 to 1000 feet long. There is a nominal 1% slope to the side where a french drain system is installed with a nominal slope of 0.3% end to end. Sampling points are installed every 100 feet along the french drain with a sump installed every 500 feet. Prior to initial waste disposed in a trench, 2 - 3 feet of pervious sand is backfilled in the bottom of the trench to allow any liquid to transgress to french drain system and to allow several feet of safety margin between the first detection of water and the actual waste level, allowing time to take steps to prevent the emersion of the waste.

All trench design, details and construction activities must be approved and inspected by the State.

Remote hook-up and release techniques are utilized routinely in the offloading evolutions as a means of minimizing personnel exposure. Another special technique utilized is what we call "Toner tubes". These are tube structures placed in the waste material during filling operations which are utilized at a later date for the offloading of high exposure rate drums. The restricted opening provides substantial shielding as does the slit trench design. The design shown in the picture has recently been modified to prevent the entry of rain and surface water as well as provide more positive contamination control. The filled tube is capped with a shield drum of concrete, the guide structure is removed and the opening filled and capped the same as all trench areas.

Since migration via the hydrological pathway is so important, trenches are constructed, filled and maintained in a manner which retards the entry of surface water into or through the trench. The original sand layer on top of the site is removed and the walls of the trenches are made of compacted clay. After the waste is placed in the trench, we fill the void spaces with sand backfill and use a 10,000 pound vibrating compactor to accelerate the settling process. A minimum of two (2) feet of compacted clay is then placed over the trench to act as a moisture barrier. Our license dictates a minimal addition of three (3) feet of earth cover over the clay. In actual practice, 5 to 10 feet of additional cover is provided. Then, with the addition of topsoil and fertilizer, the area is contoured and seeded to enhance the surface runoff while minimizing erosion. License requirements specifying specially approved solidification media for fuel cycle liquids and specially designated packaging requirements for non-fuel cycle liquids further reduces the potential for migration from the trenches.

There are two major inhibitors to more efficient operation. Since operations on the site are almost exclusively conducted outdoors, the weather can have a significant impact. For example, last week we had a day of freezing cold weather, coupled with an ice storm which shut down operations by noon. The next day, it had warmed up some, the power was on and the ice was gone but what remained was at least 12 inches of very slippery, gooee red clay. The red clay has fair ion-exchange properties - a good location for a burial ground, but really hard to work in when muddy. Just plain rain muddies up the site and lightning storms shut down the cranes. During the summer, the heat, insects and dust also leave something to be desired.

The other major inhibitor to efficient orderly operations at the site is, unfortunately, the customers. There are an average of 480 shipments arriving at the site each month. During the last four weeks, there were sixty-four waste shipment discrepancies noted: Ten against the site criteria, sixteen related to the paperwork, eleven dealing with equipment, eighteen related to improper packaging and nine for contamination levels. Waste not solidified or packaged properly, shipping papers which are either not legible or incomplete, and contamination problems all hold up and add costs to the disposal of your waste.

The environmental monitoring program consists of soil and vegetation samples, environmental TLD's, air samples and well water samples. The major route of migration off site would be via the water migration path. For this reason, there is an extensive well monitoring program. In addition to the sumps and sample points at the bottom of the trenches, there are also monitoring wells scattered throughout the active burial area, around the perimeter of the site and off site. The initial monitoring well at a specific location extends to

the water table and is core sampled while drilling. The core samples provide not only analytical data but also shows the depth of any additional saturated sand layers that may run through the clay bed. Monitor wells are installed at the same location for each saturated layer. All wells are sealed, grouted and capped to preclude any entry of contamination from the surface. Prior to each sampling, the wells are pumped down and allowed to recharge to provide a more accurate indication of the present water condition. To date, only very limited migration has been detected. Very close to one trench, samples have shown organics and tritium contamination only.

The burial area (approximately 235 acres) is owned by the State of South Carolina and leased to Chem-Nuclear for operation. The State's extensive environmental monitoring program in the area will be maintained into the future through the perpetual care fund. This fund was established at the time of initial licensing, is controlled by the State and is based upon the volume of waste disposed.

All areas - trench and well designs and locations; solidification and packaging methods, environmental monitoring and modeling, the perpetual care fund, offloading, safety and health physics procedures, special techniques for volume reduction and for increased efficiency in trench space utilization are in a continuous state of review and improvement by both CNSI and State personnel.

I would like to extend an invitation to you to visit our facilities in Barnwell if you are ever in the area. If anyone has any questions, I will be glad to try to answer them.

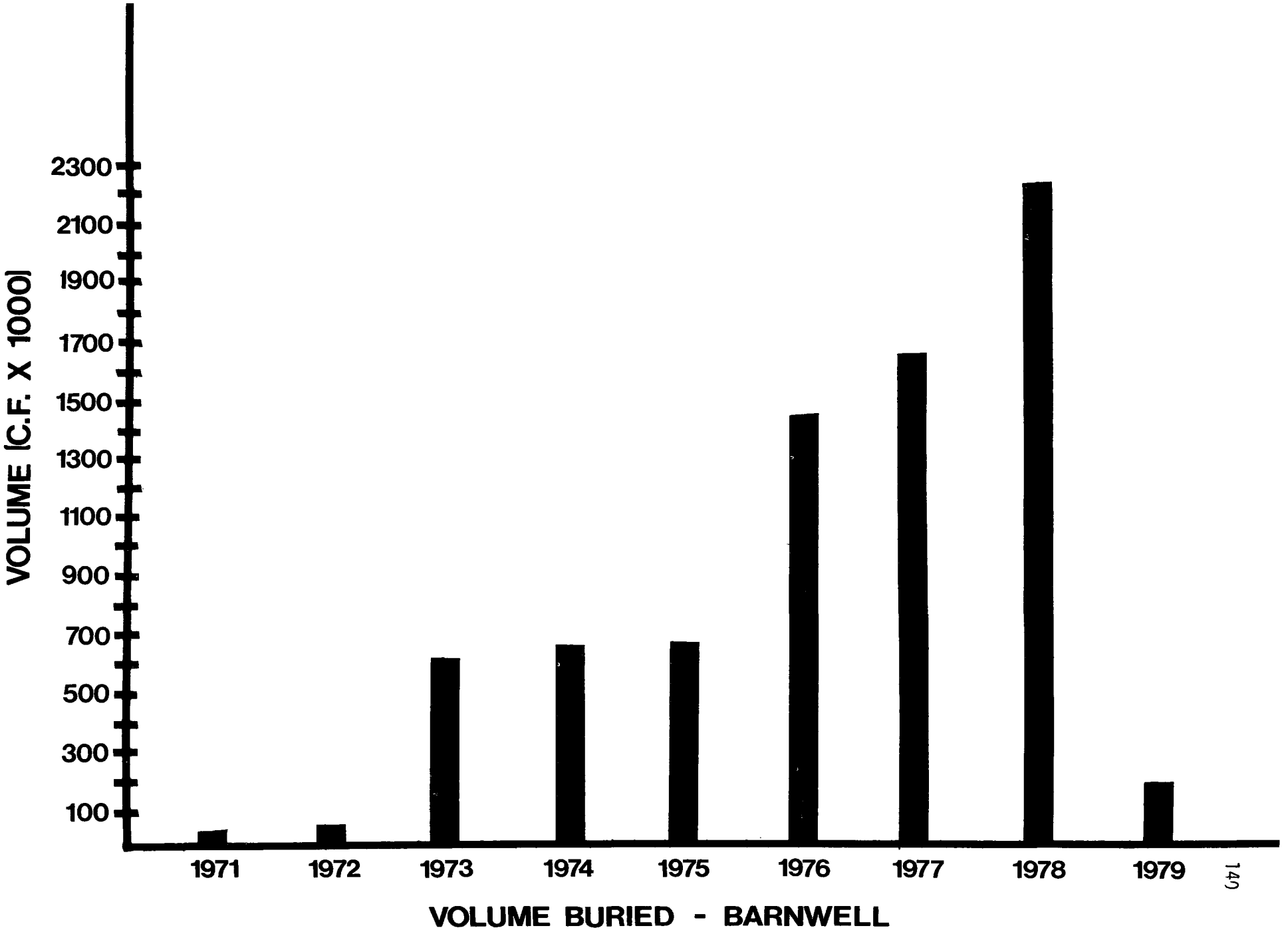
Based on 10.5 month, 1978:

Average Exposure

	No.	Avg. (mrem/month)
H.P. Techs.	4	198
Office, Mgm., Supv. (Includes Dispatcher, Janitor, Warehouseman)	9	26
Offloaders	15	174
Truck Drivers	8	21
Equipment Operators	6	176
Maintenance Personnel (Mechanics, Welder etc)	6	6

CUBIC FEET/BY STATE/6 MONTHS ---- JULY, 1978 THRU DECEMBER, 1978

STATE	VOLUME	%
Alabama	57,121.04	5.2
Arkansas	1,830.12	.2
Arizona	9.0	.0
Colorado	200.95	.0
California	516.8	.0
Connecticut	34,829.16	3.2
Delaware	249.0	.0
Florida	42,932.75	3.9
Georgia	14,775.25	1.3
Hawaii	2.0	.0
Illinois	90,020.11	8.2
Indiana	4.0	.0
Iowa	21,214.75	1.9
Kentucky	3,990.56	.4
Louisiana	732.3	.1
Maryland	38,619.02	3.5
Massachusetts	80,375.25	7.3
Maine	18,876.24	1.7
Michigan	31,119.28	2.8
Missouri	2,761.57	.3
Minnesota	7,681.57	.7
Mississippi	805.5	.1
Montana	21.0	.0
New Jersey	60,585.72	5.5
New York	165,504.6	15.0
Nevada	4.0	.0
North Carolina	75,462.28	6.8
Nebraska	9,738.8	.9
North Dakota	36.9	.0
New Hampshire	3,307.7	.3
Oregon	82.5	.0
Ohio	42,739.61	3.9
Oklahoma	32.38	.0
Pennsylvania	115,947.06	10.5
Puerto Rico	448.35	.0
Rhode Island	1,314.9	.1
South Carolina	79,517.57	7.2
South Dakota	.6	.0
Tennessee	46,862.47	4.3
Texas	5,469.63	.5
Utah	11.9	.0
Virginia	33,431.76	3.0
Vermont	5,683.6	.5
Washington	88.2	.0
Washington, D.C.	1,947.89	.2
West Virginia	1,576.13	.1
Utah	40.0	.0
Wisconsin	3,381.98	.3
TOTAL	1,101,903.75	99.9%



VOLUME BURIED - BARNWELL

MANAGEMENT AND SURVEILLANCE OF A UNIVERSITY RADIOACTIVE WASTE BURIAL SITE

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Abstract

A radioactive waste burial site is operated at the University of Missouri under the conditions of 10 CFR 20.304. Over 90% of the radioactive wastes generated by the laboratories and clinics, exclusive of the Research Reactor Facility, are economically disposed at this site. During the seven year operation about 200 cubic meters of low-level wastes containing about 1.5×10^{11} becquerels have been buried in 3.6 meter deep and 0.6 meter wide trenches. A radiation surveillance program confirms that radiation levels in the vicinity of the burial site are well within acceptable limits.

Discussion

All licensees of the Nuclear Regulatory Commission are permitted to dispose of small activities of radioactive wastes by local land burial. General provisions contained in Section 20.304 of Title 10 of the Code of Federal Regulations (10 CFR 20.304) stipulate allowable burial conditions such as nuclide activities, burial depths, and burial frequencies for the local land burials. Recently it has been announced that the provisions for generally permitted land disposals are under discussion and may be modified or withdrawn. Hopefully any changes in the regulations will not completely eliminate the opportunity for institutions to operate a local radioactive waste site for the efficient disposal of low-level solid wastes.

The University of Missouri has operated a radioactive burial site within the conditions of 10 CFR 20.304 for the past seven years. This designated burial site is a nine-tenth acre fenced plot on a University research farm located about five miles from the main portion of the Columbia Campus. A metal building at the site is used for temporary waste storage and for processing radioactive wastes scheduled for transfer to commercial firms for off-site disposal. This building is also used to a limited extent for the temporary storage of radioactive wastes and there is a freezer in the building for storing contaminated animal carcasses that are to be buried.

The soil at the site consists of a water permeable clay layer to a depth of about three meters. Below the surface soil layer is about one meter of water impermeable clay and then about a one-half meter layer of weathered limestone and clay over the limestone bedrock. Burial trenches which are about four meters deep may penetrate into the impermeable clay region but will not break through it. Surface water drains to the north across the site into a natural drainage ravine near the farm boundary about 200 meters from the north end of the burial site.

Most of the radioactive wastes generated at the University research and clinical laboratories consist of slightly contaminated paper trash, plastic vials, animal bedding, and animal carcasses. This high volume of low activity waste is well suited for shallow land burial disposal. The wastes are collected from the campus locations and accumulated in the waste storage areas on campus

or in the storage building at the waste site. Burials are scheduled with respect to weather conditions and the amounts of accumulated wastes.

Burial trenches are dug to a depth of about four meters by a University operated backhoe. The width and length of an individual burial trench is dependent upon the volume and types of wastes to be buried. Usually the trenches are about sixty centimeters wide and four meters long; however, trenches double this normal width are convenient for burial of large animal carcasses. The location and size of a particular burial trench is recorded on the map of the burial site as shown in figure 1 and referenced by a number code giving the calendar year and burial number for that year, such that the number 78-6 would indicate the sixth burial at the site in 1978.

Each item placed in the burial trench, whether a bag of laboratory trash, animal carcasses, or container of animal bedding, is identified according to the contaminating nuclide and activity. Records for each burial indicate the activities of all radioisotopes, total volume of uncompacted wastes, numbers of items buried and the fraction of an allowable burial in reference to the allowable activity limits defined in 10 CFR 20.304. Other waste records can be used, if required, to trace an item buried to its source laboratory. Following burial the wastes are covered with at least 1.2 meter of dirt and the burial located and coded on the site map.

There have been 40 burials at the waste site as of the end of 1978. These burials represent over 95% of the solid radioactive wastes generated at the Columbia Campus over the past seven years. The present waste site should accommodate the University for the next fifteen years; however, the area can be expanded if needed.

A total of 1.4×10^{11} Bq (3.87×10^{3} mCi) have been disposed by land burial in the 40 burials over the past seven years. The uncompacted volume of the disposed wastes is estimated to be 199 m^3 (7027 ft^3). Each burial is distinctively different but an average burial would be 3.58×10^{9} Bq (99.9 mCi) with a volume of 4.96 m^3 (175 ft^3) and would represent 47.4% of an allowable burial as defined in 10 CFR 20.304. A summary of the annual burial totals is given in table 1.

The volume of waste buried each year has been fairly consistant ranging from about 11 m^3 to 39 m^3 for the calendar year as shown in figure 2. However, there has been a decrease in the activities from about 4.8×10^{10} Bq being disposed in 1973 to only 4×10^{9} to 9×10^{9} Bq being disposed in each of the past three years as shown in figure 3. One of the reasons for the decrease in disposed activity is that labeling procedures requiring relatively large amounts of tritium are not being performed by campus users to the extent of several years ago.

The residual activity of previously buried wastes is continuously being reduced by radioactive decay. A computer program calculates this decay reduction and indicates the radioisotope inventories in the site. Residual activities have reached a plateau over the past two years due to decreased levels of activities per burial and the decay of the previously disposed radionuclides. The variation in residual activity over the past seven years is indicated by figure 4. At the end of 1978 the total activity of the buried wastes was calculated as being 1.11×10^{11} Bq (3.0 Ci). Of the total activity

tritium accounted for 97.4% and carbon-14 accounted for 2.49%. Other radionuclides which were minor contributors to the residual activity include scandium-46 at 0.04%, iodine-125 at 0.03%, calcium-45 at 0.02% and strontium-85 at 0.02%. Twenty-three other radionuclides are identified by the disposal records as being represented at very low activities in the burial site. The total activities of the different radioisotopes in the burial site at the end of 1978 are listed in table 2.

Radiation surveys and environmental samplings have been conducted in the waste site area to confirm that radiation levels are maintained within acceptable limits. After the backfill of each burial, the surface exposure rate is measured with a portable survey meter. There was only one measurement above normal background recorded during a post burial survey. This elevated reading was due to some items that had not been properly covered by the backfill operation. The materials were collected and were properly disposed in a later burial.

Surveys are also taken monthly at fifteen identified locations around the site boundary and in the waste storage building. Elevated exposure rates in and around the waste storage building are produced by radioactive wastes stored in the building. These wastes are primarily from the Research Reactor Facility and are being stored and processed before transfer to a commercial waste disposal vendor. Exposure rates within the building range up to 40 mR/h while the exposure rates at the site boundary nearest to the building have been as high as 0.15 mR/h when the building is full of stored wastes. Other survey points beyond the influence of the radioactive material in the storage building are at normal background levels of less than 0.05 mR/h. Thermoluminescent dosimeters positioned at the monitoring locations for extended time periods confirm the exposure rate measurements.

Soil and grass samples were obtained in 1978 at nine of the monitoring locations at the site and analyzed for radioactivity. No activity was observed above 1 Bq/g of gamma activity or above 1 Bq/g of beta activity on any of the samples. Soil samples from depths ranging to three meters were also taken at the middle of the north boundary of the waste site. These soil samples, taken in 1977, were also below 1 Bq/g of beta activity. The environmental sample measurements confirm that the soil and vegetation have not been contaminated with radionuclides from the burial site.

Water samples taken from the surface of the waste site and in the drainage area below the site showed neither gamma nor beta activity above background levels nor any tritium activity above the 400 Bq/ℓ detection level. Ground water samples from three meter deep sample holes at the north edge of the site did show slight positive tritium activity in 1977 when heavy rains probably flushed the tritium from the burial pit near the sample point. Concentrations of 10^4 Bq/ℓ of tritium were observed in the ground water from the site at that time.

Operating costs of the waste site are minimal. The major expense is the digging and backfilling charges from the University service furnishing the backhoe equipment. These charges average about $40 per burial which are small in comparison to the approximate $800 expense of having the wastes removed by a commercial disposal firm. Personnel time and efforts for a local

burial would be approximately the same as preparing the waste for shipment. This time is estimated to be about 1.5 man days per burial. Packaging expenses would be greater for a commercial shipment because of the cost of the containers. Containment or packaging costs for local burial are minimal because certified shipping containers are not required.

The local shallow land burial site has proven to be an effective means for disposal of almost all of the low-level solid wastes generated by the University campus. It has been demonstrated that a low-level waste disposal site properly maintained and operated within the conditions of 10 CFR 20.304 is a safe, efficient, and economical disposal method for large quantities of low-level radioactive wastes. It is recommended that continued use of this type of burial sites be allowed for institutions willing to accept the responsibility for proper custodial maintenance.

TABLE 1

Summary of Radioactive Burials

University of Missouri

Year (Calendar)	Burials (Number)	Volume (Cubic Meters)	Activity (Becquerels)
1972	1	11.5	1.23×10^{9}
1973	6	30.6	4.84×10^{10}
1974	3	13.4	3.37×10^{10}
1975	6	31.2	4.08×10^{10}
1976	9	35.2	4.76×10^{9}
1977	8	39.1	5.65×10^{9}
1978	7	37.5	8.84×10^{9}
Totals	40	198.5	1.43×10^{11}

TABLE 2

Residual Activities at Burial Site
(December 29, 1978)

Radioisotope	Activity (becquerels)
Hydrogen-3	1.08×10^{11}
Carbon-14	2.77×10^{9}
Sodium-22	4.51×10^{5}
Phosphorus-32	1.01×10^{7}
Sulphur-35	9.51×10^{6}
Chlorine-36	6.99×10^{6}
Calcium-45	2.64×10^{7}
Scandium-46	4.96×10^{7}
Chromium-51	7.20×10^{6}
Magnesium-54	1.33×10^{4}
Iron-55	5.40×10^{5}
Iron-59	$<1.0\times10^{2}$
Cobalt-57	3.96×10^{2}
Cobalt-60	2.82×10^{6}
Zinc-65	4.44×10^{5}
Selenium-75	2.96×10^{5}
Arsenic-76	$<1.0\times10^{2}$
Strontium-85	2.36×10^{7}
Rubidium-86	$<1.0\times10^{2}$
Molybdenum-99	$<1.0\times10^{2}$
Technetium-99m	$<1.0\times10^{2}$
Cadmium-109	2.02×10^{5}
Iodine-125	3.06×10^{7}
Iodine-131	2.07×10^{6}
Cerium-144	7.40×10^{6}
Terbium-160	7.10×10^{5}
Mercury-203	$<1.0\times10^{2}$
Lead-210	$<1.0\times10^{2}$
Unknown Beta Emitters	1.11×10^{5}
Totals	1.11×10^{11}

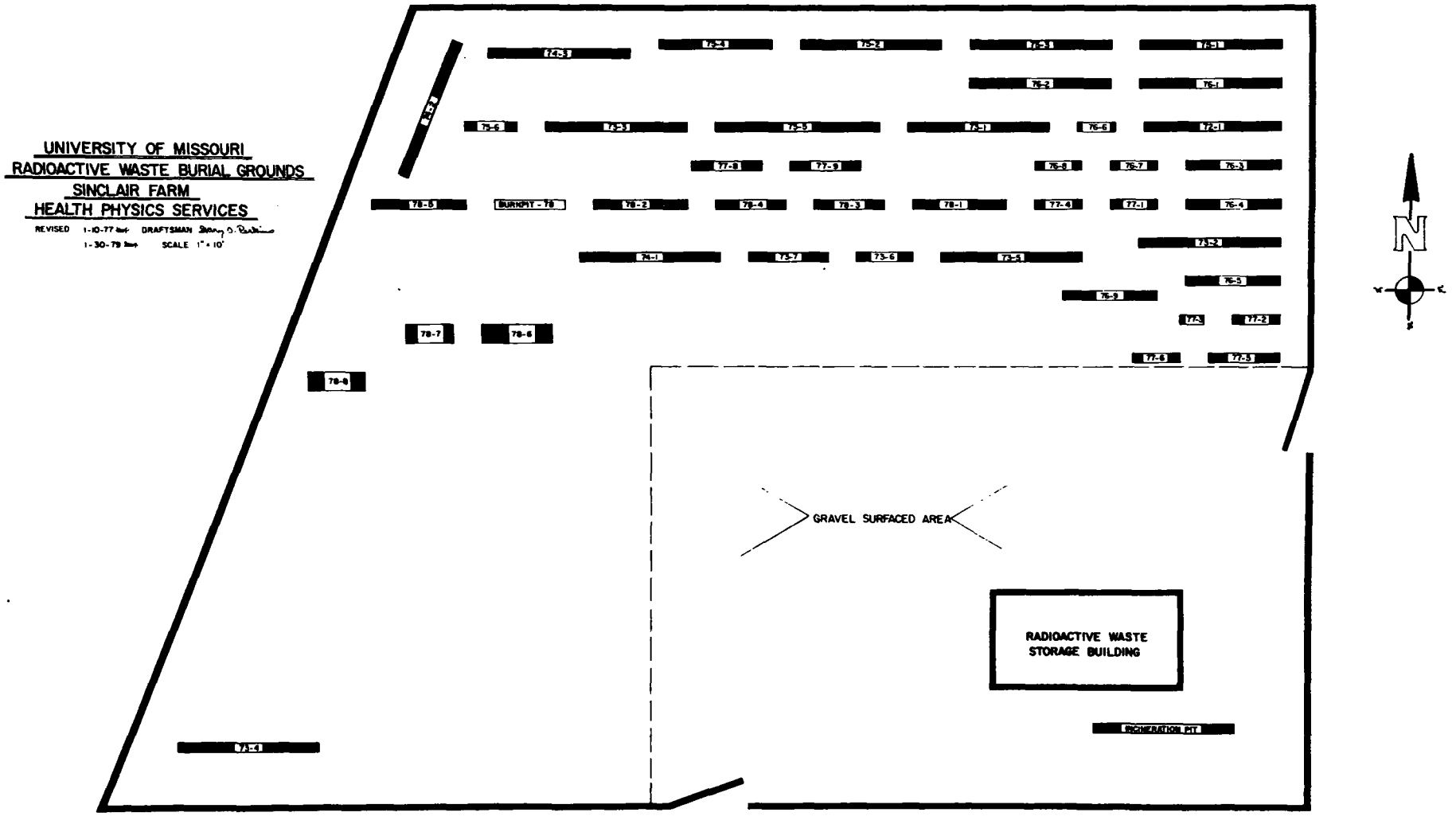

UNIVERSITY OF MISSOURI
RADIOACTIVE WASTE BURIAL GROUNDS
SINCLAIR FARM
HEALTH PHYSICS SERVICES
REVISED 1-10-77 DRAFTSMAN
1-30-79 SCALE 1" = 10'
GRAVEL SURFACED AREA
RADIOACTIVE WASTE
STORAGE BUILDING
INCINERATION PIT
BURNPIT - 78
N

FIGURE 1

FIGURE 2

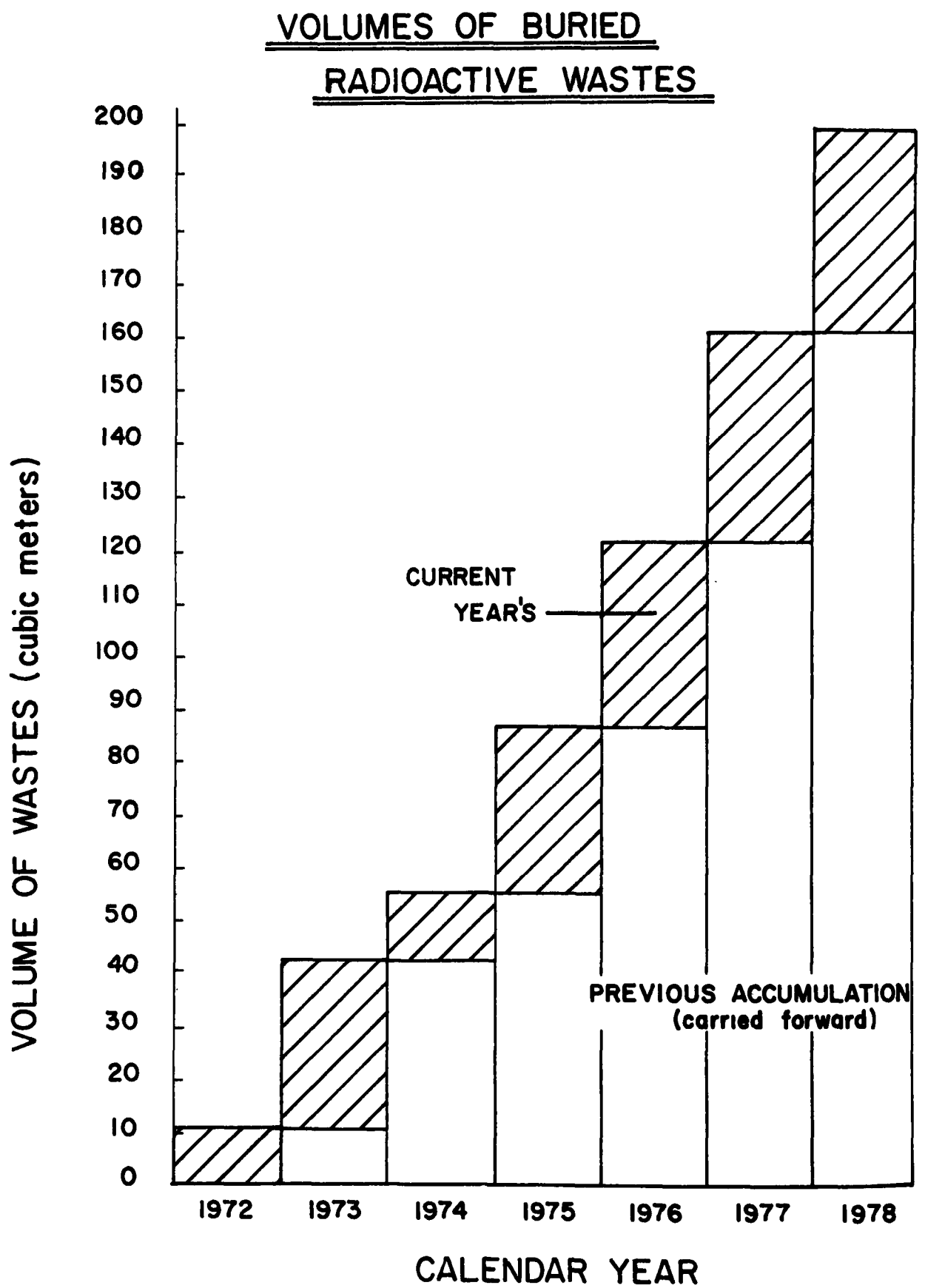

VOLUMES OF BURIED
RADIOACTIVE WASTES
VOLUME OF WASTES (cubic meters)
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
CURRENT
YEAR'S
PREVIOUS ACCUMULATION
(carried forward)
1972
1973
1974
1975
1976
1977
1978
CALENDAR YEAR

FIGURE 3

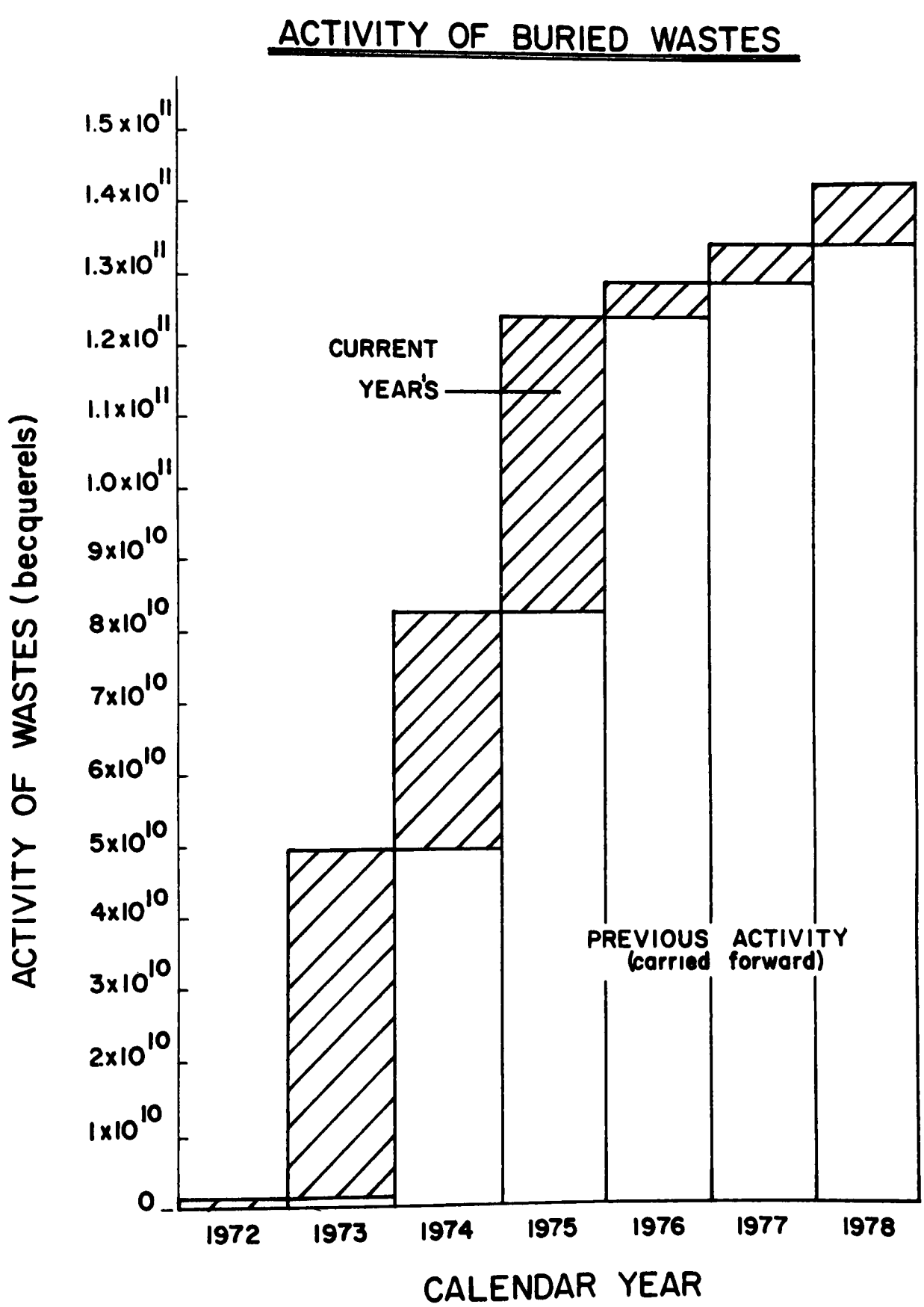
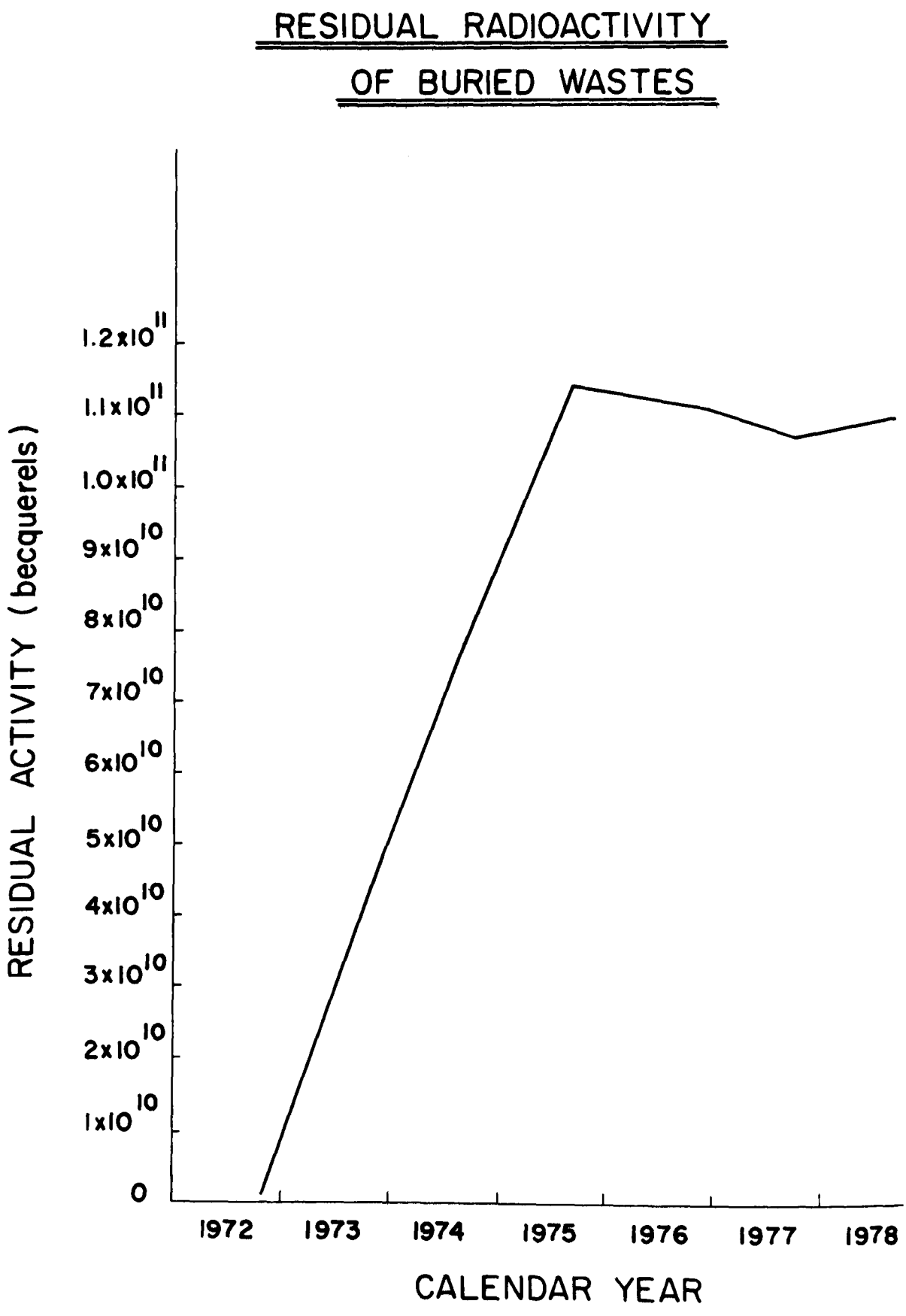

ACTIVITY OF BURIED WASTES
ACTIVITY OF WASTES (becquerels)
1.5 x 10^11
1.4x10^11
1.3x10^11
1.2x10^11
1.1x10^11
1.0x10^11
9x10^10
8x10^10
7x10^10
6x10^10
5x10^10
4x10^10
3x10^10
2x10^10
1x10^10
0
CURRENT YEAR'S
PREVIOUS ACTIVITY (carried forward)
1972
1973
1974
1975
1976
1977
1978
CALENDAR YEAR

FIGURE 4


RESIDUAL RADIOACTIVITY
OF BURIED WASTES
RESIDUAL ACTIVITY (becquerels)
1.2x10^11
1.1x10^11
1.0x10^11
9x10^10
8x10^10
7x10^10
6x10^10
5x10^10
4x10^10
3x10^10
2x10^10
1x10^10
0
1972
1973
1974
1975
1976
1977
1978
CALENDAR YEAR

A PRELIMINARY IMPACT ASSESSMENT OF INSTITUTIONAL RADIOACTIVE WASTE DISPOSAL

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Abstract

A significant fraction of the low level radioactive wastes which are buried in the commercial, shallow land burial sites in the U.S. originate from non-fuel cycle sources. The primary radwaste producers in this category include large medical and academic institutions. This paper considers, in a preliminary manner, some of the impacts of disposing of institutional radwastes via the same methods and systems as are used to dispose of fuel cycle radwastes.

Nuclide content and activity concentrations of institutional and reactor radwastes differ greatly. The varied physical and chemical forms of institutional radwastes may not be compatible for burial with reactor radwastes. Animal carcasses and other biological materials may adversely affect containment and transport parameters in the burial trenches. The time and expense involved in the commercial burial of institutional radwastes may not be commensurate with the actual radiological content of the materials. Finally, this method of disposal may not represent an optimum use of limited burial space.

Introduction

In 1975, 12% of the low level radioactive wastes which were buried at shallow land burial sites in the U.S. were institutional radwastes (An78). The total percentage of low level radwastes which were buried from all non-fuel cycle sources was 39% (Ho79). This paper will discuss the impacts of institutional radwastes only, but many of the points that are made could equally apply to all non-fuel cycle radwastes (but with 3.25 times the quantitative significance).

Method of Identifying Impacts

Impacts are the result or consequence of utilizing a material item within a particular process (e.g. driving an automobile, mining coal, or generating electricity with a nuclear power plant). In this case, the impacts that are considered are the result of utilizing the material, institutional radwaste, within the process of disposal by shallow land burial at commercial sites. It is convenient to break down the process and the material into distinct elements, and then to identify specific impacts within a matrix of these elements. This is shown in Figure 1.

The disposal process is broken down into three elemnts: Collection, transport and burial. Collection includes picking up the radwastes at a use point within the institution (e.g. university lab), processing and packaging the material at a central collection point (e.g. health physics lab), and storing the material for shipment. the extent of these processes varies according to the type of institution and scope of the radionuclide use program.

Transport includes either direct shipment of the radwastes from the institution to the burial site, or, shipment to an intermediate facility (i.e. commercial radwaste disposal company warehouse) for consolidation into a larger shipment to the burial site. Burial includes offloading the material into the burial trench, backfilling over the material, and the residence of the material at the site.

The institutional radwaste material in defined with three parameters: volume, form and radioactivity. The volume refers to the volume of the disposal package, and not necessarily the volume of the contents, unless otherwise specified.

Form refers to the physical or chemical form of the waste. This includes scintillation vials, packaged as glass or plastic vials containing organic scintillation fluids; biological materials, including animal carcasses, excreta, tissue cultures, blood samples, etc.; solid wastes, including paper, gloves, labware, etc.; and solidified or adsorbed liquids, which may include aqueous solutions or organic fluids. Radioactivity refers to the radionuclide and activity content of the waste.

Once the specific impacts have been identified, they are grouped together into more general impact categories for a discussion of their significance and possible alternatives. A schematic example is shown in Figure 2.

Impacts of Institutional Radwaste Disposal

In Figure 3, the specific impacts have been grouped together into three general impact categories: A discussion will be made of each category. Where possible, quantitative data will be cited or estimated for specific impacts in order to discuss their significance.

Resources impacts include direct and indirect costs, materials consumed in the disposal process, and limited or non-renewable resources used in the disposal process.

The acquisition and establishment of facilities and equipment for processing, packaging and storing radwastes pose unique problems for institutions. These items are considered as impacts because the largest part of their expense is met through public funds (either in the form of overhead in research money allocations, or as allocated budget items from state or federal funds) or in medical fees (in the case of hospitals). These funding resources are becoming limited through inflation and leglislated restrictions upon taxes, state and federal budgets and medical costs.

Comprehensive data on moneys spent on facilities which are specifically intended for radwaste management within institutions is not available. However, costs for radwaste processing facilities built in 1975-77 at several major universities ranged from $95,000 to $165,000 (An77).

A lack of adequate facilities for compacting solid radioactive wastes, processing liquids, and consolidating radwastes into large shipments, particularly at smaller institutions, yields secondary impacts as the result of increased volume, more unprocessed liquids or organics at the burial site, and increased transport costs (which are typically a function of the quantity of

Personnel time expended in the collection phase of radwaste disposal has impacts similar to those of facilities and equipment. Funds are derived from the same sources. A lack of adequate staff results in the same secondary impacts discussed above. It should be noted that often, the unavoidable demands of radwaste disposal, in terms of time and money, often restrict other, "less necessary", radiation safety functions. "Less necessary" in this case is defined by the respective radiation safety officer.

During 1977 interviews with 15 institutional radiation safety officers (An77), data was obtained regarding the amount of personnel time spent on the collection phase of radwaste disposal (at the institution). Dividing the time spent by the total volume shipped by that respective institution in the preceeding twelve months yielded an average time allotment of 0.6 person-hours/ft^3 of waste shipped. Because all of these programs were at major institutions, an extrapolation to the time expended by the entire institutional population cannot appropriately be made.

Containers used in institutional radioactive waste disposal are typically 55 gallon steel drums (An78). Thirty gallon steel drums, fiberboard or wood boxes, or steel pails are also used. In order to conceptualize the number of containers that are consumed in the institutional radwaste disposal process, it is convenient to translate the total volume of radioactive wastes shipped for burial in 1977 (274,433 ft^3 - Be79) into an equivalent number of 55 gallon drums. This yields 37,388 drums, or, a quantity which if placed in a single line, would extend for more than fourteen miles.

The cost of disposal containers varies according to the type of container, whether it is new or reconditioned, and the quantity purchased at one time (which is often a function of storage facilities). Prices for 55 gallon drums ranges from $15 to $25 each. Using the drum equivalency figure, above, yields a container cost of $560,000 to $935,000 for 1977.

Shipping radioactive wastes consumes fuel, both for a local radwaste pickup and for a cross country trip to a burial site. Because there are now only three sites available, some institutions ship radwastes as far as 1,500 miles. Shipments may be made as part of a combined load (with non-radioactive materials) via a local shipper, or may be made in truckload quantities (76 to 154 55 gallon drums) by the institution or a local radwaste disposal company. Quantification of the amount of fuel expended in this phase of disposal cannot be made with the available data.

The final resources impact to be considered is use of burial space. In view of increased public opposition to the establishment of burial sites within their locality (e.g. as in New Mexico), and the increased time and capital requirements for establishing a site, burial space must be considered a limited resource. Space which is utilized for institutional radwastes cannot be used for the disposal of other radwastes (e.g. reactor wastes).

In view of the low activity concentrations of institutional radwastes, consisting primarily of ^{3}H, ^{14}C and ^{125}I, it is not clear that the disposal of these wastes in a burial site represents an optimum use of [illegible] activity concentration for institutional

radwastes shipped for burial in 1975 was less than 0.03 Ci/m^3 (An78). By comparison, the average activity concentration of reactor low level radwastes shipped for burial in the same period was 1.49 Ci/m^3, consisting primarily of mixed activation and fission products (Ep77).

The impact which is the least quantifiable at this point, but may be the most significant, is that of environment. Although much research is underway to assess qualitatively and quantitatively the effects of organic solvents, liquids, and biological materials upon radwaste containment and transport parameters in the burial trench (e.g. Co-- and Ep--), the full significance of introducing these materials into the sites may not be known for some time.

What is known is that institutional radwastes are a source of liquids, organics, and biological materials in the trenches. In 1977, approximately 486,134 liters of scintillation fluids, in the form of scintillation vials, 31,834 ft^3 of adsorbed and solidified liquids, and 27,718 ft^3 of biological wastes were shipped to the burial sites (Be79). It should be noted that the burial facility at Barnwell, SC, is currently burying non-fuel cycle radwaste in seperate trenches from fuel cycle radwaste (Oa79).

Health and safety impacts include personnel exposures to radiation, chemicals and bio-materials and the potential for accidents, including fire, expolosion or traffic accidents. Radiation exposure may be the least significant of these. Typical annual exposures of personnel handling radwastes at 15 institutions were less than 100 mRem/yr (An77). Although such a small sample cannot be extrapolated with great confidence upon the total institutional population, such small exposures do seem consistent with the low activity concentrations of beta and low energy gamma emitters in the waste.

The noxious and toxic properties of handling toluene are well documented (He73). A lack of adequate facilities to provide personnel protection from organic solvent vapors or noxious fumes from biological wastes often results minimal handling and processing of these materials at the institution. This is reflected in drums of unabsorbed/unsolidified liquids which occasionally reach the burial site (An79 and Oa79).

Fire, explosion and traffic accidents also occur in the disposal of institutional radioactive wastes. Two examples from first-hand experience as an employee of a commercial radwaste disposal company will typify these impacts. A fire in a radwaste storage area at a research institution caused several thousand dollars of damage to the facilities. Fire personnel attributed the cause of the fire to spontaneous ignition of toluene vapors from the thirty drums of scintillation vials stored in the area.

In a second instance, a tractor trailer, carrying a load of institutional radwaste to the burial site, wrecked in the Washington, D.C. area. Although the release of materials within the trailer was minimal, and no radioactive contamination outside of the trailer was noted, a national newspaper headlined the event the following day as "Atom Truck Crashes on Capitol Beltway".

Alternatives

A comprehensive assesment of alternatives is beyond the scope of this paper. However, three alternative do warrant a brief discussion.

In addition to its impact at the burial site, the disposal of nominally contaminated scintillation fluid represents an opportunity cost. Recycle of scintillation fluid would reduce the impact of the material at the site, as well as reduce the demands for production of the scintillation fluids. Industrial scale recycling of the fluid is not currently noted. However, small scale recycle through evaporation-distillation has been used at at least two universities (Lu77 and Ha77). No quantitative data on the efficiency of this method is available.

A less sophisticated recycle procedure was noted at several universities (An77), and could easily be applied in many situations where scintllation counting is used. This involves a reuse of scintillation vials (and fluid) which contain activity levels below a predetermined limit (dependent upon the application for which the recycled vials are intended. Where the samples contain non-miscible materials (e.g. contamination survey wipes), a recycle of 50% of the vials is common.

Incineration of waste scintillation fluid is another alternative. The potential impacts of the organics in the trenches and a reduction in volume of waste buried would result. If the incineration facilities were on-site or within the region, transportation impacts would be reduced. Additionally, packaging the material for incineration, rather than containment, would allow the use of less expensive, or recyclable shipping containers.

Estimated activity concentrations in waste scintillation fluid are 7×10^{-3} Ci/ml of ^{3}H and 0.24 Ci/ml of ^{14}C (Gr77). Applying these activity concentrations to the entire volume of scintillation fluid (in vials) shipped in 1977 (486,134 liters) results in a total activity of 3.4 Ci of ^{3}H and 0.24 Ci of ^{14}C. These levels of activity would seem to make incineration a reasonable alternative, at least from a radiological point of view.

Local or regional burial sites for non-fuel cycle radwastes is another alternative. Some institutions already practice this by on-site burial, or by burying radwastes in a local landfill (An78). Regional waste facilities would reduce or eliminate the transport and burial impacts. They could include an incineration facility to reduce some of the collection impacts.

Conclusion

The current direct cost of institutional radwaste disposal is $\$10.90/ft^3$ (Be79). This means a total 1977 expenditure of nearly $3 million for containers, transport and burial. Assuming that institutional radwastes still only comprise 30% of all non-fuel cycle radwastes, as they did in 1975 (An78), then the total direct costs for non-fuel cycle radwaste disposal would exceed $10 million. This does not include personnel, facility and equipment costs, much of which [illegible] public sector.

Assuming that only 35% - 40% of the total monies spent on non-fuel cycle radwaste disposal are for burial, this yields a total burial expenditure of no more than $4 million spent in burial costs in 1977 by non-fuel cycle sources. Regional waste disposal concepts, whether burial, incineration, or some combination of the two, would further reduce these monies to about $800,000/site, assuming only five regional sites. Considering only institutional radwaste sources would reduce this to $250,000/site. Whichever figure is used, the question is, is this revenue source sufficient incentive for industry to do the planning, evaluation and capital outlay necessary for the establishment of such sites?

Another alternative would be to reduce bural site impacts by restricting, through regulation, the allowable forms of radwaste for burial (e.g. solidification of scintillation fluids). This would make impacts within the institution, in the form of additional personnel and facilities requirements, more acute. More likely, such waste processing as would be required would be taken over by intermediate radwaste disposal companies, which would greatly inflate radwaste disposal costs.

It is clear that more consideration of this problem is needed. That is why this paper is only "A Preliminary Assessment . . ."

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Gr77 Granlund, R., Incineration of Waste Scintillation Fluid", presented at "Management of Low Level Radioactive Waste Symposium", Atlanta, GA, May 23-27,1977.

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Oa77 Oakley, H., Chem Nuclear Systems, Barnwell, SC, personal communication, 1979.

DISPOSAL PROCESSES	RADWASTE PARAMETERS Volume	Form	Activity
Collection	Facilities Personnel Containers	Chem Exp. Fire/Expl.	Rad. Exp.
Transport	Fuel Accidents	Fire/Expl.	
Burial	Bur. Space	Bur. Trench Integrity	

Figure 1. Matrix of Radwaste Parameters and Disposal Processes with Specific Impacts

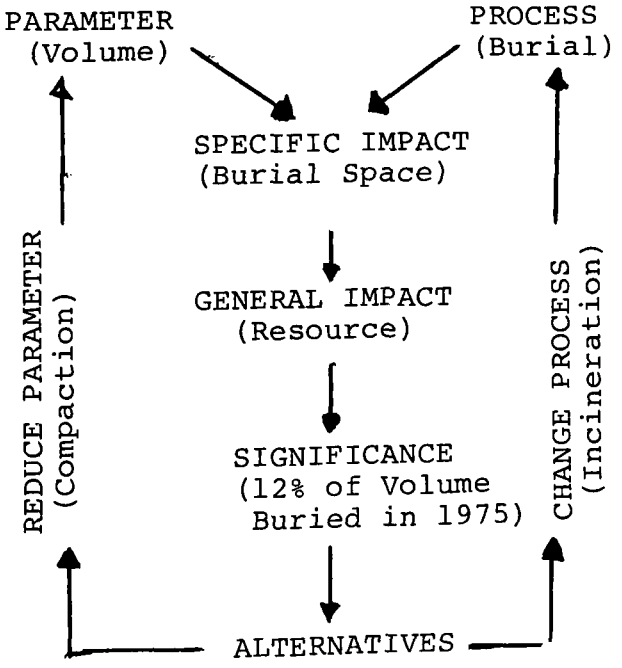

Figure 2. Schematic Diagram of Impact Assessment (with Eaxample)

RESOURCES	ENVIRONMENT	HEALTH & SAFETY
Personnel	Burial Trench	Chem. Exposure
Facilities	Integrity	Fire/Expl.
Containers		Accidents
Fuel		
Burial Space		

Figure 3. Impacts of Institutional Radioactive Waste Disposal

THE STATUS OF LOW-LEVEL RADIOACTIVE WASTE DISPOSAL-- HOW TO PLAN A DISASTER!

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Abstract

The nuclear industry is faced with serious problems in the transportation and burial of low-level radioactive wastes. Soaring burial costs, state regulations regarding transportation routes, and lack of direction from regulatory agencies are problems that must quickly be resolved.

In order to gain control of this situation four major steps must be taken. First, states must accept their fair share of responsibility in the "waste" problem. Regulatory agencies must recognize the seriousness of the problem and develop a schedule for action. The nuclear industry must assert itself in a positive manner regarding the safety of nuclear power, and the low-level waste burial ground situation must improve.

Introduction

The present status of the low-level waste scene is quite complicated and appears to be headed in the direction of more confusion.

Table I outlines the current nuclear reactor status given by the Atomic Industry Forum (AIF) as of November 29, 1978. There are a total of 203 reactors which have operating licenses, construction permits, or are on order. These 203 reactors total 197,918 MWe. In addition, Commonwealth Edison Company (CECO) recently placed an order for two 1,150 MWe nuclear units from Westinghouse. However, at the same time, Public Service Electric and Gas Company cancelled all four of its floating platform nuclear plants. Projections had previously indicated that only two domestic nuclear reactors would be ordered during 1979; those two were the CECO plants which have already been ordered. Therefore, the prospects for nuclear steam supply system vendors during 1979 appear to be bleak indeed.

In a recent AIF study, it was indicated that a normal PWR produced 40,000 ft^3 of solid low-level waste per year. A BWR produces 55,000 ft^3 per year. Considering the number of plants now having operating licenses, this would result in a range of 2.8 to 4.0 x 10^6 ft^3 per year to be shipped to licensed burial grounds. As a comparison, the amount of low-level solid waste produced by a PWR in a 40-year life of a plant is 1.6 x 10^6 ft^3. An average-size PWR containment vessel has approximately five million cubic feet of space. The low-level solid waste produced by this plant would fill approximately 32 percent of the containment in 40 years. One may ask why such a comparison? The response is that a recent on-site storage study considered a decommissioning alternative of placing the low-level solid waste in "fixed" burial facilities, i.e., the containment, after retrieval from on-site burial facilities. This simple arithmetic is provided so that low-level waste from an operating nuclear power plant can be gained.

The attitude in our industry today is very similar to that of being in "limbo." For example, the regulatory agencies appear to be moving slowly in making decisions regarding low-level waste. One only has to look at the status of high level waste to know that there is chaos; and low-level waste is following suit. It is my feeling that low-level waste could be the "tail wagging the dog." As an example of the lack of recognition of the magnitude of the problem, one DOT official was recently asked, and it was reported in Nucleonics Week, why there was not an official government position regarding transportation of radioactive waste. His response was, "I personally have not seen anything to indicate an emergency. I doubt that the DOT will jump into the fray to make a precipitous decision." Further, in Nucleonics Week a utility executive recently stated that, "The nuclear option is dead, there is no question about it. We at Consumers Power continue to plan nuclear plants but each year we wipe them off the planning board--they are too expensive." It is obvious that there is confusion and lack of direction in regard to low-level waste.

Burial Grounds

What are the problem areas facing this industry? First and foremost, there must be a burial site for low-level waste if we are to bury the waste. Those that are familiar with this industry know that the low-level waste burial ground history has been quite confusing. Table II shows the history of low-level waste burial sites. For example, in 1975, there were six operating burial sites. Currently, only three of these sites are operating; two west of the Continental Divide and one in the eastern part of the United States. The Barnwell site located in South Carolina recently considered placing a limitation of 135,000 ft^3/month on shipments into the burial site. Although this limitation has not as yet been imposed there is always the possibility that the State of South Carolina will become more concerned about taking most of the waste east of the Mississippi. Also, the State of South Carolina is beginning to scrutinize what comes into the site. For example, recently, the State placed a hold on receiving oily waste shipments. The State is currently evaluating the various methods of solidifying oily waste and feel that until such criteria is set, the site can no longer receive this type of waste. The State of South Carolina is also beginning to look at organics contained in liquid scintillation vials. There is always the impending threat of placing a hold on burial of urea-formaldehyde solidified low-level waste. In addition, Chem Nuclear Systems, Inc. (CNSI), recently suspended efforts to open a burial site in New Mexico due to licensing problems. Nuclear Engineering Company (NECO) has run into continual red tape in attempting to license additional space for Sheffield. It was reported in Nucleonics Week that one NECO official said, "...it's paralysis by analysis." There are two reasons--safety and financial--why Maxey Flats will probably remain closed. First, Kentucky will insist that state and federal studies, now under way, "prove that it is 100 percent safe to bury low-level waste at the site before operations resume. Second, the state would have to charge taxes high enough to discourage private industry from operating the site in order to maintain a profit and ensure long-term care.

However, there is some attempt to open a low-level waste burial site in Lyons, Kansas at the site of the old high-level waste salt mines. The opening of other low-level waste burial sites would be of benefit to the nuclear industry.

Figure I shows the increase in burial costs, another problem faced by the industry.* During 1978, burial costs at the Barnwell, South Carolina site increased approximately 85 percent for waste in the 0-200 mRr/hr range. This is a significant price increase. On December 28, 1978, users of the Barnwell site were notified of additional price increases, primarily for higher level waste, various handling and surcharges. At this point, the cost of burial is becoming an important factor in the budgets for operating nuclear power plants.

Transportation

A second and perhaps even more significant problem, is that of transportation. The routes to get to the burial sites are being threatened and costs are increasing. Restrictive statutes and ordinances have been adopted in over 50 states and localities. For example, Clergy and Laity Concerned (CALC), a group of northwest nuclear opponents, hope to choke off transportation routes to Hanford. Their aim is the Hanford facility itself, but the impact will be to stop the receipt of radioactive waste at the low-level burial grounds.

Figure II shows the increase in the cost of transportation. As one can see, the increase from 1973 to 1978 has been approximately 30 percent. As a comparison, Table III gives a simple analysis of the impact of the cost of transportation of low-level radioactive waste for one particular nuclear plant located in the midwest. This analysis is for a shipping cask that contains approximately 170 ft^3 of waste. Before the Sheffield, Illinois facility was closed, the plant was paying about $400 for tranporting the waste to the burial site. When Sheffield closed the cost for transportation resulted in a four-fold increase for shipments to Barnwell. If the Barnwell facility were to close, the cost for transportation would be about an eight-fold increase. This analysis does not include the increased probability of an accident on the highways. This cost increase does not parallel the cost increase received from the burial grounds. It is my feeling that the transportation problem could become the major problem facing the industry due to the fact that many states are considering the banning of routes for transporting any type of radioactive waste through their localities and states.

Equipment Costs

Due to significant efforts, and changes in design, one radwaste hauling vendor has been able to control or practically maintain the costs for some of his shipping casks over a five-year period. This, of course, is not the case with most pieces of equipment. The escalation rate has been averaging approximately eight percent per year.

Economics

Table IV is a recent analysis of how different costs have risen since 1967 and since one year ago this January. The point is that transportation

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*The top curve is the price increases for western burial sites operated by NECO. The cottom curve is the price increases for the Barnwell site operated by CNSI.

and equipment costs are falling within the ranges shown in Table IV. The recent burial price increases have indicated a potential "run-away" situation. With increases of approximately 85 percent for the 0-200 mR/hr range waste occurring during 1978, one must keep a cautious eye on the future if he is to budget this portion of operating costs adequately.

Solutions

What must be done to effectively get the nuclear industry out of the dilemma that we now face? I believe that there are basically four major steps that must be taken.

1. First of all, states must accept the fact that they are involved in the production of radioactive waste if nuclear plants are located in their states. Most states produce medical and institutional radioactive waste and all states produce some toxic chemical wastes. An official of the State of South Carolina recently related that the state was doing its share by accepting low-level radioactive waste and it was time that someone else handled the other types of waste, such as toxic chemical waste. I am suggesting that a serious look be taken by the states and that their involvement in production of radioactive and toxic chemical wastes be reviewed. The states must somehow become involved; either by providing a low-level waste burial ground, a medical/institutional radioactive waste burial ground, a toxic waste burial ground, or by entering some cooperative venture. A great deal of cooperation would be required by the states, but it is a simple fact that all of the states make a contribution to the waste produced and they should share in the handling and burial of these wastes.

2. The second major step is that the regulatory agencies must not only recognize the peril of not solving the low-level waste problem, but that they must develop a schedule to seek answers to these problems before the problem becomes even more serious. For example, the DOE is considering a contingency plan to take low-level waste at its burial sites in emergency situations. However, to my knowledge, no real plan has been placed in effect and if Barnwell were to close for some particular reason, the availability of casks to ship low-level waste to Beatty and Hanford would create a real problem for the nuclear industry.

3. The third major step is that the nuclear industry must go on the "warpath." There is enough evidence to show the economic advantages and safety of nuclear power that a more positive attitude must be taken. Is nuclear power really cheaper? The New England power plants have been producing power for 1.293 cents/kwh over the past two years compared with 2.662 cents for oil-fired capacity. Table V gives recent cost estimates from Ebasco Services, Inc. indicating their estimates of nuclear versus fossil over a 10-year period. The nuclear plants are currently estimated to cost approximately 1,648/kw vs. 1,226/kw for fossil plants. Most people recognize that the nuclear power question is now a political rather than a technical or environmental one. We must direct our warpath towards the political arena and become as positive and outspoken as the anti-nuclear proponents.

4. The fourth major step is to improve the burial ground situation. There are basically two alternatives as far as I can see. First, we can continue with commercially-operated sites and, second we could proceed with government-controlled burial sites. At this time, I am not sure which is the best direction to take. It is obvious that there could be some advantage now to having government-controlled burial grounds because the ability to open government burial sites would be easier. However, there are disadvantages that must be considered. This question must be studied and a decision made prior to a decision being forced upon the industry.

Conclusion

The bottom line is that unless some steps are taken, an already serious problem could become a disaster. There are power plants that have such limited storage capacity that if Barnwell were to close down, and if radwaste disposal vendors could not provide adequate shipping casks for hauling the waste to the West Coast, these plants would be faced with closing down due to radwaste disposal problems. It is about time that we take this situation in hand before it becomes a much more serious problem. We have an opportunity to do just this if we will step forward and speak out, talk to our Congressmen and write letters to the appropriate authorities making these decisions.

TABLE I
NUCLEAR REACTOR STATUS REPORT

72	Reactors with operating licenses		52,273 MWe
90	Reactors with construction permits		98,968 MWe
37	Reactors on order		42,565 MWe
0	Letters of intent/options		0
203		Total	197,918 MWe

TABLE II
HISTORY OF LOW-LEVEL WASTE BURIAL SITES

Name	Location	Operating	Comments
West Valley	New York	No	Operations suspended in March, 1975
Maxey Flats	Kentucky	No	Operations suspended in December, 1977
Sheffield	Illinois	No	Operations suspended in April, 1978
Barnwell	South Carolina	Yes	135,000 ft^3 per month limitation imposed
Hanford	Washington	Yes	
Beatty	Nevada	Yes	
Chem Nuclear	New Mexico	No	Licensing effort suspended
SWECo	Kansas	No	Licensing application in

TABLE III
TRANSPORTATION COSTS FROM A MIDWEST PLANT

	Approximate Cost
Plant to Sheffield, Illinois	$ 400
Plant to Barnwell, South Carolina	1,700
Plant to Beatty, Nevada	3,400

TABLE IV
HOW DIFFERENT COSTS HAVE RISEN

Item	Percent since 1967	Percent since Year Ago
Overall living costs	101	8.8
Food, including meals out	112	11.3
Housing and operations	110	9.8
Transportation	90	6.6
Clothing and upkeep	63	4.1
Medical care	125	8.5
Personal care	86	6.7
Entertainment	79	4.7

TABLE V
COST ESTIMATES*

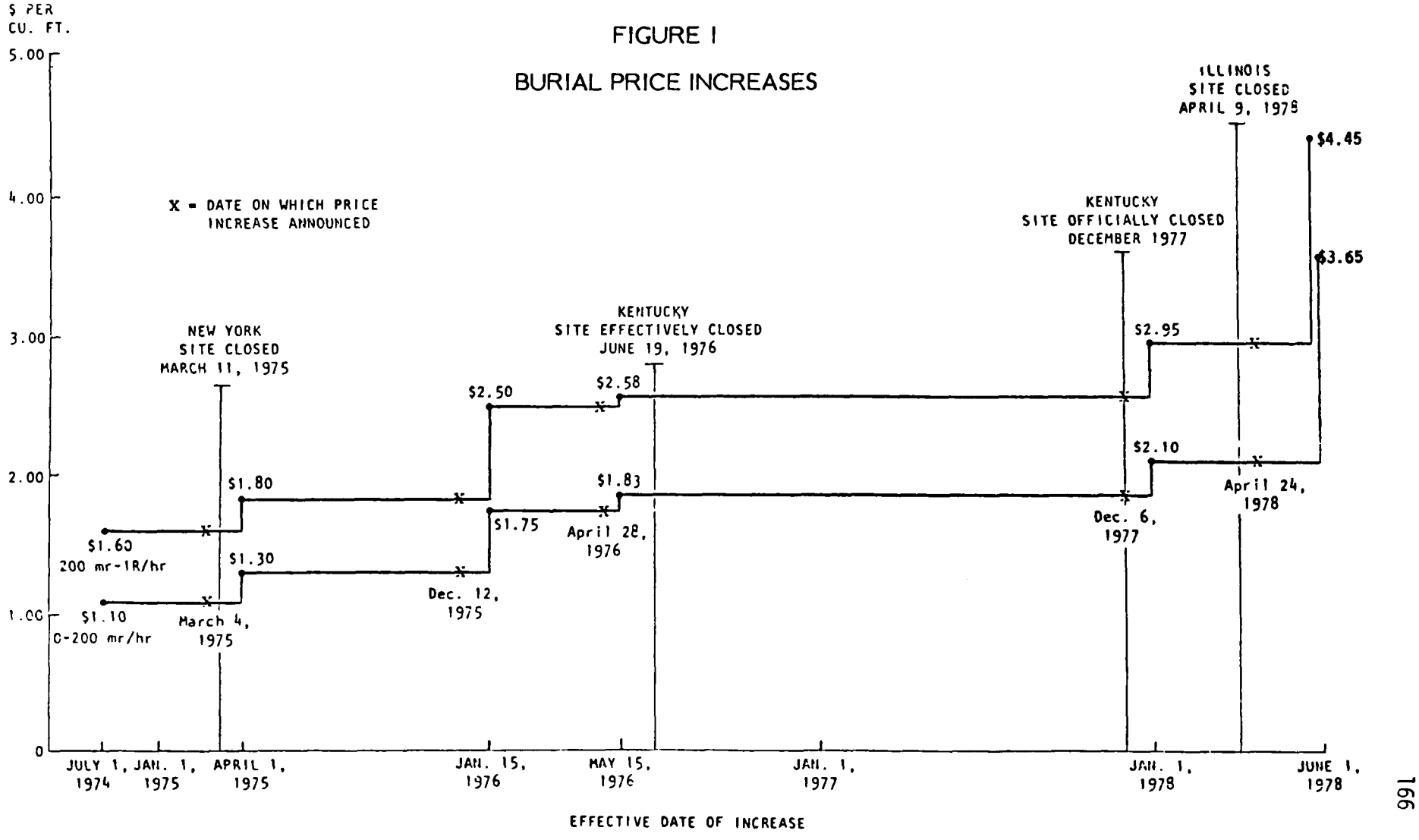
Year	Nuclear**	Fossil
1969	226/kw	183/kw
1978	1,648/kw	1,266/kw

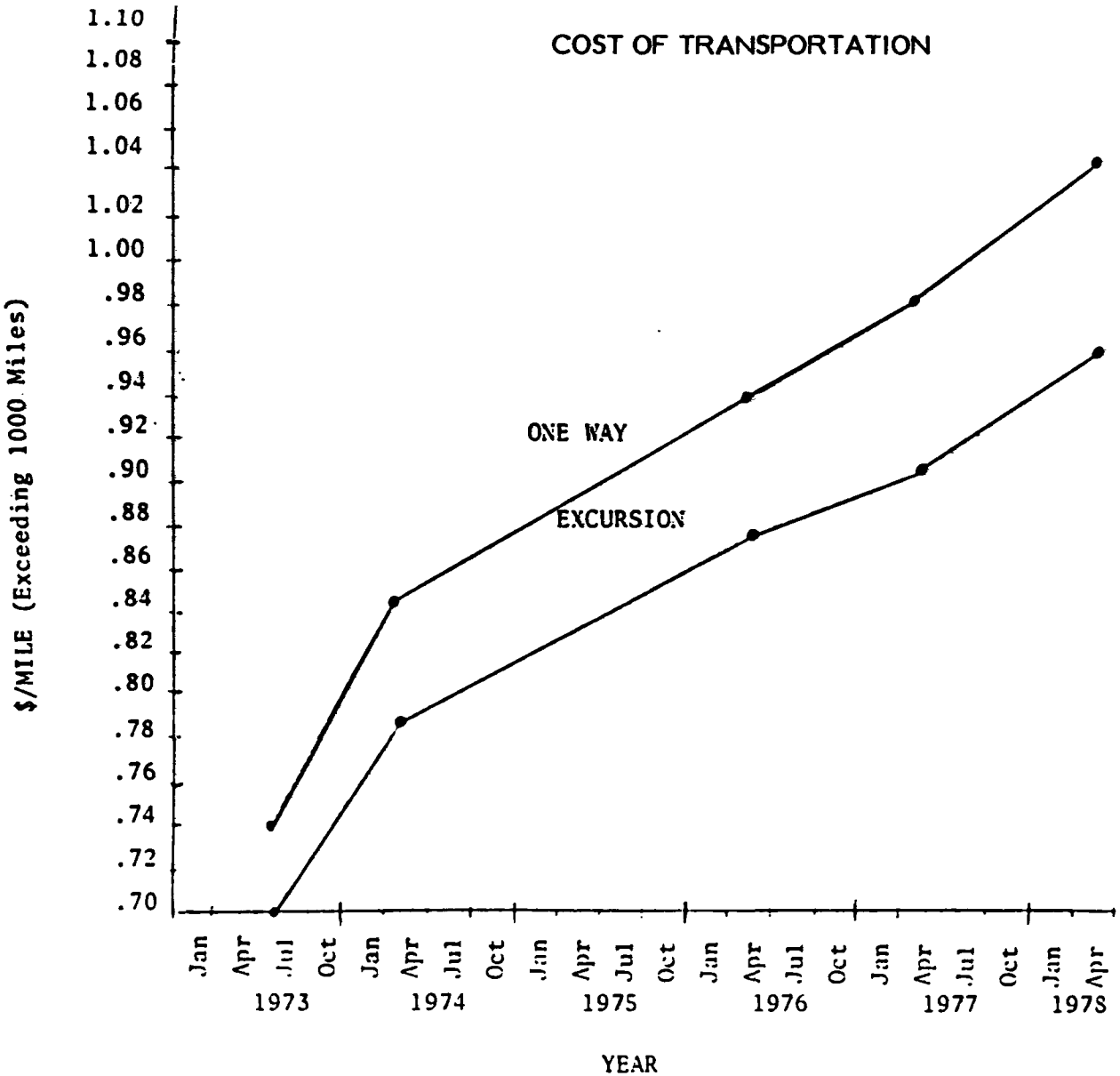
*Ebasco Services, Inc.

**Regulatory changes account for 78 percent of soaring nuclear plant costs.

FIGURE I

BURIAL PRICE INCREASES




COST OF TRANSPORTATION
$/MILE (Exceeding 1000 Miles)
1.10
1.08
1.06
1.04
1.02
1.00
.98
.96
.94
.92
.90
.88
.86
.84
.82
.80
.78
.76
.74
.72
.70
ONE WAY
EXCURSION
Jan
Apr
Jul
Oct
1973
1974
1975
1976
1977
1978
YEAR

SHALLOW LAND BURIAL--WHY OR WHY NOT?

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Abstract

This paper summarizes a master's thesis on the state-of-the-art for shallow land burial of solid low-level radioactive wastes. The coverage of the thesis, which is condensed for this paper, ranges from site selection to problem case histories. Inherent in such coverage is the assessment of risk, the discussion of operational and management problems and the real significance of off-site migration--this topic will be discussed in light of the stands taken that the migration is a serious problem and that it is not. Emphasis is on the engineering parameters of importance in site selection, and what pretreatment, if any, is needed.

Introduction

This paper will present considerations which should be included in making the political decision to bury or not to bury. Such a decision should be made with a background of what has been done, what problems have occurred, what alternatives exist, and what risks are entailed.

History of Shallow Land Burial

Shallow land burial (SLB) of low-level radioactive wastes has been practiced in the United States since the advent of the atomic age in the 1940's. SLB competed with disposal at sea until the 1960's when the latter practice was abandoned (Len67). Starting in 1960, the Atomic Energy Commission (AEC) set up criteria for licensing commercial land burial sites (USC76). These criteria were:

1. "A written commitment from a responsible state official that the state would assume control over the burial site in event of default or abandonment of the site by the commercial operator."

2. "The geological and hydrological characteristics of the site must be such that containment of the waste materials is assured in a manner that will not endanger public health and safety."

3. "The waste must be in solid form prior to burial."

4. "Establishment of an environmental monitoring program."

5. "The packages in which the wastes are transported to the burial site meet the NRC and DOT standards for packaging and transportation." (Note: NRC is the Nuclear Regulatory Commission and DOT is the Department of Transportation.)

6. "As part of the licensing process, any future burial grounds licensed by the NRC would require a review under the provisions of the National Environmental Policy Act (NEPA)."

There have been 6 commercial sites licensed since September 1962; they are: Beatty, Nevada; Maxey Flats, Kentucky; Richland, Washington; and Sheffield, Illinois (Nuclear Engineering Co., Inc.); West Valley, New York (Nuclear Fuel Services, Inc.); and Barnwell, South Carolina (Chem-Nuclear Systems, Inc.) The Barnwell site is now receiving about 80% of all the wastes being buried commercially.

West Valley and Maxey Flats are closed due to operational problems and Sheffield has used up its authorized space. In addition to the commercial sites, there are 5 major Department of Energy facilities which have shallow land burial. They are Oak Ridge National Laboratory, Oak Ridge, Tennessee; Los Alamos Scientific Laboratory, Los Alamos, New Mexico; Idaho National Engineering Laboratory; Idaho; Hanford, Richland, Washington; and Savannah River Plant, South Carolina.

The restrictions on wastes accepted at commercial sites include packaging of the wastes, solidification of liquids and sludges, gases only in containers (if at all), limits on transuranics, limits on fissile material, and limits on chemical toxicity.

Problems With Shallow Land Burial

Problems arising at low-level solid radioactive waste burial grounds may be site-specific, dependent on the regional hydrologic, geologic, and climatic conditions, or they may be common to all sites. The major problem has been the migration of radioactivity away from the burial sites in concentrations that have caused political concerns although knowledgeable radiological health professionals have stated that the observed levels of contamination posed no threat to the safety and health of the public (Me76b). Migrations have occurred at 4 sites: 2 commercial, Maxey Flats and West Valley; and 2 Federal facilities, Oak Ridge National Laboratory and Idaho National Engineering Laboratory (USC76; Me76b).

The migration problems have probably been a result of the emphasis on economics and convenience in the selection and operation of the disposal sites. The criteria utilized for site selection included factors such as isolation of the site from population centers and water supplies, depth of burial to minimize radiation at surface, limits on form and type of waste, and operational procedures. The only reference to geologic or hydrologic considerations was the specification of the depth of the groundwater table (USC76).

The migration of radioactivity at Maxey Flats has been attributed primarily to the accumulation of water in the trenches, both those which had been completed and those still receiving waste, and the subsequent hydrological transport of the radioactivity from the site. In the period from 1972 to 1975, the State of Kentucky and the licensee were able to correct most of the problems (Co76).

A problem of diversion of contaminated equipment that was to be buried has received wide publicity in connection with the Beatty site. The State of

Nevada found that tools, clocks, and other contaminated equipment had been removed from the site by employees. The license was suspended until the situation was corrected (USC76).

Other problems have related primarily to poor management procedures in that maps and records do not exist; they were never prepared or they burned. There have been some problems with fires and explosions. There is no record of any problems of migration or effects from the possible emanation of gases from burial sites. Shrinkage of the buried wastes resulted in accumulation of surface water in the burial trenches at West Valley (Mo68).

Mobilization of the Waste

Solid radioactive waste disposal in terrestrial environments is subject to infiltration and leaching by ground and surface waters. The initial result of water contacting the waste would most likely be formation of leachate having the approximate characteristics of ordinary commercial landfill leachate plus the presence of radioactivity.

Because of the low density of the waste, oxygen is available for aerobic decomposition during the early stages of organic breakdown; this decomposition yields carbon dioxide, water, and nitrate. As the oxygen supply diminishes, anaerobic decomposition produces methane, carbon dioxide, water, organic acids, nitrogen, ammonia, and sulfides of iron, manganese, and hydrogen. Carbon dioxide production, as an example, could dissolve strontium in the bicarbonate form (Gia77; Ma65).

Other characteristics of landfill leachate include high chemical oxygen demand (COD, up to many 1000's of mg/l), volatile acids, and a pH in the range of 3.7 to 8.5. Constituents which release or mobilize contaminants from the waste include acetic, propionic, isobutyric, and valeric acids. In brief, landfill leachate contains agents capable of solubilizing cobalt, strontium, cesium, and other radionuclides, including some of those generally considered relatively insoluble such as plutonium (Me76a,b).

Alternatives for Waste Disposal

Treating and/or packaging of low-level radioactive waste prior to burial can serve several purposes, including (1) volume reduction; (2) the reduction of the mobility of the radioactivity; (3) enhancement of the safety associated with the handling and disposal; and (4) recovery of plutonium and other transuranics (Gil77). The most burdensome problem facing SLB is not the radioactivity of the waste but the large quantity of waste (NRC76). This problem is likely to get worse because of opposition to the establishment of new sites (NY76).

Several methods are available for the pretreatment of waste before disposal. These include sorting, incineration with or without pneumatic classification beforehand, baling and compaction with or without prior shredding, and acid digestion (Gil77; NRC76). Wastes may be hand sorted at its source or at the disposal site into combustible/noncombustible glass, metals, hazardous chemicals/radioactive, and short/long lived radionuclides (Gil77; NRC76).

Incineration is perhaps the ultimate volume reduction technique short of chemical separation and concentration. Volume reductions of 70 to 90 percent can be achieved (Mc70). Other advantages of incineration include the stabilization of the organic waste, a product with better understood properties for better leachate prediction and/or prevention, and the end of the threats of long-burning, subterranean fires at the sites (NRC76). Incineration has received widespread acceptance in Europe, but in the Unitd States, the attitude exists that the extra volume reduction often does not justify the additional cost (Mc70). Because baling and compaction offer relatively low cost volume reductions to 75 percent while requiring little segregation or sorting, they have been more attractive than incineration in the United States (Mc70; St75).

Acid digestion is still in the pilot plant research stage (Gil77).

In addition to pretreatment schemes, there are probably viable alternatives of engineering the containment of the wastes regardless of whether any pretreatment has been applied. These alternatives include the catchment and treatment of the leachate (especially the early flows), the interruption of the water access to the burial trenches and/or the wastes and the sorption of the radionuclides from the migrating water.

The catchment and treatment of leachate can be accomplished by preleaching the wastes in containers or pits, by putting the trenches on an impervious bed of clay or plastic, by pumping the waters in or beneath the trenches, or by leaching the pretreated waste, especially the ashes from incineration. The interruption of the water access to the wastes can be accomplished by cutoff structures or by encapsulation of the wastes. The sorption of the radionuclides from the migrating water cannot be relied on to provide total containment of all the waste radionuclides; however, sorption may give enough holdup to reduce the migration rate to acceptable values.

Current research at several locations throughout the United States, including the University of Texas at Austin, is aimed at providing satisfactory engineering designs for shallow land burial of low level radioactive wastes.

Risk and Risk Assessment

The design of a waste disposal site is based on either complete containment of the waste, thereby allowing for zero planned discharge to the environment and subsequently low risk to the environment, or the design may be based on a finite amount of radioactive contamination leakage. The leakage would have maximum limits based on the existing or proposed risk criteria developed by the regulatory agency. The level of risk allowed will tend to dictate the disposal practice for low-level radioactive waste.

Two of the major problems with risk assessment of a radioactive waste burial site are: first, scientist and health physicists are unable to determine the exact dose which would prove to be innocuous by both the genetic and somatic definitions (NAS72; Led65). This problem centers around the "how safe is safe enough" cliche. Secondly, the design period of the burial sites can only be verified by historical data, accelerated life testing, or models based upon the experience of similar systems. This is a major problem with radioactive waste burial sites due to the long hazardous lifetimes of the radioactive waste (USE77; USE78a,b).

The range of possible risk associated with the disposal of low-level radioactive waste is related to the amount of money spent on ensuring containment of the radioactivity. The range of risk could be extreme. High risk could be the result of very little research and maintenance of a disposal site, and low risk could be the result of encapsulation and/or solar disposal. Outside of the possibility of a moratorium being placed on all waste production, the risk from future waste cannot be entirely eliminated.

Conclusion

Why or why not utilize SLB as a method for disposing of low-level radioactive waste must be decided in the near future. Maxey Flats, Sheffield, and West Valley, three of the four eastern disposal sites, have already quit receiving waste, resulting in 80% of all low-level radioactive waste being sent to Barnwell.

The reasons why SLB should be utilized include economics and simplicity of site operations. Yet, these two advantages have also been major sources of problems at existing sites. Economics, being the culprit, in that dollars were saved by not practicing thorough site selection techniques, and by inadequate engineering design of the burial sites. Simplicity of site operations has resulted in improper disposal practices and lax operational management programs.

The past operational practices and problems, together with results from ongoing research should provide enough data to allow for the proper design and operation of disposal sites so as to ensure safe containment of the radioactivity. Utilization of pretreatment and volume reduction techniques together with proper site management should allow for optimal use of areas allocated for disposal operations, and result in minimum risks to public health and the environment.

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WASTE MANAGEMENT OF URANIUM MINING AND MILLING OPERATIONS

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ABSTRACT

Uranium mining and milling operations associated with waste practices are somewhat different from other nuclear operations in that no new radioactive material is generated.

The methods and procedures of uranium mining and milling operations using the acid-leach solvent extraction method are described using the Sherwood Project of Western Nuclear, Inc., as an example.

DISCUSSION

Uranium mining and milling operations remove, pulverize, and disperse radioactivity which is naturally deposited in the Earth's crust. No new radioactivity is created through these operations. The uranium and its daughters, including radon a gasseous radionuclide, are merely converted to a more dispersible form. It is the large volume of this low concentration waste coupled with its readily dispersible chemical and/or physical form which makes the safe disposal of uranium tailings a unique challenge.

Most currently extracted ores contain uranium concentrations of from 0.05% to 1% (IA76), with the more recently constructed U.S. mills utilizing the lower concentrations. This presentation will attempt to describe the various sources of waste from Uranium mining and milling. Western Nuclear, Inc., Sherwood Project will be an example of how a typical open pit mine and acid leach mill manages its waste.

SHERWOOD - GENERAL DESCRIPTION

Each site has particular characteristics which affect how it manages its waste. The Sherwood Project is located on the Spokane Indian Reservation, approximately 33 miles north, northwest from Spokane, Washington. Its location high atop a bluff overlooking Lake Roosevelt affords it one of the most picturesque settings for a uranium milling operation in the U.S.. The topography is generally hilly. Annual rainfall at the site is estimated to be 20 inches (50 cm) per year, slightly more than Spokane. The average temperature is 44.7°F (9.3°c). The immediate vicinity of the mill is sparcely populated with approximately one person (0.9) per square mile (0.5 person/km^2). The predominant industries are ranching and multi-use forest with farming and recreation minimally represented. The only groundwater on the site consists of low volume springs used previously for livestock waterings (US76). Lake Roosevelt is the major source of water in the near vicinity of the mill.

MINE WASTE SOURCES AND CONTROLS

At Sherwood's open pit mine, waste rock is the largest volume mine waste, and is anticipated to be 88% of the volume of the mined rock. (Fi78)

The ore is routinely sorted using its radiation emission properties. A portable GM probe is used while the ore is still in the ground. A second GM probe tower is employed to measure the average ore content of each truck load of excavated material. From these two determinations, the material is divided into three piles: waste rock (or overburden), protore (or ore that is not yet economic to mill), and ore destined for the milling operation. Part of the overburden has already been used as a source of crushed basalt for construction materials for the mill access road and the horizontal drain blanket at the base of the tailings dam. Most of the overburden, however, is transferred from one area to another where it is known there is no underlying ore body.

Low grade ore or protore is another source of mine waste and the definition of this ore varies from one mill to another. For the Sherwood Project, protore contains less than 0.06% and more than 0.035% uranium (Fi76). This protore is piled on 2 pads to await changes in the economics of the uranium industry. Ore, that with 0.06% and higher uranium content, is stockpiled on one pad near the crusher building to await processing in the mill.

Erosion during ore storage is controlled by two means (US76). The ore pads themselves are sloped towards the mill so as to impound any surface runoff. There will also be some terracing on the slopes towards Lake Roosevelt so as to alleviate erosion. Currently all ore dumps are sloped into the mine site to allow any runoff to be reabsorbed on-site.

Because of the low concentration of uranium in this ore body, the Sherwood Project is not expected to generate a large volume of mining equipment which cannot be decontaminated to acceptable levels for release to uncontrolled areas. Any unrecoverable contaminated material will be put into the tailings retention system. No such waste has been generated to date.

Most of the liquid mine effluent is due to surface runoff of the seasonal rains. Very little water is used for drilling. The Sherwood Project has encountered little groundwater in the mine area, and that water which has been found is very localized ground water (Fi76).

Wind erosion tends to be naturally controlled. The stockpiled material breaks down easily; however, its high moisture content tends to control dusting naturally. The Sherwood Project has a system for water spraying the mine areas to control the dust but, as yet, it has not been necessary to use the system for control of wind erosion.

The stockpiled ore next enters the crushing circuit. Once crushed, the fine ore storage building protects the majority of the crushed ore from erosion by wind and rain. Some minimal erosion can occur as the building has been erected on pedestals for ease of maintenance. The expected lifetime of the ore body is 10.6 years (US76). After that time, erosion of the mine area is planned to be controlled by limited restoration of the topography with overburden, sandy surface soil, and seeding with native vegetation.

Potential sources of airborne contaminants from the open pit mine are primarily the operation of excavation equipment and trucking on unsurfaced roads. In comparison, blasting is a small source of dust (FI78). Non-sanitary waste water from the mill has been used to control dusting in the mine area.

A limited airborne monitoring program had been operating for two years prior to the beginning of mining operations. A more elaborate monitoring program begun in April, 1977 has seen no appreciable increase in activity since mining operations commenced (Me78).

MILL WASTE SOURCES

A short discussion of the mill process will aid understanding of waste sources. The Sherwood mill employs an acid leach process using a combination of Sulfuric Acid and Sodium chlorate at a pH of about one to dissolve the uranium from the crude ore (US76). This slurry is then treated with polymeric fluculent to separate the uranium bearing solution from the barren waste by the process of counter current decantation. The dissolved uranium then enters the solvent extraction process which uses high flash point kerosene as the carrier for the active extractant - a tertiary amine. Following extraction into the solvent, followed by an ammonium sulfate strip of the solvent, the concentrated uranium in an aqueous phase is neutralized by anhydrous ammonia, yielding the yellowcake product [ammonium diuranate $(NH_4)_2\ U_2O_7$] (Mi78). The remaining water content of the yellowcake is removed through a thickener process, centrifuge, washing the sulfates out, and finally evaporative heating in the roaster. The dried yellowcake is then transferred to a hopper to await packaging in 55 gallon drums.

This typical milling process, therefore, generates the following wastes:

unreacted, acidic, barren ore
flocculated and barren precipitates
waste organic solution
neutralized, barren ammonium sulfate solution
aqueous wastes from the thickeners and centrifuge
contaminated runoff
dusts from the crushing circuit
sulfuric acid vapors
dusts from dryers and packaging areas
radon emmissions from radium bearing wastes

Non-routine wastes also result from:

emergency spills or overflows
decommissioning of plant
long term emissions and erosions from tailings area

Chemical constituents other than Uranium of the ore body which need to be considered include:

Molybdenum
Vanadium
Zinc
Calcium

As with most acid leach mills (IA76), salts are a major threat to the ground water. Fortunately, there is minimal ground water at the Sherwood Project and the sorptive capacities of the soil are natural barriers to control migration of the salts (US76).

WASTE HANDLING SYSTEM

Tailings area. The most prominent feature of the Sherwood Project and the feature which will handle the major share of all wastes generated is the tailings retention system. The mill tailings are deposited in an above ground dammed structure. This tailings area is located in a valley which drains eventually to Lake Roosevelt (the backwaters of the dammed Spokane River). The 163 acre (63 hectares) dammed tailings area (WN76) is surrounded by a diversion ditch designed to divert the projected 100 year flood which was postulated to follow the day when the projected 50 year flood occurred. This tailings retention area is created by a 63 foot (19 m) high, 2400 foot (740 m) long dike which is designed to contain approximately two years of tailings (US76). The dike will be increased in stages to an eventual height of 108 feet (32 m) to contain the tailings from the projected 10.6 years of mill operations.

The entire tailings area is, or will be, lined with a 30 mil (1.7 mm) polyester reinforced hypalon liner. Hypalon is a synthetic rubber which is reasonably resistant to weathering, ozone, and sunlight. It has high resistability to acids and alkalis. It is also moderately resistant to various organic solvents and biological degredation. It is usually supplied in the unvulcanized form and can be seamed by heat sealing or solvent welding (St78).

In 1974-1975 when this waste retention system was first proposed, storage of the tailings in the mined out portion of the mine area was considered, but was rejected because of the increased hazard to any ground water sources in the area and to Lake Roosevelt. The use of the above ground lined tailings system was preferred at that time because it essentially removed the aqueous wastes from the environment (US76). A small portion of the hypalon liner was strength tested in place using heavy construction equipment. Aside from minor punctures from pointed rocks, no tears or major flaws were noted.

The synthetic liner on the waste ponds is designed to contain all liquid effluents from the mill, thus facilitating the re-use of water and storage of rain water. The liner should also prevent migration of this contaminated water through the underlying silty sands. Thus the large amount of salts normally generated through the acid leach process should remain adequately sequestered in the lined tailings structure.

The tailings retention system receives the neutralized tailings slurry. The previously acid tailings are neutralized with calcium oxide. The addition of the calcium oxide is beneficial because the chemical similarities between calcium and radium reduces mobility of the radium in the tailings. Also, the majority of the heavy metals are also settled out during this process. Since water is at a premium at the Sherwood mill, a pontoon mounted pump is floated in the tailings pond to pump the neutralized water back into the mill process following treatment with Barium chloride for radium removal in the aggitated barium chloride treatment tank.

This removal is accomplished in the primary and secondary precipitation ponds. These ponds comprise a total area of 3/4 acre (3000 m^2) (US76) and are lined with the same reinforced hypalon liner material used for the main tailings pond (WN76). The supernatent which is drawn from the ponds is routed back to the mill process. Any excess water which cannot be routed back to the mill is allowed to overflow into the unlined seepage/evaporation pond.

An industrial waste pond of approximately 1/8 acre (500 m^2) has also been constructed and lined with the reinforced hypalon (WN76). This pond has, so far, been used only to contain and evaporate the backwash water generated during cleaning of the filter media from the drinking water treatment plant. It has the capability, however, of containing wastes from any of the processes in the plant, but particularly those from the solvent extraction process, truck maintenance bays, and laboratory areas.

OTHER WASTES

One major feature of the Sherwood mill is that it has reclaimed a major fraction of its waste streams. Since the mill is processing low grade ore, average 0.089% uranium oxide (Fi78), and since the site has

no substantive on-site water supply and since the costs of pumping water from Lake Roosevelt are high, it is economically critical to recycle. Thus waste streams from the acid leach process, the barren ammonium sulfate (counter-current decantation) circuit, and any waste organic solution are fed back into the mill process. Aside from the economic considerations, recycling has the obvious benefit of minimizing water pollution.

The mill has been designed so that no liquid can escape the process area during normal operation as a result of any foreseeable tank or pipe failure (WN76). The floor grates, building walls, sumps and berms were designed so that there will be adequate volume to contain spills within the buildings or process areas. For example, the mill building will contain the contents of two leaching tanks which burst simultaneously.

Contaminated runoff is unlikely to occur from the mill area since the tailings area is sloped to contain any such runoff which is generated within the tailings area. In addition, the walls of the diversion ditches, much of which is comprised of granite, are expected to divert any surface runoff from intruding into the tailings area (US76).

Airborne wastes are controlled by the use of a wet scrubber atop the crusher and bag houses above the crushed ore storage buildings and conveyor transfer points. Dust control may be enhanced in dry weather by the use of water sprays. The operation utilizes a wet grinding system, thus eliminating the need for dust collection equipment (WN76).

Acid vapors in the leaching circuit, if generated, are collected through a common header and vented through demisters to trap acid and chlorine gas. Any water collected is then routed back to the process. A forced air ventilation system is used to collect and vent to the outside organic vapors generated in the solvent extraction portion of the process. The precipitation and dewatering of the yellowcake is essentially a wet process, and thus the airborne effluent control at this stage (WN76) is relatively simple.

The roaster drying area is equipped with a wet venturi scrubber to collect greater than 99% of the uranium particles contained in this process effluent. The packaging area is equipped with a micro pulsair bag house collector (WN76).

Radon emissions from radium bearing wastes are of most concern in the milling process prior to the transfer of tailings to the tailings pond. During tne acid leaching process, radon is controlled by ventillation of the mill building itself; the Counter Current Decantation process is outside, therefore, no emission control system is utilized in this wet process (WN76). Currently, the fine ore storage bin is the largest radon source at the mill site (Me78, Mi78).

Radon emanations from the operating tailings pond are generally uncontrolled, using only fresh air dilution. Calculations have been made to estimate the population doses from ^{222}Rn emissions per year to the population of a 50-mile radius. This dose is estimated to be 4.79 man-rem to the lung (US76).

RECLAMATION OF THE TAILINGS AREA

A tailings area stabilization plan has been proposed which includes provisions for both stabilization and for long term maintenance. Estimates of the amount of cover (overburden) needed to reduce radon emissions to approximately twice the background emanation rate were made and were calculated to be approximately 13 feet (4 m) (WN76, Cu76, Kr63).

One essential feature of the stabilization plan is the grading of the tailings and its 13 feet of cover to blend with the surrounding area so as to minimize erosion. The operators of the Sherwood Project have secured a surety bond from the Bureau of Indian Affairs in the amount of $6 million (April, 1978) to cover mill decommissioning and tailings stabilization costs. A separate bond was also secured from the Bureau of Indian Affairs in the amount of $176,000 to cover maintenance and monitoring of the reclaimed and stabilized area for a period of 50 years. Provision was made to re-evaluate the adequacy of both bonds periodically. This will probably be done at the time of radioactive materials license renewals.

SUMMARY

Each uranium extraction method, acid leach, basic leach, heap leach, in-situ leach, has its own characteristic waste problems (IA76). These problems may be allevaited or enlarged by the characteristics of a particular location. For instance, in-situ leaching by its very nature has solved the problem of what to do with large volumes of solid waste which is so typical of acid leach mills. However, in-situ leaching has perhaps a more environmentally significant problem with the large volume of liquid waste which is generated which may threaten the ground water (US78a). In Texas (US78a), such waste has been allowed to be discharged into more or less sequestered underground formations. This may or may not be an acceptable method of disposal elsewhere, depending on the characteristics of the particular site.

The Sherwood Project endeavored to employ state-of-the-art technology to solve their typical acid leach waste problems. The unique features of Sherwood's system are the hypalon liner used in the tailings retention system and their water conservation techniques. Both of these approaches have resulted from the site specific characteristics of minimal ground water, the height of the mill from Lake Roosevelt, and its remote location from natural tailings system liners, such as bentonite clay.

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DISCLAIMER

This paper was prepared while one of the authors (A.A.M.) was at Georgia Institute of Technology, Atlanta, Georgia. Mention of commercial products in this paper does not constitute an endorsement by the State of Washington, Georgia Institute of Technology, U.S. Environmental Protection Agency or Western Nuclear, Inc.

MANAGEMENT OF LOW-LEVEL NATURAL RADIOACTIVITY WASTES OF PHOSPHATE MINING AND PROCESSING*

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Abstract - Redistribution of the uranium-series radionuclides associated with phosphate deposits produces what are, in effect, low-level radioactive wastes. Proposed management of these materials involved (1) restricting by-product uses to applications not causing significant exposures to man; and (2) returning other material to the land with simulation of the natural radioactivity depth profile by covering high activity overburden, tailings, clays, gypsum and other radioactivity-bearing wastes with a lower activity overburden layer. Radon flux models are being developed to aid design of materials placement.

Introduction

Increasing attention is being directed to the potential radiation exposure from naturally-occurring radionuclides at concentrations only an order of magnitude above the average values at the earth's surface. This is leading to a new dimension in radioactive waste management - large quantities of relatively low activity materials, involvement of industries other than the traditional nuclear industries and impingement on established commerce.

One of these natural radioactivity sources is the phosphate industry.[+] Naturally-occurring uranium series radioactivity associated with phosphate deposits of marine origin is redistributed in mining, beneficiation, chemical processing, distribution and use of products and by-products and waste management (Gu75, Ro78).

One of the more significant exposure routes to man from this radioactivity source is that of inhalation of airborne radon progeny in structures that (a) are built in a closely-coupled fashion over lands containing elevated near-surface concentrations of ^{226}Ra or (b) incorporate radium-bearing building materials (US75). Consequently, this paper will concentrate on the fate of radium and on measures that limit radon exhalation.

Historically, there has been little concern and no regulatory constraints on the potential radioactive wastes from the phosphate industry. Recently, however, recommendations have appeared calling for licensing, restriction of the use of certain by-products, and increased monitoring and regulation to control natural radioactivity from the phosphate and other industries (Boo77, Boh78). Then in late 1978, proposed hazardous waste regulations of the U.S. Environmental Protection Agency designated overburden and slimes from phosphate surface mining, gypsum from wet-process phosphoric acid production and slag from elemental phosphorus production as hazardous solid wastes (US78).

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+The scope of this paper will be limited to the Florida phosphate industry.

The Radioactivity Source

In the typical undisturbed, pre-mining profile, ^{226}Ra concentrations are on the order of 0.5 pCi/g at the surface, increase gradually with depth through the 1 to 20 m overburden and reach values on the order of 40 pCi/g near and in the ore or "matrix" (Ro78).* In mixed overburden that has been cast aside to expose the matrix, concentrations are typically an order of magnitude higher than in the original surface soil but local concentrations may be found ranging up to that of the matrix.

For the purpose of this paper, phosphate operations can be conveniently divided into three categories - mining, wet-process chemical operations and the thermal process. Mining includes overburden removal, excavation and transport of the exposed matrix and beneficiation of the matrix to separate the phosphate rock product from sand and clay. In wet-process chemical operations, phosphate rock is processed to produce phosphoric acid and phosphate products such as fertilizer and animal feed ingredients. In the thermal process, phosphate rock is reduced in an electric furnace to produce elemental phosphorus. The partitioning of radioactivity in phosphate mining and chemical operation has been described previously (Gu75, Ro77, Ro78). The flow of materials and radioactivity in mining and beneficiation, phosphoric acid production and electric furnace operations are summarized in Figures 1-3. The ^{226}Ra concentrations of various fractions are presented in Table 1, column 2. Included in the table but not identified in a figure is "debris" which consists of higher radioactivity spoils on lands mined prior to the adoption of the flotation process (i.e. prior to the 1940's). At that time, only the coarse, pebble product fraction was recovered and all the balance of the input matrix was returned to the land. Also included in the table is an entry for the sediments and scales (gypsum and other impurities) that occur in phosphoric acid reactors, filters, piping and tanks; these contain the highest ^{226}Ra concentrations observed in the industry.

Rough estimates were made of the production of these wastes annually by a nominal facility, annually by the entire Central Florida industry and cumulatively to date. Table 2 presents volumes, quantities and radium activities associated with each of the major waste types and the areas committed to overburden spoils, clay settling ponds, tailings dumps and gypsum piles. While the radionuclide quantities are significant, most occur as tremendous volumes of low concentration materials.

As summarized in Table 3, most of these "waste" materials have an onsite use, have entered into commerce or have potential commercial use. While uses do represent potential routes for radiation exposure to the public, they also factor into the costs associated with management alternatives, and certain uses may even present a positive benefit to society. All of these facets must be considered and balanced in developing a reasonable posture concerning large volume materials of low activity concentrations.

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*Radioactivity concentrations given in this paper are current values for Central Florida. In the North Florida phosphate mining region, surface soil concentrations are comparable but those in matrix and various products are 25 to 50% of those in Central Florida.

General Considerations for Waste Management

In view of the large quantities of relatively low activity materials and considerations such as the need for overburden and tailings as replacement for mined volumes, the potentially recoverable phosphate and uranium resources of dewatered clays, and the superiority of furnace slag to other materials locally available as aggregates, it is neither practical nor prudent to ship these materials to off-site burial grounds as is done with small quantities of "conventional" low level radioactive waste from the nuclear industry and nuclear research. Rather the solution lies in first developing criteria or standards for population exposure from technologically enhanced natural radiation, next designating limits for unrestricted use of materials and land and then pursuing one or both of two options:

1) restricting land use and commercial utilization of materials according to radioactivity parameters, and/or

2) employing *in situ* management of materials in mining, reclamation and disposal where necessary to meet criteria for the desired land use.

Criteria for Materials and Land Use

Establishment of an indoor airborne radon progeny standard is a prerequisite for regulating materials for radioactivity in order to limit exposure by this route. Values ranging from 0.005 WL to 0.05 WL above background have been proposed; hopefully a standard will soon be established (St71, US77, HRS78). Most recently, the hazardous waste regulations propose a maximum addition to background of 0.03 WL (US78).

With an airborne radon progeny standard, models and empirical observations can be used to derive standards in terms of measureable lands and materials parameters. The indoor airborne radon progeny concentration is a function of a number of variables including the radon exhalation rate or flux at the inputing surface. Flux, in turn, is a function of the radon generating capacity, thickness and radon transport characteristics of the underlying radium-bearing material and of transmission characteristics of any cover (such as a covering over a wall or a floor over fill and soil). Thus for both building materials and under-floor soil and fill, the radon-generating capacity of the materials is a fundamental parameter. This radon generating capacity can be expressed as the "emanating radium concentration", C^{e}_{Ra}, which can be defined:

$$C^{e}_{Ra} = C_{Ra} \cdot E,$$

where C_{Ra} is the materials radium concentration which is also the radon production rate per unit mass, and

E is the "emanating power" or "emanating fraction", i.e., the fraction of the produced radon that is released to void space.

Radioactivity limits for building materials, land and fill, adopted for the purpose of limiting exposure by the airborne radon route, should be based on C^{e}_{Ra} or jointly on its two components, C_{Ra} and E. Values of E and C^{e}_{Ra} for various phosphate waste and by-product materials are listed in Table 1, columns 3 and 4.

The relationship between average indoor radon progeny concentrations in existing structures and measureable land radiological parameters has been quite variable in field studies in Central Florida. In University of Florida studies, the soil radioactivity concentration predicting an indoor radon progeny concentration of 0.03 WL in conventionally-constructed slab-on-grade structures was on the order of 6 to 19 pCi/g for various data-fitting models. Based on studies in Central Florida, the proposed EPA hazardous waste regulations specify a ^{226}Ra concentration limit of 5 pCi/g for land and building materials

If the nominal emanating fraction of typical Florida soils is taken to be about 0.20, the 5 pCi/g standard corresponds to a C_{Ra}^{e} value of about 1 pCi/g. If this concept is extended to other materials, absolute radium concentrations corresponding to this emanating radium value can be calculated as indicated in Table 1, column 5.

Some of the Table 1 materials meet the 1 pCi/g emanating radium criterion and can be used as construction materials or as lands or fill for residential construction without incorporating any special measures for limiting radon progeny exposure. On the other hand, the radiological characteristics of land reclamation materials are highly variable and individual cases would have to be evaluated. Under the proposed criteria, use as building material would be contraindicated for some of the materials such as by-product gypsum and some parcels of land would require special restrictions on the type of construction. The status of slag will be uncertain until the wide disparity in reported emanating fraction values can be resolved.

Materials Management

Since some of the materials do not meet the proposed criteria for unrestricted use, some form of management in place would be indicated. This consists of either mixing higher and lower radioactivity materials to reduce the concentration or selective placement in mining, disposal, reclamation and construction site preparation to more nearly simulate the natural radioactivity concentration profile with the higher activity materials under a low radioactivity cover.

Mathematical modeling of radon flux can be used as an aid in planning materials management schemes to be tested in the field. The expected flux at the surface of an emanating layer can be calculated from the finite single layer model, equation A-1 in Table 4. For most of the materials of concern here, the thickness approaches an essentially infinite value after several meters and the maximum flux can be calculated from the simpler infinite single layer model, equation A-2 in the table. If the values of all the other parameters can be anticipated, this equation reduces to:

$$J_{\infty} = K\, C_{Ra},$$

where K is a constant for the specified material and conditions.

Values of K for some conditions expected in Florida have been computed as shown in Table 1, column 6.

For the indoor radon progeny inhalation exposure pathway, limiting conditions are based on the projected indoor radon progeny levels in future

structures built over the site of concern. In the field studies, the soil surface radon flux corresponding to 0.034 WL (0.03 WL above an average background of 0.004 WL) for existing structures of conventional slab-on-grade construction ranged from 4 to 19 $pCi/m^2 \cdot s$, depending upon the model used to fit the data. A conservative approach would limit flux to 4 or 5 $pCi/m^2 \cdot s$.

Covering a lower layer of more active, radon source materials with an upper, attenuating layer of available lower activity material can be represented by the bi-layer model, equation B in Table 4. For the bi-layer situation, any required reduction from the flux predicted for the uncovered case can be expressed as an "apparent transmission" (acutally the net effect of the attenuation of radon originating in the lower layer and the contribution of radon from the cover material itself):

$$T = J_2/J_d, \text{ or, in the maximum case, } T = J_2/J_\infty.$$

The number of combinations expressing the range of expected conditions can be reduced by presenting the transmission as a function of the ratio of the soil radium concentrations in the two layers and constructing families of attenuation curves for pairs of materials. Figure 4 is a set of curves for high activity overburden covered by low activity upper layer overburden.

As an example, consider a thick layer of 11.7 pCi/g high activity overburden at 10% moisture. The infinite single layer model predicts a flux of 7.4 $pCi/m^2 \cdot s$. Using a design objective of 4 $pCi/m^2 \cdot s$ this requires a layering with T = 0.54. Using 2.3 pCi/g overburden as cover, the activity ratio is 0.2; from the curve the required cover thickness is 0.7 m. This is an attainable condition. Similar types of analysis can be performed for placement of materials produced in mining or for covering chemical plant waste and by-products.

The models suggest that placing the higher activity materials under reasonable thicknesses of available low activity cover (upper layer overburden) will achieve radon fluxes acceptable for unrestricted use. This can be achieved directly in mining if it is feasible to separate the overburden and place the higher activity lower layer (with its leached zone and adventitious matrix) and other waste materials in the bottom of mine cuts with a covering of upper layer overburden. Incidently, such selective placement also reduces possible radionuclide uptake by plants and minimizes the potential for contamination of near-surface ground water and run-off.

The modeling calculations are being extended to various concentrations of other materials and simulations of materials placement in test columns are currently in progress. The ultimate test of these methods will be the extent to which they are verified in future field operations.

Summary and Conclusions

The naturally occurring uranium series radioactivity appearing in phosphate mining spoils and the waste and by-products of beneficiation and chemical plant operations is for the most part contained in large volumes of relatively low radioactivity concentration. The solution to management of materials and lands with radioactivity levels that do not meet unrestricted use criteria consists of 1) restricting land and materials use and/or 2) selective *in situ* placement of higher radioactivity materials in a manner that meets criteria for the desired use.

Inhalation exposure to airborne radon progeny originiating in building materials and in lands used for construction purposes constitutes one of the more significant exposure pathways and this paper concentrated on limitation of exposure by this route. The definitive approach to this problem awaits an adopted indoor airborne radon progeny standard. The development of criteria for materials and lands is further complicated by the observed high variability in the relationship between airborne radon progeny concentrations and radiological characteristics of lands.

The emanating radium concentration is a more meaningful single parameter for evaluating materials than the absolute radium concentration. A method for estimating cover thickness requirements was presented.

Concepts have been formulated and models are showing directions to proceed. However, empirical observations show large variability in the various parameters needed to predict radiation exposure and design mitigative measures. Much work still needs to be done to measure basic parameters of the materials in question, fine tune mathematical models and perform testing and validations.

A complete approach involves evaluation of other nuclides of the uranium series and other exposure pathways including suspension of airborne particulates, uptake by crops from the root zone of the soil with subsequent tranmission through the agricultural food chain and transfer to surface and ground water.

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TABLE 1. Some Radon-generating Properties of Florida Phosphate-related Materials
Numerical Values are for Central Florida

Material	^{226}Ra (C_{Ra}) pCi/g a)	Emanating Fraction (E) b)	Emanating Radium (C^e_{Ra}) pCi/g	C_{Ra} for C^e_{Ra} = 1 pCi/g	J_∞/C_{Ra} g/m^2·s c)
A. MINING AND BENEFICIATION					
Overburden					
Range through profile	0.2-46	--	--	--	--
Leached zone	9 (2-46)	0.16 d)	1.4 (0.2-7.4)	6.3	0.63
Mixed overburden lands	5 (1-35)	0.16	0.8 (0.2-5.6)	6.3	0.63
Clays	28 (14-52)	0.20	5.6 (2.8-10)	5.0	0.24
Sand Tailings	5 (2-12)	0.10	0.5 (0.2-1.2)	10.0	0.45
Debris Lands	10 (3-32)	0.12	1.2 (0.4-2.8)	8.3	0.73
B. WET PROCESS PHOSPHORIC ACID PRODUCTION					
Gypsum	32 (21-65)	0.12	3.8 (2.5-7.8)	8.3	--
Misc. Sediment, Scale	(80-380)	--	--	--	--
C. THERMAL PROCESS					
Slag	69 (45-101)	0.05 (0.0002-0.004) e)	3.5 (2.2-5.1) (0.01-0.4)	20.0 (200-400)	--

a) For comparison, some ^{226}Ra concentrations in products are:
Mining and beneficiation: matrix (ore), 38 (18-84); pebble (product), 57 (45-97); rock concentrate (product), 35 (24-50);
Wet process phosphoric acid and fertilizer production: phosphoric acid, <1 pCi/g; ammoniated phosphates, 4 (1-12); triple superphosphates, 20 (15-32).

b) Standard deviations of the distributions of mean emanation power for multiple samples of the various materials were: overburden and debris 50-60%; clays and tailings 13-15%; gypsum 30%; and slag 60%, expressed as percent of the category mean.

c) Clays calculated at 33% moisture, all others calculated at 10% moisture.

d) Assumed to be similar to mixed overburden.

e) Analysis by second laboratory; discrepancy unexplained.

-- indicates not measured or not calculated.

TABLE 2. Estimated Inventory of Florida Phosphate Solid Wastes and By-products*

Material (Form, ^{226}Ra)	Parameter	Facility Annual	Florida Annual	Florida Cumulative
	A. MINING AND BENEFICIATION			
Overburden: (spoils, 5 pCi/g)	Volume, m^3	9.9×10^6	1.3×10^8	2.5×10^9
	Mass, tons (metric)	1.4×10^7	1.9×10^8	3.6×10^9
	Area, hectares	80	1000	26,000
	^{226}Ra activity, Ci	70	940	18,000
Clays:+ (settling ponds, 28 pCi/g)	Volume, m^3	2.0×10^6	2.5×10^7	2.8×10^8
	Mass, tons (metric)	2.8×10^6	3.6×10^7	4.0×10^8
	Area, hectares	50	660	17,000
	^{226}Ra activity, Ci	80	1000	11,000
Sand Tailings: (spoils, 5 pCi/g)	Volume, m^3	2.3×10^6	3.0×10^7	1.1×10^9
	Mass, tons (metric)	4.2×10^6	5.5×10^7	6.0×10^8
	Area, hectares	30	350	9000
	^{226}Ra activity, Ci	20	270	3000
	B. WET PROCESS PHOSPHORIC ACID PRODUCTION			
Gypsum: (gypsum piles, 5pCi/g)	Volume, m^3	2.3×10^6	2.1×10^7	1.3×10^8
	Mass, tons (metric)	2.5×10^6	2.3×10^7	1.4×10^8
	Area, hectares	10	100	730
	^{226}Ra activity, Ci	80	730	4400
Sediments & Scales: (80-380 pCi/g)		No Quantity Estimate		
	C. THERMAL PROCESS			
Slag: (slag pit, 60 pCi/g)	Volume, m^3	1.1×10^5	2.1×10^5	6.7×10^6
	Mass, tons (metric)	1.4×10^5	2.7×10^5	8.6×10^6
	^{226}Ra activity, Ci	10	17	550

*Quantities and areas were back-calculated from published mining and chemical plant production data and represent only rough estimates.

+Clays volume and mass reported on dry weight basis. Settled clays actually contains 15% solids.

TABLE 3. Current and Potential Values and Uses of Phosphate Industry Wastes

A. MINING AND BENEFICIATION
Overburden - Replacement for volume removed in mining; land reclaimed for development, agriculture, recreation.
Clays - Replacement volume (30% of removed matrix); potential phosphate and uranium resource if recovery technology developed; potential soil conditioner to improve fertility, exchange capacity, moisture retention; potential commercial uses.
Sand Tailings - Replacement volume (40% of removed matrix); fill materials.
B. WET PROCESS PHOSPHORIC ACID PRODUCTION
Gypsum - Soil conditioner and Ca source (Li78); building materials use outside U.S. (Fi78); potential chemical raw material.
Sediments and Scales - No known use or values.
C. THERMAL PROCESS
Slag - Crushed and to commercial use as aggregate; or as light weight aggregate (expanded form).

Table 4. General Models for Soil Surface Radon Flux*

Definitions		
Description	Symbol	Units
Radon flux at the soil-air interface	J	pCi/m^2-s
Radium concentration in the soil media (grams, dry weight)	C_{Ra}	pCi/g
Bulk density, volume per unit of dry weight	ρ	g/cm^3
Emanating power, amount of radon released to void space per unit of radium (or radon)	E	pCi/pCi
Decay constant for radon-222	λ	s^{-1}
Porosity	p	-
Media diffusion coefficient $D = 5.0 \times 10^{-2} e^{-0.1}$ for values of D from 5×10^{-2} to 5×10^{-3}	D	cm^2/s
Depth of media	d	cm

A. Single Layer Models

1. Finite Single Layer

$$J_d = 10^4 \cdot C_{Ra} \rho E \sqrt{\frac{\lambda D}{p}} \cdot \tanh d\sqrt{\frac{\lambda p}{D}}$$

2. Infinite Single Layer - special case, very thick active layer ($d \to \infty$)

$$J_\infty = 10^4 C_{Ra} \rho E \sqrt{\frac{\lambda D}{p}}$$

B. Bilayer Model

$$J_2 = -10^4 m_2 D_2 (2A_2 e^w + k_2)$$

where

$$A_2 = \frac{(k_2 - k_1) - k_2 k_4 e^{w-y}}{(k_4 - 2)e^y + k_4 e^{2w-y}}$$

$w = m_2 d_2$ $\quad m_1 = \sqrt{\lambda p_1 / D_1}$ $\quad k_1 = C_{Ra1} \rho_1 E_1 / p_1$

$z = m_1 d_1$ $\quad m_2 = \sqrt{\lambda p_2 / D_2}$ $\quad k_2 = C_{Ra2} \rho_2 E_2 / p_2$

$y = m_2 d_1$ $\quad k_4 = \frac{m_2 D_2}{m_1 D_1 \tanh z} + 1$

* From Bolch et al.(Bo78).

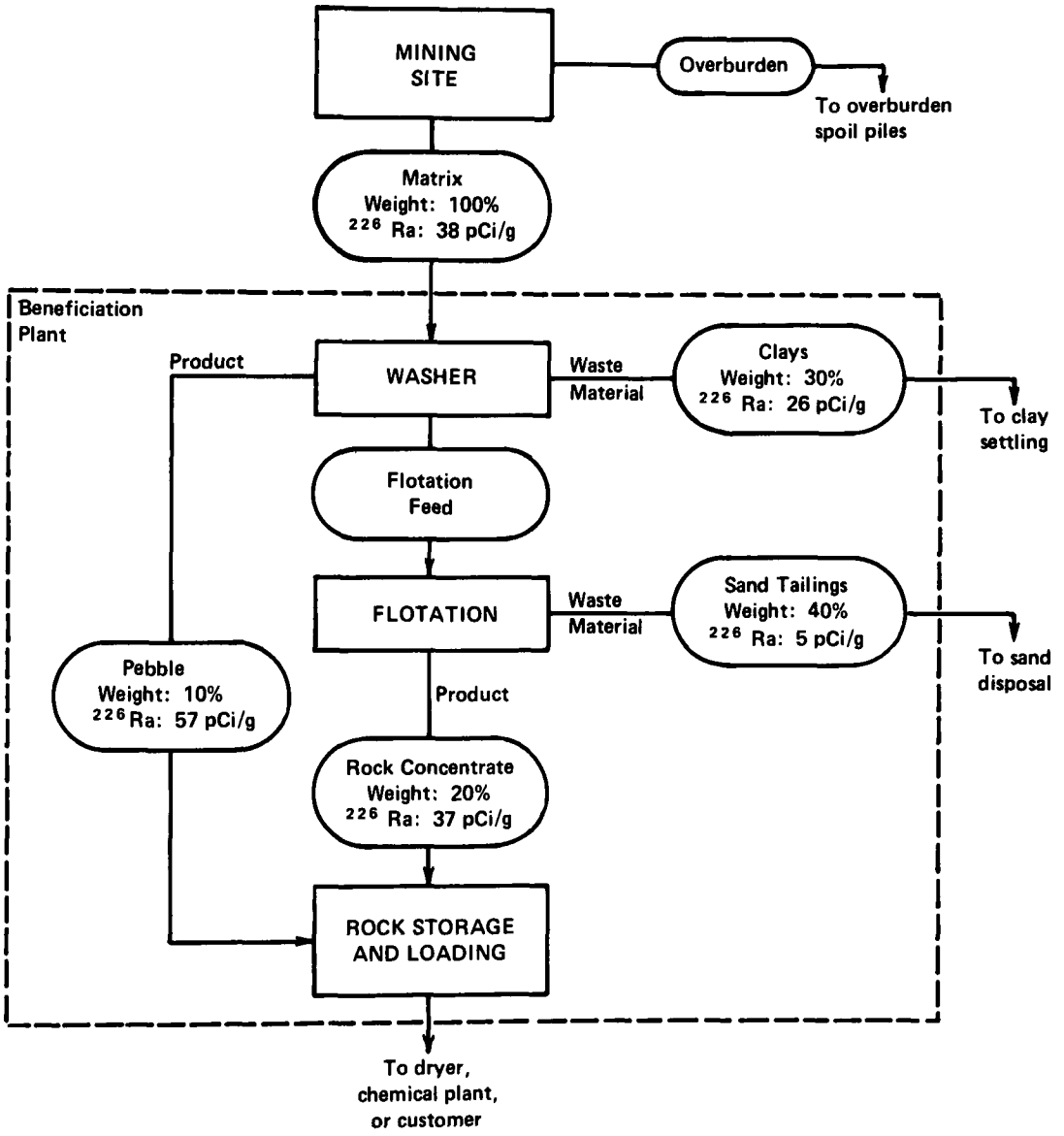

Fig. 1. Simplified Flow Diagram for Phosphate Mining and Beneficiation. Radium Concentrations are for Central Florida.

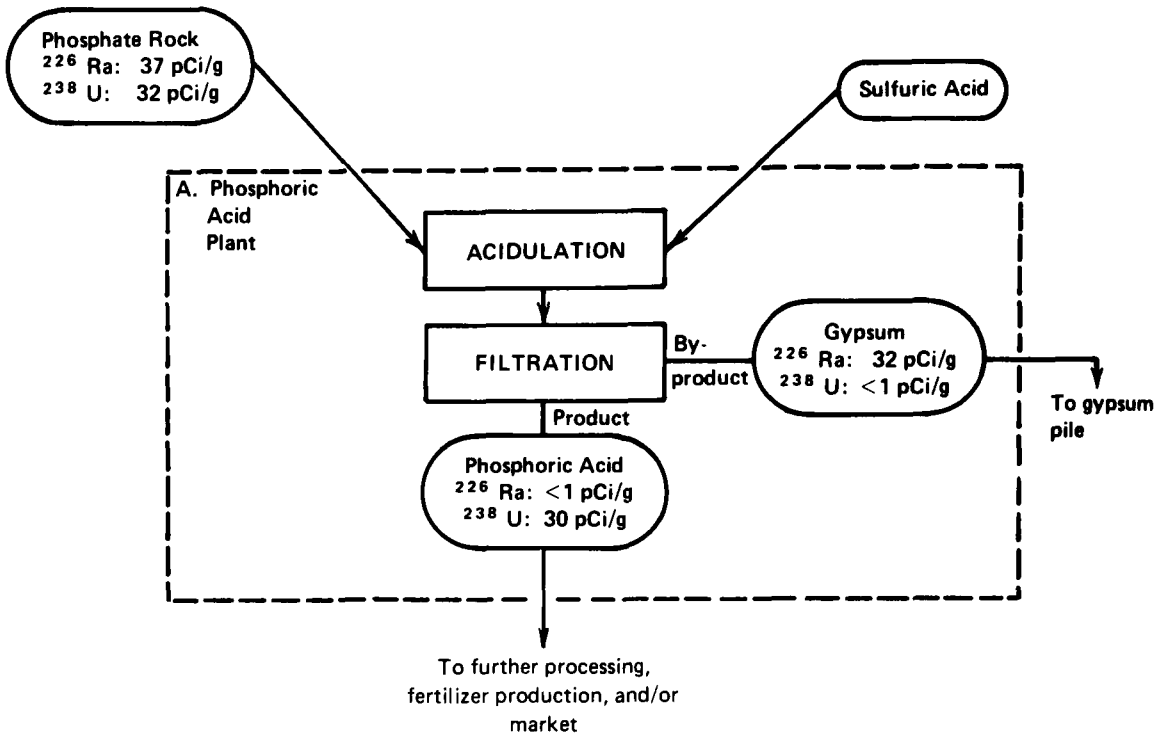

Fig. 2. Simplified Flow Chart for Wet Process Phosphoric Acid Production. Radionuclide Concentrations are for Central Florida.

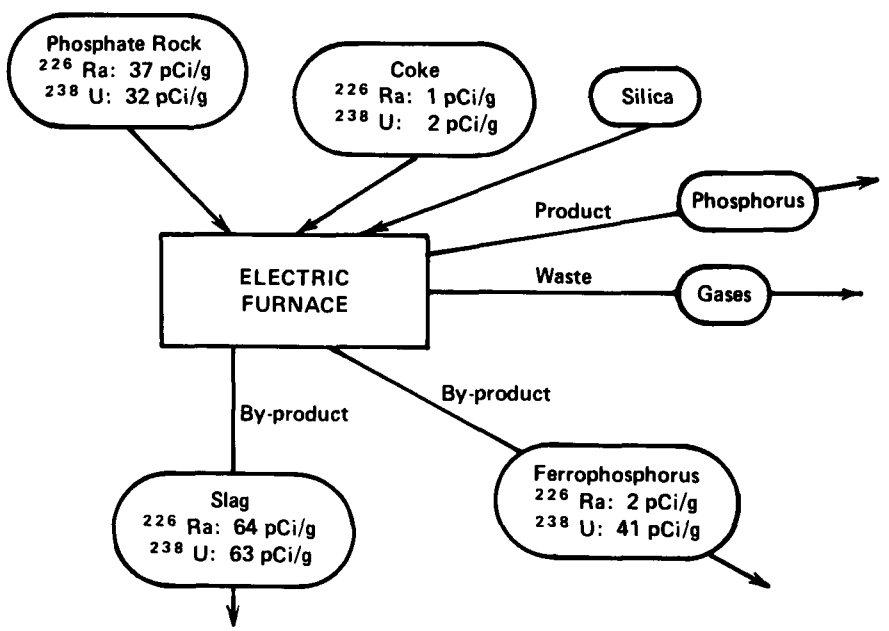

Fig. 3. Simplified Flow Diagram for Thermal Process Elemental Phosphorus Production. Radionuclide Concentrations are for Central Florida.

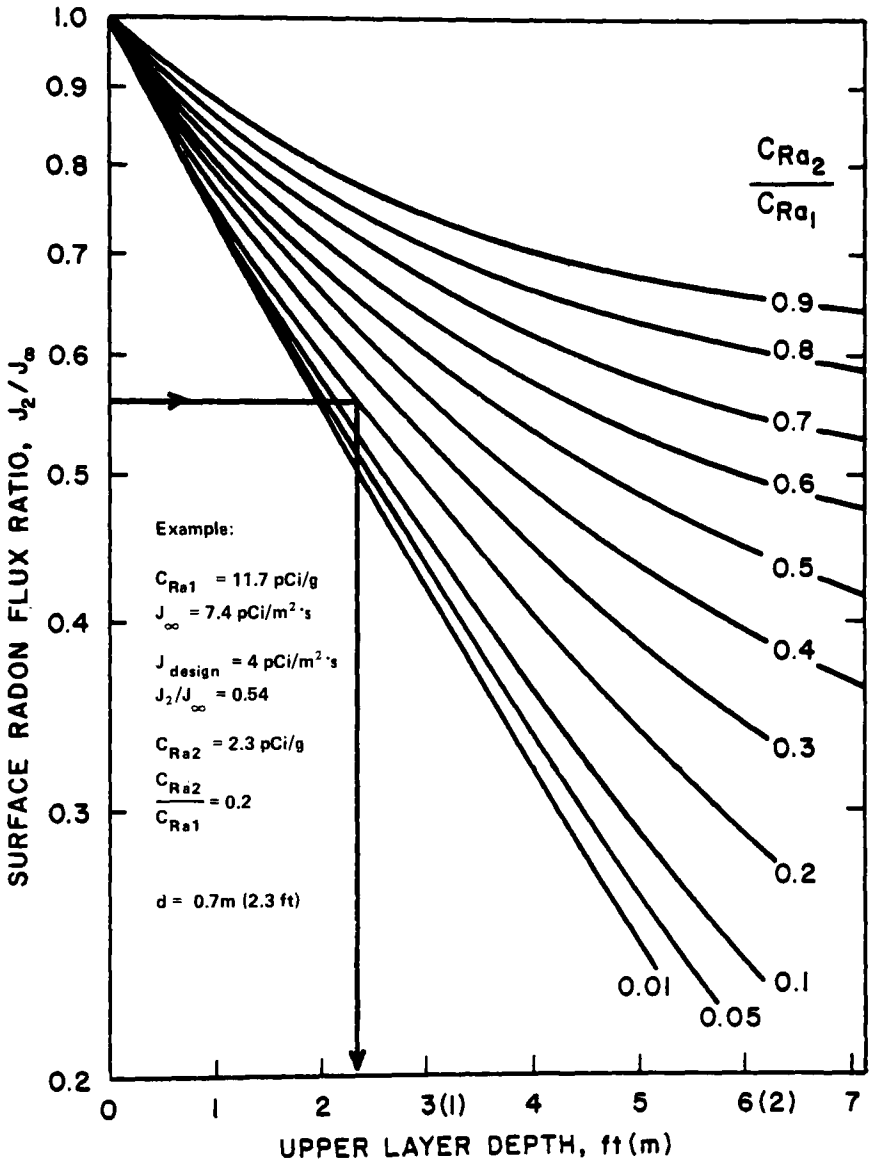

Fig. 4. Typical Design Curve for Attenuation of Radon Flux. Low Activity Overburden over High Activity Overburden.

ACTIVITY MEASUREMENTS AT A WASTE VOLUME REDUCTION FACILITY

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Abstract

The monitoring program for Ontario Hydro's radioactive waste management site will be described, several aspects of which will be discussed in detail. The program at this facility includes categorization, volume reduction processing, and storage of solid radioactive wastes from nuclear generating stations of the CANDU type. At the present time, two types of volume reduction process are in operation - incineration and compaction. Following categorization and processing, wastes are stored in in-ground concrete trenches or tile-holes, or in above-ground quadricells.

The monitoring program is divided into three areas: public safety, worker safety, and structural integrity. Development projects with respect to the monitoring program have been undertaken to achieve activity accounting for the total waste management program. In particular, a field measurement for the radioactivity content of radioactive ash containers and compacted waste drums.

General

The radioactive waste management site currently operated by Ontario Hydro is located at the Bruce Nuclear Power Development on the east shoreline of Lake Huron about 140 miles north-west of Toronto. The site consists of waste processing facilities, namely an incinerator and compactor, and various types of storage structures for the different categories of solid radioactive wastes. The actual site layout was developed in the following stages:

- Stage 1 - concrete trench and tile hole storage structures.
- Stage 2 - waste volume reduction facility.
- Stage 3 - concrete trench and tile hole storage structures.
- Stage 4 - concrete quadricell storage structures for bulk resin storage.

The waste volume reduction facility or W.V.R.F. includes a starved-air type radioactive waste incinerator (Trecan, Canada) and a compactor (Stock Equip. Co.) for processing the large volume, low specific activity wastes prior to storage.

The various concrete storage structures have the following major design criteria:

- minimum operational life of 50 years.
- radioactive contents retrievable.
- isolated from ground water.
- adequate shielding.

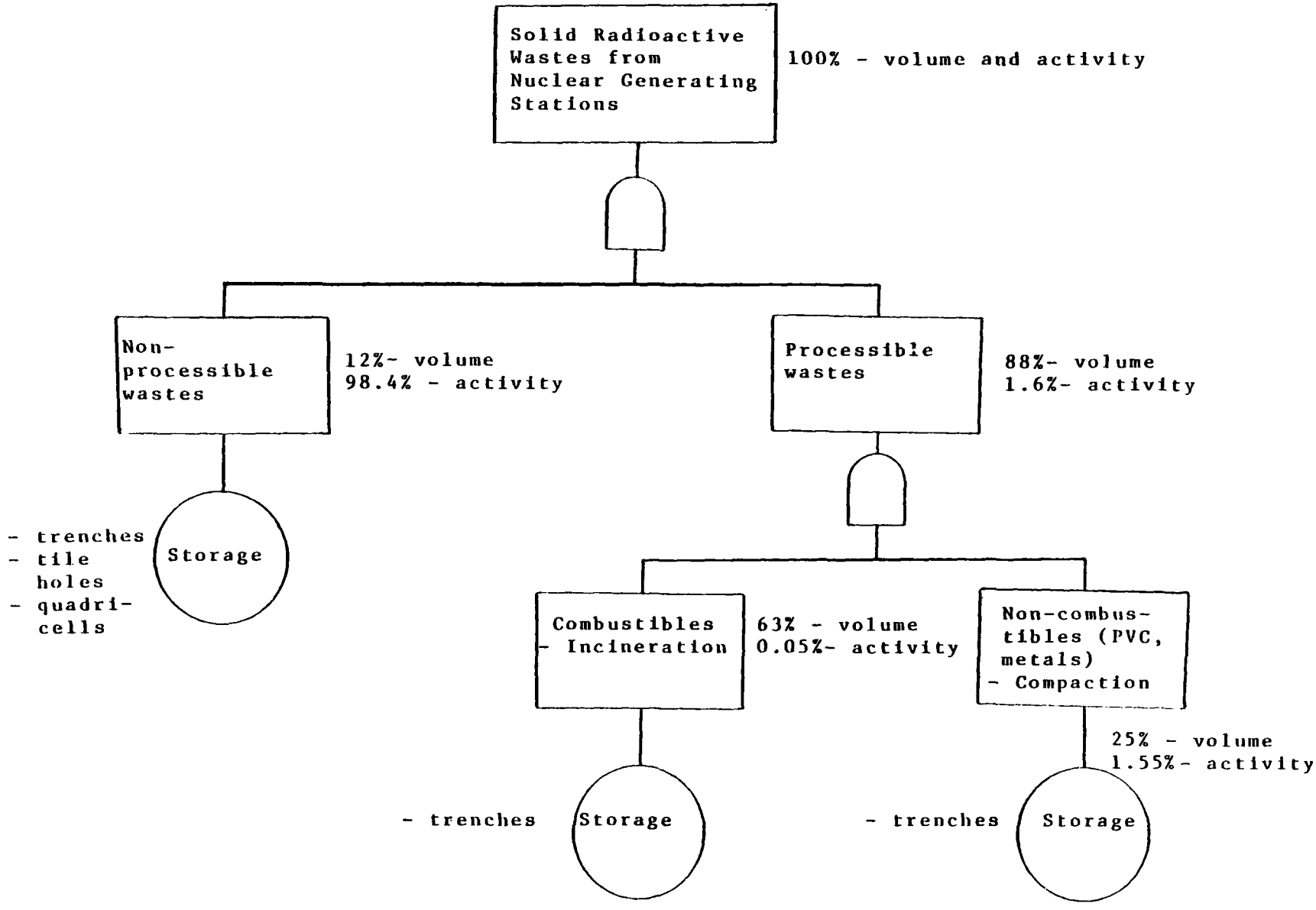

FIG 1 - RADIOACTIVE WASTE MANAGEMENT PROCESS

The radioactive wastes currently processed and stored at this site originate from the nuclear generating stations at B.N.P.D. and Pickering.

Waste categories

The solid radioactive wastes originating from the nuclear generating stations are classified into the following categories prior to any volume reduction processes:

Solid Waste Category	Beta-Gamma Activity (Nominal Ci/m^3 [1])
Type 1	< 0.1
Type 2	0.1 to 100
Type 3	> 100

For the purpose of segregating the waste packages, direct gamma dose rate measurements are used with Type 1 being equivalent to a contact dose rate of less than 200 m rad/h, Type 2 less than 15 rad/h and Type 3 in excess of 15 rad/h.

Type 1 wastes are typically the general dry garbage with low levels of radioactive contamination such as waste paper, used protective clothing, metal and plastic scrap materials. The type 2 and 3 radioactive wastes are usually associated with the reactor system and are typically discarded filters, ion-exchange columns, bulk ion-exchange resin and reactor components. In general, the Type 1 and 2 level wastes are stored in the concrete trenches and the Type 3 in the concrete tile holes and quadricells.

Waste management process

The overall radioactive waste management process for the site is illustrated in Fig. 1. The Type 2 and 3 wastes are normally regarded as non-processible and transferred directly to the storage structures. This accounts for about 12% of the wastes by volume and greater than 98% of the wastes by activity content. The majority of the Type 1 wastes are processed in the W.V.R.F. prior to storage.

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(1) Nominal curie - that quantity of beta gamma emitting radioactive material which emits 3.7×10^{10} photons of 0.8 MeV per second.

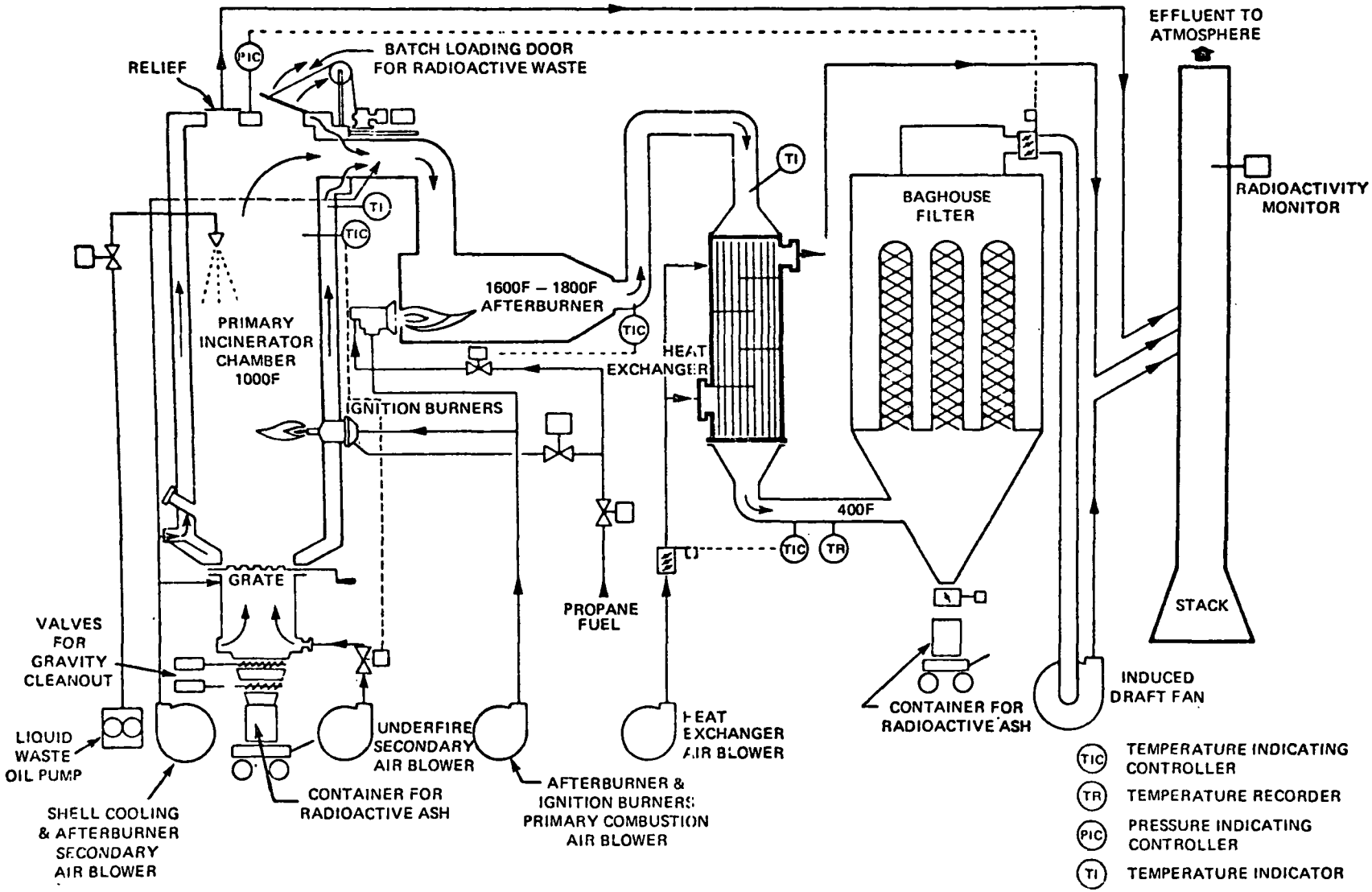

FIGURE 2 INCINERATOR SCHEMATIC

These wastes are further segregated prior to processing into combustible wastes for the incinerator and non-combustible wastes, such as P.V.C. materials and metals, for the compactor. The combustible wastes account for about 63% of the total by volume and less than 0.05% by activity content. The non-combustible or compacted wastes account for 25% of the total by volume and about 1.55% by activity content.

The current solid waste volume processed at the site is about 3000 m^3 per year with an estimated total activity content of about 500 Ci. The incinerator provides an overall volume reduction of about 25 and the compactor about 2.

Monitoring program - general

The monitoring program for the storage site and waste volume reduction facility (W.V.R.F.) has been designed to monitor performance in the areas of public safety, worker safety, and structural integrity of storage facilities. The associated activities are broken down as follows:

Public safety - radioactivity of liquid effluent
- radioactivity of airborne effluent
- radiation fields at fence line
- radioactivity in ground water

Worker Safety - hazard identification
- working environment surveys
- internal and external dosimetry
- worker knowledge

Structural Integrity - record of stored activity
- radioactivity in ground water

Activity measurements - radioactive incineration

Figure 2 is a schematic of the radioactive waste incinerator. Routine radioactivity measurements have been made on samples of the incinerator ash, heat exchanger deposits, baghouse ash, and stack monitor filters. The stack monitor, supplied by Radeco, provides continuous on-line monitoring for operational control. The particulate filter, charcoal cartridge, and tritium bubbler samples are analyzed in the site Health Physics laboratory for compliance monitoring purposes.

The filters and ash samples are analyzed with a computer - based gamma spectrometry system. The stack bubbler samples are analyzed for tritium using an automatic liquid scintillation counter.

The incinerator is loaded with 20 to 22 m^3 of low level radioactive waste in 0.05 m^3 plastic bags. The waste averages about 1% of the activity limit defined for the type 1 waste category (excluding tritium).

TABLE 1: LOW LEVEL RAD-WASTE INCINERATION - "TYPICAL"

	PARTICULATE ACTIVITY (µCi/burn)	(%)	I-131 (µCi/burn)	H-3 (µCi/burn)
Incinerator Ash	19000	90	--	--
Baghouse Ash	2000	9.6	--	--
Stack Effluent	75	0.4	50	5×10^6

TABLE 2: RADIONUCLIDE DISTRIBUTION FOR INCINERATION

RADIONUCLIDE	INCINERATION ASH (%)	BAGHOUSE ASH (%)	STACK EFFLUENT (%)
Ce-144	15.1	< 1	1.7
Sb-124	N.D.	10	N.D.
Cr-51	4.5	N.D.	N.D.
Ru-103	3.0	< 1	4.3
Cs-134	1.8	10	11.7
Ru-106	5.2	7	< 1
Cs-137	5.6	49	54.8
Zr-95	9.1	< 1	< 1
Nb-95	16.0	< 1	N.D.
Zn-65	5.0	20	25.1
Co-60	14.8	3	< 1
La-140	15.2	N.D.	N.D.
Others (Gd-153, Ce-141, Sn-113, Sb-125, Mn-54, Fe-59, Ba-140)	4.7	1	2.4
	100%	100%	100%

The process reduces the waste to about 0.5 m^3 of ash of which 90% remains in the incinerator primary chamber and 10% is trapped in the baghouse filters.

The distribution of the waste activity following a typical burn is shown in table 1. 100% of the radioiodine and tritium is driven off in the stack emissions. About 0.4% of the total particulate activity is observed in the stack emissions. As in the case of the radioiodine and tritium, the final distribution of radionuclides in the particulate activity depends on the physical and chemical properties of the elements involved (see table 2). For instance, cesium, which has relatively low melting point (29°C) and boiling point (670°C), constitutes a large fraction of the activity found in the ash in the baghouse (operates at 200°C) and in the effluent gases. In the other extreme, chromium has a melting point of 1900°C and a boiling point of 2200°C and is not detectable in the baghouse ash or in the effluent gases, but remains in the primary chamber.

Field measurements

In order to facilitate the routine measurement and accounting of curie content of processed wastes under field conditions, relationships have been determined for field measurements using gamma dose-rate survey meters.

Ash from the incinerator is discharged into 2.5 m^3 steel cubic containers prior to storage. Equation (1) is the relationship determined for this type of container.

$$\text{Curie content } (\mu Ci/m^3) = 10^4 \times \text{mrem/h (@ 1 ft)} \quad ---(1)$$

Non-combustibles are compacted into 0.2 m^3 drums for which the relationship in equation (2) was determined.

$$\text{Curie content } (\mu Ci/m^3) = 2 \times \text{mrem/h (contact)} \quad ---(2)$$

Future programs

No attempt has been made to date to analyze for Carbon-14 or alpha-emitters. This work is currently in the development stage.

A COMPARISON OF ALTERNATIVES FOR LOW-LEVEL RADIOACTIVE WASTE DISPOSAL

By

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Ford, Bacon & Davis Utah, Inc.

ABSTRACT

A comparative analysis of alternatives for disposal of low-level radioactive wastes has been performed for the U.S. NRC. A systematic evaluation of all possible disposal mechanisms identifying options most viable for further analysis is presented. Generic reference disposal facility concepts for each viable alternative are evaluated to provide a consistent, meaningful comparison based on technological, economic, and sociopolitical factors. The results of the comparative analysis are presented in a convenient matrix format to facilitate intercomparisons and to promote understanding of the complexities of the tradeoffs involved in selecting waste disposal options.

The concepts judged to be the most viable alternatives to the current practice in this country of disposal of low-level wastes by shallow land burial include improvements to shallow land burial, ocean disposal, intermediate depth burial (10-15 m deep), disposal in natural or mined cavities, and disposal in exposed or covered structures. Representative waste disposal facility concepts for each of these alternatives were analyzed as the basis of the evaluation, using reference waste volumes and facility lifetimes.

I. INTRODUCTION

This paper describes an evaluation of alternative methods for the disposal of low-level radioactive wastes performed for the U.S. Nuclear Regulatory Commission (NRC) by Ford, Bacon & Davis Utah, Inc. (FBDU). Alternative methods for waste disposal must be evaluated to assure that safer or more effective techniques are not overlooked.

II. SCREENING OF ALTERNATIVES

A comprehensive review of all possible methods which have been identified or proposed for low-level radioactive waste disposal was performed, based on a systematic methodology for identifying disposal options ensuring that no viable choices have been overlooked. This first study objective included systematically identifying, cataloging and describing possible low-level waste disposal alternatives.

The range of all possible low-level radioactive waste disposal alternatives was divided into categories to provide a systematic means for identifying all options. The categories were subdivided to arrive at specific disposal methods, shown in Figure 1. After analysis and review, those alternatives warranting further evaluation were selected. The selected alternatives are the basis for this report, and include the base case of typical shallow land burial, improvements to present practices, deeper burial, disposal in mined cavities, disposal in engineered structures, and disposal in the ocean.

To assure completeness of the initial listing and adequacy of the selection of viable alternatives, a panel of technically competent individuals of recognized waste management expertise was consulted for review and guidance. A formal report of the results from this phase of the study has been published.[1]

The most viable alternatives selected are compared with current solid low-level waste disposal by shallow land burial using a rigorous and detailed analysis. The results of this effort are presented in a convenient matrix format to facilitate their use in decisions pertaining to selection of viable alternatives in national low-level waste management programs.

For the generic alternatives selected for further evaluation, several additional factors require specification to allow a meaningful comparative analysis. These factors include the location, size, and type of disposal facility designed for each alternative method. For this study generic Eastern U.S. and Western U.S. locations and possible ocean disposal sites were assumed to obtain transportation factors, a volume of waste to be accommodated was given, and the disposal facilities were conceptually designed to reasonably accommodate and contain the wastes.

Based on the reference disposal facilities for each alternative method studied, technical, sociopolitical and economic factors were evaluated as the basis of a comparative analysis. Values for the parameters required for performing the evaluations are specified; but it should be understood that the performance of any actual waste disposal facility will depend on the conditions that exist at the real site, which may vary from those assumed for this study. By changing some of the site- and facility- specific factors, some of the conclusions of this comparative analysis could be changed. However, a uniform, consistent approach has been taken for all alternatives evaluated in this report, which provides a rational basis for waste management decisions and allows appropriate evaluation of the tradeoffs involved in disposal option selection.

III. EVALUATIONS

Reference disposal facilities for each alternative have been selected as a base case in performing comparative analyses

of the different types of disposal options. These reference facilities are all based on disposal capacity for a constant volume of waste having a given radioactivity inventory. The conceptual designs of the facilities represent an estimation of the types of design criteria that may be required in the future for waste disposal sites. The results from disposal of waste in the reference facility provide a uniform basis for comparison of the alternatives with both costs and effects being appropriate indices for the comparison. Other approaches for the comparative analysis coudl be selected. For instance, either the costs for construction or the resultant effects from waste disposal at a reference facility could be held constant, the designs varied, and the comparison based on non-fixed variables. The approach taken in this study, however, provides a consistent basis for comparing alternatives, and is appropriate for the preconceptual design stage of development of the reference disposal facilities.

The volume of waste to be disposed of was assumed to be 630,000 m3, which roughly corresponds to the expected output of 1,000 typical light-water reactors for one year (1,000 Reference Reactor Years (RRY) of low-level waste).[2] This volume of waste would correspond to roughly 800,000 megawatt-years of electricity production (MW(e)-yr). Wastes from non-fuel cycle sources will also be accommodated in the reference facilities. The generic inventory has been adjusted to account for wastes from both sources. The generic reference facilities were assumed to handle this volume of waste in a twenty-year operating period.

The environmental effects are subdivided into non-radiological and radiological impacts. The non-radiological effects include impacts on construction and waste management workers. Radiological impacts include direct radiation exposures to workers and the public along the transportation route and in the area of the disposal facility.

The exposure pathways calculated for the various alternatives may not necessarily be an exhaustive listing of all the possible mechanisms for human exposure at each site. However, they do provide a consistent basis for comparison of the alternatives and are representative of the most important types of impact that would be expected from implementation of waste disposal operations. Obviously, by changing the conceptual design, the radiological impacts would be changed, as would be the associated costs. Prior to implementation of any of the alternatives, it is anticipated that formal cost-benefit analyses and trade-offs would be performed to optimize the results from the selected option. The analyses reported in this study are useful, however, in providing perspective and guidance for selection among the various alternatives, and are presented for that purpose.

Institutional control over disposal sites is assumed to be maintained for 150 years after operations cease. Any future site reclamation efforts would occur after that time period.

The assessment of sociopolitical implications is somewhat subjective. However, available published research and information on the topic[3,4] has been used for guidance. Additionally, many of the social acceptance issues depend on adequate demonstration that the technological problems have been appropriately solved. Assuming that the disposal alternatives meet the minimum constraints of being technically sound, the sociopolitical issues hinge mainly on requirements for governmental agreement and control, as is the case with ocean disposal in international waters. These issues are considered in the weighting factors used in the comparative analysis.

IV. COMPARATIVE ANALYSES

After completion of the technical, sociopolitical and cost evaluations for each concept, the major factors relating to each of these areas are quantified. Some of the items important to the comparison of alternatives, however, can be quantified only by subjectively ranking one concept against another. Other items (such as cost, for example) are quantified during the course of the evaluation. Care must therefore be exercised to assure that the different alternatives are uniformly assessed.

Once the important evaluation factors and parameters have been quantified for each of the disposal alternatives, the factors and the alternatives are jointly displayed in a convenient matrix format. One additional factor in the comparison is an estimate of the relative importance (weighting) of the evaluation factors and parameters used in the comparison. For instance, an estimate of how heavily costs should be considered in relation to sociopolitical issues allows comparison of the different concepts.

The weighting factors are somewhat subjective. The weighting factors used in this report were determined from a survey of knowledgeable persons serving as advisors on this study. The use of these weighting factors allows quantitative comparison of the alternatives, but does not mean that others may not be more appropriate for different circumstances.

The comparative analysis is useful in demonstrating that the selection of best or optimum alternatives for low-level radioactive waste disposal involves complex tradeoffs among several factors. It also shows that there is more than one method of safely handling low-level radioactive wastes. However, going from the generic generalized concepts studied in this project to specific designs at real sites will lead to important differences in the values of the evaluation parameters. This comparative analysis should, therefore, be used with care for guidance in selecting optimum choices, and not be directly applied to specific, actual disposal operations.

The radiological effects from waste disposal are summarized in Table 1. Increased transportation exposures to the western site account for the differences between eastern and western

locations. Improving the base case would reduce exposures by about 20%. Disposal in mined cavities would eliminate about 90% of the potential doses. Disposal in structures would increase potential exposures by about 20% because of greater availability of the waste to future reclamation activities. Ocean disposal would eliminate all but about 20% of the potential exposures from the base case. In general, it can be noted that reclamation events contribute the most significant portion of the potential doses. Elimination of the possibility of reclamation after only 150 years beyond disposal would effectively reduce the consequences of waste disposal activities.

Non-radiological effects are based on accident statistics from comparable industries,[5] and are summarized in Table 2. Crew sizes are estimated, and transportation risks are calculated based on shipping distances.[6] Because of the larger construction crew sizes, the structural disposal concepts provide approximately twice as large an impact as the base case.

Cost estimates for the preconceptual design facilities are presented in Table 3. Capital and operating costs are differentiated, and profit, escallation and financing charges included. Monitoring and surveillance activities for 150 years have been included in the operating budgets.

Table 4 contains the overall comparative analysis of all alternatives. Weighting factors are included for the various evaluation parameters. Selection of other weighting factors could change some of the conclusions of this analysis. However, a consistent approach has been taken and some insight into the tradeoffs involved obtained.

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TABLE 1

SUMMARY OF RADIOLOGICAL EFFECT FOR ALTERNATIVES

Alternative	Reclamation Events			Short Term Events			Total Cummulative Effect	Normalize Effect
	Inhalation/Direct	Gamma/Food		Transportation/	Well Water Consumption/	Single Container Accidents		
Shallow-Land Burial-Eastern Site	110	340	625	9.5	80	200	1364	1.0
Shallow-Land Burial-Western Site	110	340	625	33	80	200	1388	1.0
Improved Burial-Eastern Site	99	310	560	9.5	1.5	150	1130	0.8
Improved Burial-Western Site	99	310	560	33	1.5	150	1154	0.8
Deeper Burial-Eastern Site	0	0	0	9.5	1.5	150	161	0.1
Deeper Burial-Western Site	0	0	0	33	1.5	150	184	0.1
Abandoned Mine-Eastern Site	0	0	0	14	0	100	114	0.1
Abandoned Mine-Western Site	0	0	0	38	0	100	138	0.1
New Horizontal Shaft Mine-Eastern Site	0	0	0	14	0	100	114	0.1
New Horizontal Shaft Mine-Western Site	0	0	0	38	0	100	138	0.1
New Vertical Shaft Mine-Eastern Site	0	0	0	14	0	100	114	0.1
New Vertical Shaft Mine-Western Site	0	0	0	38	0	100	138	0.1
Above Grade Structure-Eastern Site	220	670	625	9.5	1.5	100	1626	1.2
Above Grade Structure-Western Site	220	670	625	33	1.5	100	1650	1.2
Buried Structure-Eastern Site	220	670	625	9.5	1.5	100	1626	1.2
Buried Structure-Western Site	220	670	625	33	1.5	100	1650	1.2
Direct Ocean Dumping	0	0	1	38	0	200	239	0.2
Ocean Projectile Disposal	0	0	0	38	0	200	238	0.2

TABLE 2

SUMMARY OF NON-RADIOLOGICAL EFFECTS FOR ALTERNATIVES

Alternatives	Transportation			Construction			Operations			Cummulative Effects
	Total Train Car Miles	Total Injuries	Total Fatalities	Crew Size (My)	Total Injuries	Total Fatalities	Crew Size (My)	Total Injuries	Total Fatalities	
Shallow-Land Burial-Eastern Site	528,000	0.20	0.002	20	0.61	0.01	265	8.06	0.09	9.9
Shallow-Land Burial-Western Site	1,848,000	0.70	0.01	17	0.52	0.01	265	8.06	0.09	10.4
Improved Burial-Eastern Site	528,000	0.20	0.002	22	0.67	0.01	265	8.06	0.09	10.0
Improved Burial-Western Site	1,848,000	0.70	0.01	20	0.61	0.01	265	8.06	0.09	10.5
Deeper Burial-Eastern Site	528,000	0.20	0.002	31	0.95	0.01	265	8.06	0.09	10.2
Deeper Burial-Western Site	1,848,000	0.70	0.01	28	0.85	0.01	265	8.06	0.09	10.7
Abandoned Mine-Eastern Site	792,000	0.30	0.003	12	0.63	0.01	265	8.06	0.09	10.0
Abandoned Mine-Western Site	2,112,000	0.80	0.01	12	0.63	0.01	265	8.06	0.09	10.6
New Horizontal Shaft Mine-Eastern Site	792,000	0.30	0.003	54	2.84	0.06	265	8.06	0.09	12.7
New Horizontal Shaft Mine-Western Site	2,112,000	0.80	0.01	54	2.84	0.06	265	8.06	0.09	13.3
New Vertical Shaft Mine-Eastern Site	792,000	0.30	0.003	62	3.26	0.07	265	8.06	0.09	13.3
New Vertical Shaft Mine-Western Site	2,112,000	0.80	0.01	62	3.26	0.07	265	8.06	0.09	13.8
Above Grade Structure-Eastern Site	528,000	0.20	0.002	330	10.06	0.12	265	8.06	0.09	20.4
Above Grade Structure-Western Site	1,848,000	0.70	0.01	326	9.94	0.12	265	8.06	0.09	20.9
Buried Structure-Eastern Site	528,000	0.20	0.002	363	11.07	0.13	265	8.06	0.09	21.6
Buried Structure-Western Site	1,848,000	0.70	0.01	352	10.73	0.12	265	8.06	0.09	21.7
Direct Ocean Dumping	2,112,000	0.80	0.01	NA	NA	NA	300	9.15	0.11	11.2
Ocean Projectile Disposal	2,112,000	0.80	0.01	NA	NA	NA	300	9.15	0.11	11.2

TABLE 3

COST ESTIMATE SUMMARY FOR ALTERNATIVES ($MILLIONS)

ALTERNATIVE	Capital Costs	Operating Costs	Summary Contingency	Financing Escallation & Profit	Total Facility Costs	Transportation Costs	Total Costs	Total Unit Costs ($/m^3)	Normalization Costs
1. Shallow-Land Burial-Eastern Site	12.26	23.65	10.77	35.22	81.90	67.74	149.64	238	1.0
2. Shallow-Land Burial-Western Site	9.42	23.65	9.92	29.44	72.44	237.10	309.54	491	2.1
3. Improved Burial-Eastern Site	13.81	23.65	11.24	38.29	86.99	67.74	154.73	246	1.0
4. Improved Burial-Western Site	10.98	23.65	10.39	32.58	77.60	237.10	314.70	499	2.1
5. Deeper Burial-Eastern Site	18.70	23.75	12.74	48.18	103.37	67.74	171.11	272	1.1
6. Deeper Burial-Western Site	15.87	23.75	11.89	42.48	93.99	237.10	331.09	525	2.2
7. Abandoned Mine-Eastern Site	7.06	24.47	9.46	22.55	63.54	101.60	165.14	262	1.1
8. Abandoned Mine-Western Site	7.06	24.47	9.46	22.55	63.54	270.96	334.50	531	2.2
9. New Horizontal Shaft Mine -Eastern Site	29.57	24.47	16.21	70.40	140.65	101.60	242.25	384	1.6
10. New Horizontal Shaft Mine -Western Site	29.57	24.47	16.21	70.40	140.65	270.96	411.61	653	2.7
11. New Vertical Shaft Mine -Eastern Site	33.73	24.47	17.46	78.78	154.44	101.60	256.04	406	1.7
2. New Verticle Shaft Mine -Western Site	33.73	24.47	17.46	78.78	154.44	270.96	425.40	675	2.8
3. Above Grade Structure -Eastern Site	178.33	24.63	60.89	121.31	385.16	67.74	452.90	719	3.0
4. Above Grade Structure -Western Site	175.60	24.63	60.07	115.81	376.11	237.10	613.21	973	4.1
5. Buried Structure -Eastern Site	192.13	24.63	65.03	128.52	410.31	67.74	478.05	759	3.2
6. Buried Structure -Western Site	189.40	24.63	64.21	123.02	401.26	237.10	638.36	1013	4.3
7. Direct Ocean Dumping	3.65	73.94	23.28	40.04	140.91	101.60	242.51	385	1.6
8. Ocean Projectile Disposal	3.65	484.67	146.50	221.58	856.40	101.60	958.00	1521	6.4

TABLE 4 WEIGHTED COMPARATIVE ANALYSIS FOR ALTERNATIVES

	Compatibility with Waste	Site Selection	Safeguards	Env. Effects	Availability of Technique	Inst. Control	Public Accept.	Individual Costs	Industry Costs	Weighted Comparison
Weighting Factor	0.075	0.117	0.065	0.111	0.095	0.108	0.161	0.143	0.125	1.00
Alternative										
Shallow-Land Burial-Eastern Site	0.075	0.117	0.065	0.111	0.095	0.108	0.161	0.143	0.125	1.00
Shallow-Land Burial-Western Site	0.075	0.117	0.065	0.111	0.095	0.108	0.161	0.300	0.263	1.29
Improved Burial-Eastern Site	0.075	0.117	0.065	0.100	0.095	0.108	0.144	0.143	0.125	0.97
Improved Burial-Western Site	0.075	0.117	0.065	0.111	0.095	0.108	0.144	0.300	0.263	1.28
Deeper Burial-Eastern Site	0.075	0.140	0.059	0.056	0.105	0.119	0.128	0.157	0.138	0.98
Deeper Burial-Western Site	0.075	0.129	0.059	0.067	0.105	0.119	0.128	0.315	0.275	1.27
Abandoned Mine-Eastern Site	0.075	0.176	0.052	0.056	0.114	0.130	0.128	0.157	0.138	1.03
Abandoned Mine-Western Site	0.075	0.164	0.052	0.067	0.114	0.130	0.128	0.315	0.275	1.32
New Horizontal Shaft Mine-Eastern Site	0.075	0.164	0.052	0.078	0.124	0.130	0.112	0.229	0.200	1.16
New Horizontal Shaft Mine-Western Site	0.075	0.152	0.052	0.078	0.124	0.130	0.112	0.386	0.338	1.45
New Vertical Shaft Mine-Eastern Site	0.075	0.152	0.052	0.078	0.124	0.130	0.112	0.243	0.213	1.18
New Vertical Shaft Mine-Western Site	0.075	0.140	0.052	0.089	0.124	0.130	0.112	0.400	0.350	1.47
Above Grade Structure-Eastern Site	0.075	0.105	0.078	0.178	0.105	0.130	0.176	0.429	0.375	1.65
Above Grade Structure-Western Site	0.075	0.094	0.078	0.178	0.105	0.130	0.176	0.586	0.513	1.94
Buried Structure-Eastern Site	0.075	0.105	0.072	0.189	0.105	0.119	0.144	0.458	0.400	1.67
Buried Structure-Western Site	0.075	0.094	0.072	0.189	0.105	0.119	0.144	0.615	0.538	1.95
Direct Ocean Dumping	0.075	0.140	0.033	0.078	0.095	0.130	0.224	0.229	0.200	1.20
Ocean Projectile Disposal	0.075	0.140	0.033	0.078	0.124	0.130	0.192	0.915	0.800	2.49

CATEGORIZATION OF ALTERNATIVES FOR DISPOSAL OF LOW-LEVEL RADIOACTIVE WASTES

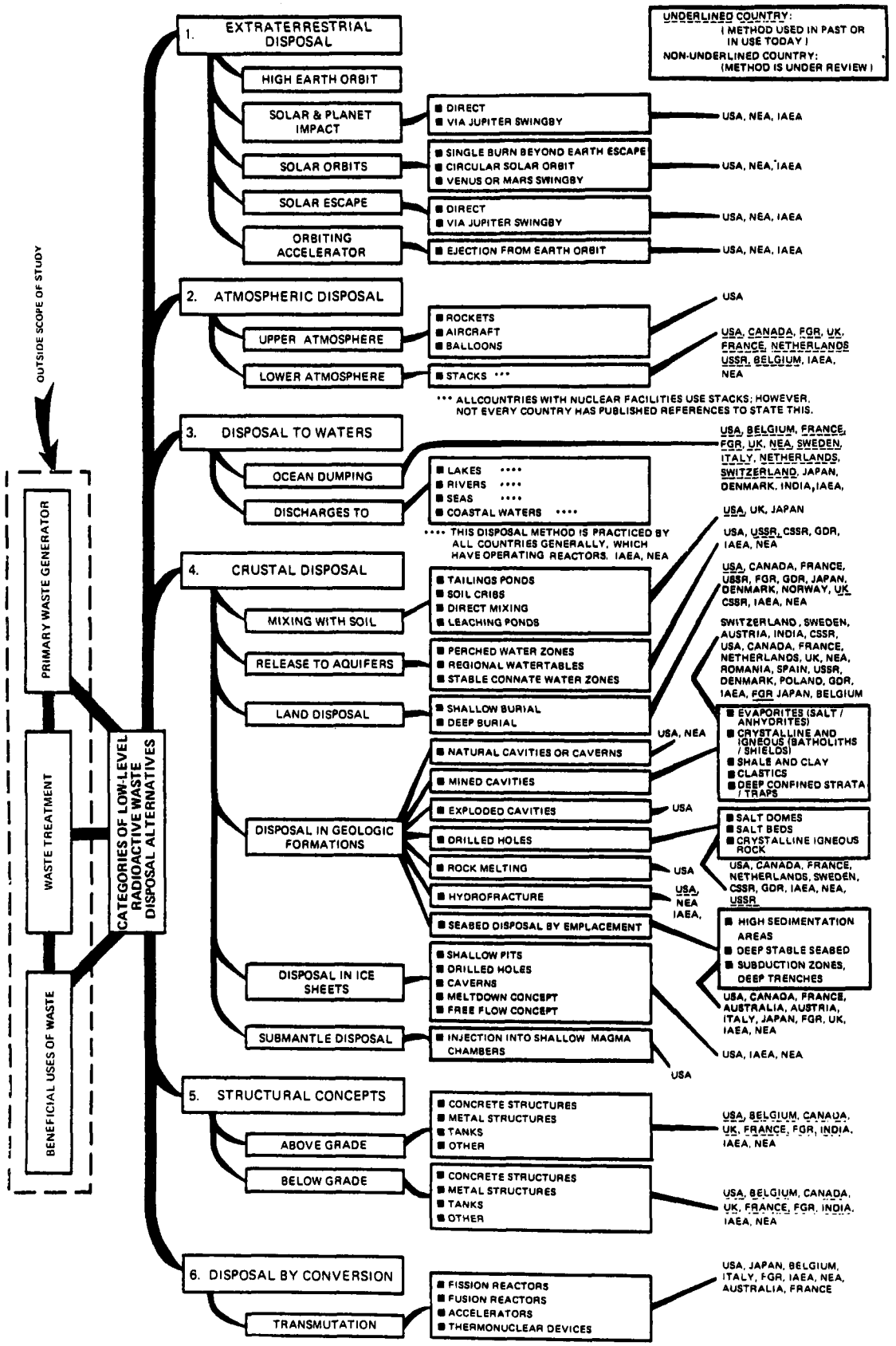

FIGURE 1.

Ford Bacon & Davis

SESSION D

REGULATORY ASPECTS

Session Chairperson

C. R. Price

State of Virginia

DISPOSAL CLASSIFICATION OF LOW-LEVEL RADIOACTIVE WASTE

J.A. Adam
Nuclear Regulatory Commission

I will be presenting a system for classifying radioactive waste that has been developed for the Nuclear Regulatory Commission (NRC) by Ford, Bacon & Davis Utah under the lead of Dr. Vern Rogers. It is a system for classifying waste based on the minimum requirements for safe disposal. It was first given public attention in NUREG-0456, "A Classification System for Radioactive Waste Disposal - What Waste Goes Where?", June 1978. That document reported a study in progress and did not represent the fully developed system. We have made considerable progress since the publication of NUREG-0456 and today I will present the complete classification system.

I would like to emphasize that the system which I am going to present is not being proposed by NRC at this time. The system will be one of several alternatives that will be considered during the development of a waste classification regulation.

Early in the development of the classification system, it was decided that the system should be based on the requirements for safe disposal. We recognized that there are varying degrees of toxicity of radioactive wastes and various degrees of confinement that can be achieved by disposal.

Three generic disposal options were considered:

- disposal as ordinary (non-radioactive) waste,
- containment in a licensed disposal facility, and
- isolation (i.e., deep geologic repository).

For the containment option, variations based on the accessibility of the waste to man, periods of administrative control or restricted land use, and presence or absence of groundwater were also considered.

The steps in developing the complete system are:

- develop study guidelines (what is safe?),
- determine the pathways to man,
- formulate the classes to be considered,
- determine disposal concentration guides, and finally
- classify the waste from the major waste stream or sources.

The first step, defining safe disposal, was accomplished by formulating a set of study guidelines. The study guidelines dealt with exposures to few and many individuals, "as low as reasonably achievable" (ALARA), positive net benefit, and periods of administrative control or restricted land use.

Two of the study guidelines turned out to be dominant; restrictions on the exposure of the maximally exposed individuals and the period of administrative control. We used 500 mrem/year, whole body or organ, as the ex-

posure limitation for the critical individuals and 150 years as the period of administrative control.

Because the analyses that were used in developing the classification system are consequence rather than risk analyses, the use of a 500 mrem per year exposure restriction for the maximally exposed individuals is considered to be conservative. We chose to use consequence rather than risk analyses because of the difficulties in assigning probabilities to the numerous events that can take place over the many years involved (risk equals probability times consequence). This is particularly true when dealing with generic disposal rather than waste and site specific disposal.

As an analogy, consider the risk from the head-on collision of two cars. The risk is the consequence of a collision times the probability of a collision. For any given speed the risk is greater on a curvy, two-lane road than on a divided interstate highway. Consequence alone is not an adequate measure of risk. However, if there is an identifiable speed at which head-on collisions would not result in any serious injury, then we could assert that driving at that speed is safe without any statement of risk. Likewise, if we can show that the consequence from waste disposal will not result in any serious injury, we can assert that such disposal is safe without statements of risk. In this context, 500 mrem per year to the maximally exposed individuals is used to define safe disposal and not to estimate the degree of risk. Further, just as the proper design of roads reduces the risk of driving, we envision that reasonable caution by future man and other waste management criteria such as careful site selection will reduce the risks far below the levels of the postulated consequences.

More simply, we can insure that wastes will be safely disposed of by insuring that they are sent to disposal facilities capable of their safe disposal. This can be done by classifying the wastes according to minimum requirements for safe disposal as determined by consequence analysis. Other criteria and standards for other functions, such as waste preparation and site selection, can be used to reduce the risks from disposal to as low as reasonably achievable. This conservative approach was adopted because of the difficulties in specifying detailed site characteristics and probabilities for a non-site specific waste classification system.

To identify an appropriate period of restricted land use a series of calculations were made. These calculations were for the change in potential exposures with various periods of decay for typical mixtures of isotopes. The results of the various calculations were fairly consistent. During the first hundred years or so, the very short lived isotopes control the level of potential exposures. After a few hundreds of years the long-lived isotopes, in particular carbon-14, control. From about one hundred years to a few hundreds of years cesium-137 controls. Typically, the decay between one hundred and one hundred and fifty years resulted in a factor of 2.5 reduction in potential exposures. The decay between one hundred and fifty years and two hundred years resulted in a reduction of 1.5. We chose one hundred and fifty years as an appropriate period for restricted land use. It should be noted that the restricted land use we postulate is passive in nature as compared to active administrative control. Restricted land use does not mean complete restriction. Use of the land, such as for use as an airfield, may be permissible or even desirable.

The pathways that were investigated were of two types; onsite reclaimer events and offsite migration. The onsite reclaimer events include:

- a worker or reclaimer digging into the waste and inhaling resuspended waste,
- consumption of food grown onsite,
- consumption of well water obtained from on or near the site, and
- direct exposure of workers or reclaimers.

Routine and accidental airborne releases during disposal operations were also considered. Because such airborne release are not directly related to the type of disposal but rather the care taken in preparing and handling the waste, airborne releases are not included in the classification system.

These generic pathways are used as surrogates for families of more specific pathway scenarios. For example, a reclaimer may be digging on a site which has been released from restricted use to dig a basement, emplace fence posts, lay sewers, or any number of reasons, including unforeseeable future activities of man.

The offsite migration events include:

- ground water migration of the waste to surface waters (rivers) and subsequent consumption; and
- surface erosion of the waste to surface waters.

Three conclusions regarding offsite migration were reached. First, offsite migrations were seldom controlling pathways. Second, groundwater migration was not affected by periods of administrative control or restricted land use. And third, offsite migration rates and consequences are strongly dependent on site specific parameters and could not be adequately addressed in generic analyses. Because offsite migration is seldom controlling and is highly site specific, it is not included in the waste classification system. However, the burden of substantiating that offsite migration would not be controlling at a given site remains with the classification study.

There are several observations regarding the onsite reclaimer events. The consequences of the reclaimer (but not worker) inhalation, direct exposure and food consumption events can be reduced through the use of a period of restricted land use, or eliminated if the wastes are buried sufficiently deep. Because the waste concentrations in well water can be close to maximum near the site, but outside of a restricted area, a period of administrative control of restricted land use may not reduce the consequences of well water consumption. Siting of disposal facilities in dry regions can eliminate the well water event for as long as the site remains dry. However, it can not be asserted, at least generically, that a site will remain dry indefinitely. Having observed that most of the decay of typical waste will occur in the first few hundred years it was concluded that assuming a dry site would remain dry for at least a few hundred years (150 years) was appropriate.

The classification system postulates five classes of terrestrial disposal. (This does not mean that NRC will consider all five classes when developing their waste classification regulations.)

CLASS A:

Isolation or the best that can be reasonably accomplished. Because isolation is intended to be the best that can be achieved, no limitations are placed on Class A waste by the classification system.

CLASS B:

Man does not have ready access to the waste (i.e., deep burial) and the site is intially dry. The well water pathway is assumed to exist after one hundred and fifty years.

CLASS C:

Man does not have ready access to the waste (i.e., deep burial) but there is ground water present. The well water pathway is assumed to exist prior to any decay of the waste.

CLASS D:

Man does have access to the waste (i.e., shallow burial) after a period of administrative control or restricted land use (150 years). The well water pathway exists prior to decay of the waste. The reclaimer digging into the waste, food consumption, and direct exposure pathways are assumed to exist after 150 years of waste decay.

CLASS E:

Man does have access to the waste from the time of disposal (i.e., sanitary landfill). The assumed pathways are; worker inhalation of resuspended waste, food and well water consumption, and direct exposure.

For class D and E, variations could have been postulated based on the absence of well water for a dry site. It was concluded that these variations would not be significant for most wastes because of the dominance of the inhalation and food pathways. However, for some isotopes, such as tritium, disposal in a dry site would offer a significant advantage.

The table shows examples of calculated allowable average concentrations for the various classes of waste. The controlling pathway is also noted.

CLASS	AVERAGE ALLOWABLE CONCENTRATIONS (Ci/m^3)			
	H-3	Sr-90	I-129	Pu-239
A	-	-	-	-
B	430,000[1]	38[1]	0.3[1]	90[1]
C	94[1]	2.4[1]	0.3[1]	90[1]
D	94[1]	0.02[2]	0.3[1]	0.1[3]
E	0.05[2]	0.00023[2]	0.14[1]	0.0003[3]

[1]well water consumption [2]food consumption [3]dust inhalation

Comparison of the allowable concentration for tritium (H-3) among the 5 classes illustrates the importance of decay and the choice of disposal options for short lived isotopes. The comparison also shows that when man has ready access to the waste and there is no decay, food rather than well water consumption can be the controlling pathway for tritium.

The allowable concentration for I-129 in class E waste is lower than for the other classes even though well water is assumed to be the controlling pathway for all the classes. This is the result of not allowing for dilution of the waste by surrounding soil for class E waste. Class E assumes uncontrolled (unlicensed) disposal without qualification as to dilution of the waste, depth of burial or thickness of cover. These may be unduly conservative assumptions.

If the development of the classification system were to stop with the calculation of average allowable concentrations the system would be unworkable for at least two reasons. First, for many important isotopes, the allowable levels are not readily detected. Second, besides allowable concentrations, there needs to be consideration of surface contamination, activation of structural materials, hot spots, and other physical properties of the waste which affects disposal. Many of these considerations were discussed in NUREG-0456.

To make the classification system workable, we are proposing to classify radioactive waste according to the source of generation. Classification by source would involve an analysis of existing and newly acquired information on the isotopic mix, statistical variances, activity levels, and physical properties of the waste from the major waste streams. As an example, the results of such an analysis could be: the waste from the clean-up of the primary cooling system of a PWR (except certain resins) are Class D, medical waste (with a few exceptions) are Class D, and the waste from the cleanup of the secondary cooling system of the PWR under normal operating conditions are Class E. For classification by source to be successful the methodology must be workable for both the waste generator and the disposal facility operator and at the same time must provide a means to insure safe disposal.

The radioactive waste disposal classification system which has been described is being proposed to NRC as a practical first step for assuring the safe disposal of radioactive waste. This system and alternative classification systems, along with the development of criteria and standards for waste disposal and other regulatory requirements, will be considered by NRC in the development of the waste classification regulation.

A HISTORICAL REVIEW OF FEDERAL/STATE ROLES IN REGULATING COMMERCIAL LOW LEVEL RADIOACTIVE WASTE BURIAL GROUNDS

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Abstract

The subject of disposal of low level radioactive waste has generated considerable interest among State and Federal agencies, State and Federal legislatures, the public and the industry in the past three to four years. The background and regulatory processes which have been followed since 1961 in regulating the low level waste burial grounds provide a useful point of reference for consideration of the currently evolving changes in this sector of the nuclear industry. The background is discussed in this paper as well as the type of activities conducted at the burial sites and the processes followed in licensing and regulating the existing commercial burial sites. The paper also discusses the possible future roles of Federal and State governments in regulating such sites.

Introduction

Almost every sector of the nuclear industry generates high interest amongst the public and the industry and in government agencies from time to time. In the Washington area almost every day some article related to nuclear matters appears in the daily papers. Subjects range from nuclear power plants, transportation of radioactive materials, high and low level waste, uranium mills to smoke detectors. The fact that this entire symposium is devoted to low level radioactive waste management is indicative of the high level of interest in this subject. The subject of waste disposal, particularly high level, but also low level to a significant extent is one of the most intractable subjects which the technical and regulatory experts must deal with. Some would argue, however, that the subject is not so much a question

regarding the technical aspects as it is the political and institutional aspects. It is also sometimes stated that it is not a lack of technical knowledge but a lack of decisions which makes the subject intractable.

The high level of public interest in this subject began to surface in late 1974 when the State of Kentucky issued a report on the results of a special six-month environmental study at the Maxey Flats waste burial ground. The report noted the contribution of radioactivity to the surrounding environs resulting from the operations at the site but concluded that it did not create a public health hazard. This was followed by an NRC report on the same site and a January 1976 report by EPA on the same subject. During early 1976, three other events took place which highlighted the attention the Federal Government was giving to this subject. First, the GAO issued a report on January 12, 1976 on land disposal of radioactive waste. Second, the Conservation, Energy and Natural Resources Subcommittee of the House Committee on Government Operations held hearings on low level waste early in 1976. During FY 1977 authorization hearings for NRC, the subject of regulation of low level sites was discussed in Congress. Congressional interest in this subject has remained high through 1978 including various proposals for organizational and institutional frameworks for addressing the subject at the Federal level. One of the most recent publications is the "Report to the President by the Interagency Review Group on Nuclear Waste Management" (DOE, 78).

Why Are There Commercial Low Level Sites?

Prior to 1960, disposal in designated areas of the Atlantic and Pacific Oceans was the conventional method for the disposal of wastes generated by

commercial users. In June 1960, the Atomic Energy Commission placed a moratorium on issuance of new licenses for sea disposal. The AEC established an interim program in 1960 whereby radioactive wastes generated by licensed users were accepted for burial at the Commission's Oak Ridge National Laboratory in Tennessee and at the National Reactor Testing Station in Idaho though existing licenses for sea disposal were permitted to remain in effect. In September 1962 the first commercial land burial facility located near Beatty, Nevada was licensed and became available for use by the private sector. Other commercial burial grounds were established in Kentucky (1962), New York (1963), Washington (1965), Illinois (1967), and South Carolina (1971). In a press release dated May 28, 1963, the Atomic Energy Commission stated that the Commission's burial grounds would no longer be available to licensees for waste material shipped on or after August 12, 1963. It further stated that "AEC's withdrawal is in line with its policy to foster industrial participation in the atomic energy program." Some of these burial grounds were presumably established because it was felt they might serve to attract other nuclear related operations to the area but this has apparently not been the case.

What is a Low Level Site?

"Low level radioactive waste" has never been formally defined. It is generally considered to be any waste other than high level wastes which are defined in Appendix F of 10 CFR Part 50 of the Commission's regulations. Therefore, low level wastes cover such things as minimally contaminated papers, metal, boxes, etc. and even "suspect" waste, but in fact can also include megacuries of radioactive material such as $^{3}_{H}$, $^{60}_{Co}$, $^{90}_{Sr}$ and others. Nevertheless, it is generally known that the six sites noted previously constitute the commercial low level burial grounds in the United States.

The sites vary in licensed acreage from 20 to 260 acres and are located in both dry areas such as Nevada and Washington to wet areas for the other four sites. Table 1 shows the volume of wastes and the amount of activity that have been buried at each since their initial operation. The physical operations conducted at a burial site are rather straightforward and do not involve complex machinery, mechanical control systems or highly skilled staff. Operations carried out are similar to a sanitary land fill operation with a greater degree of care exerted during trench excavation, trench filling and trench capping. Of course, certain specialized operating procedures must be observed during these operations. However, operations in general are rather modest when compared with the operation of a fuel fabrication plant, uranium mill or a nuclear power plant.

How Are the Sites Licensed and Regulated?

A regulatory requirement from the very beginning was that the land for a disposal site be owned by a State or the Federal Government. All are located on State-owned land except the Washington site which is on Federally-owned land. Regulatory jurisdiction for each of the sites was determined by the type of materials to be buried or handled at the site. If the site is located in an Agreement State* the regulatory authority is exercised by the State unless the quantity of special nuclear material to be handled prior to burial exceeds those quantities specified in 10 CFR 150.11. If quantities exceed that, a license for handling special nuclear material has to be obtained from the NRC.

* A State which has entered into an agreement with the AEC (now NRC) pursuant to Section 274 of the Atomic Energy Act of 1954, as amended, whereby the Commission relinquishes and the State assumes, certain regulatory authority over the use of source material, byproduct material, and small quantities of special nuclear material.

In non-Agreement States, burial grounds were and continue to be, regulated by AEC (NRC). It should be noted, however, that the burial grounds also receive non-agreement material and accelerator produced radioactive material which is not subject to control by NRC. Such material is regulated by the Agreement States and the non-Agreement State of Illinois.

The procedures for licensing sites included a submission of information by the applicant on the geological and hydrological characteristics of the site and the usual information related to qualifications of the applicant, operating procedures, and the applicant's radiation safety program. Although the information on geology and hydrology obtained on the sites at the time they were initially licensed would be considered modest by today's standards, the information obtained was evaluated by appropriate technical specialists in the State and Federal agencies. It should be noted that the National Environmental Policy Act (NEPA) process for Federal agencies was not in effect at the time any sites were originally licensed by AEC.

During the early 1960's it was felt that the disposal of waste in the ground took care of the matter. These sentiments were no doubt expressed with a degree of certainty that they did not deserve. Notwithstanding this, the States involved in all six sites, through various mechanisms, established environmental monitoring programs in the vicinity of the site. Environmental monitoring programs by nature are designed to provide verification that the "system" was performing as expected or if it was not performing as expected one could take such action as necessary to correct it before

the problem became serious. I have noted earlier in this paper that one of the initiating events for the publicity surrounding this subject was a 1974 environmental monitoring report issued by Kentucky. I believe it worth noting that the Kentucky environmental monitoring program did precisely what it was supposed to do, that is, detect a possible problem at a low threshold. Another aspect of the regulation of these sites is that all operations are subject to periodic inspection by the appropriate regulatory agency.

In each case, the States (as site owner or lessor in the case of Washington) established disposal fees which were intended to provide a perpetual care fund when the sites were closed. The funds were not expected to be used for corrective action since major problems in site performance were not expected. It is obvious that the funds accumulated to date are insufficient for major corrective action and may be even insufficient for long term maintenance. There are no uniform national standards that can be used at present regarding the establishment of such funds. At the May 1973 meeting of the National Conference of Radiation Control Program Directors, a task force on radioactive waste management presented a report and certain recommendations. Among other things, it recommended a national and/or regional coordinated approach to the question of establishing disposal sites as may be required, recognizing that proliferation of such sites may not be in the public interest. It also noted that information is required in the areas of criteria and requirements regarding perpetual care of land burial grounds and the legal and financial implications of the State's perpetual care responsibilities.

Some States have expressed the parochial view that they might establish a burial ground just to be used for wastes generated within their own borders.

If this view prevails there could be more than 50 burial grounds in the United States. I think most of us here today would agree that would be highly undesirable. In this regard the Supreme Court ruled in June 1978 that a New Jersey law prohibiting importation of wastes from neighboring States (this case involved non-radiological wastes) violates the Constitution because a State is not free to impede interstate commerce simply because that commerce is in valueless wastes. This should be considered by those who might suggest a burial ground just to handle the wastes generated within its borders. One should also question the economic viability of such an operation, at least in many States. The concept of regional sites to address the problem of adequate distribution of capacity should be pursued vigorously. This obviously will require a high degree of cooperation among several States if it is to be effective.

Thoughts for the Future

A fair question to ask is "Is low level waste burial a viable option?" I leave this to those closer to the subject technically, but would observe that there are still three low level sites operating today. Certainly the technical and regulatory climate is different today than it was in 1962 when the first site was licensed. The most notable factors are NEPA requirements, more detailed information required to evaluate site suitability, and the high level of public interest and involvement. I would judge that the use of solid land burial as a disposal method will continue although with various modifications such as new techniques, waste classification, and volume reduction. I believe that volume reduction is an area where the industry, both waste generators and waste collection and disposal firms, should take the initiative. One never knows when an action taken in some sector of the nuclear industry affects another. For example, the NRC recently received a communication from

the State of South Carolina indicating that rather large volumes (as much as 50,000 ft^3 per month) of waste in the form of spent fuel racks are being received for burial at the Barnwell, South Carolina facility due to reracking to accommodate the increasing quantities of spent fuel stored at nuclear power plants. It would seem that some effort at the waste generating sites to reduce the volume of these wastes would be desirable.

What does the future hold for Federal/State roles in this area of regulation of low level waste disposal? Although the NRC's task force report on this subject (NRC77) recommended NRC reassert regulatory authority over the burial grounds, the Commission did not adopt this as formal policy. It felt there were a number of unresolved questions on the matter and there was no compelling need to make a final decision at that time (December 1977) since the States were adequately protecting the public health and safety. The Commission also believed it was more urgent to proceed with other elements of the low level waste program including development of regulations, standards, guides and a study of alternatives to shallow land burial. The report of the task force for the review of nuclear waste management (DOE, 78a) commonly referred to as the "Deutch Report" noted various possibilities for addressing the question of regulatory control of the burial sites. At the July 1978 meeting of the National Governor's Association (NGA) they stated that "The Governors believe that long-term program plans for low-level radioactive waste which continue to permit private operation and 'agreement-state' regulation of low-level waste burial grounds on a cooperative basis with Federal authorities, wherever this is both preferred and practicable, should be finalized as expeditiously as possible." The subject was also considered by the Interagency Review Group established by President Carter to develop

recommendations for the management of nuclear waste (DOE, 78). The IRG recommended the concept of State "consultation and concurrence" for high-level waste facility siting plans and that States have the option to retain management control of existing commercial low level waste sites or to transfer such control to the Federal Government. They further recommended that the Department of Energy assume responsibility for developing and coordinating the national plan for low level waste. All of this leads to the conclusion that the concern over low level waste burial grounds is attributable to the institutional questions to an extent at least as great, if not greater than the technical questions. I previously mentioned the regulatory jurisdiction from the early days of this program. I think it is well recognized at the Federal level that the States will have a significant role to play in the regulation of low level waste burial. I am equally certain the States realize that the Federal Government has a significant role to play no matter where the jurisdiction lies. It should be noted that Section 14(b) of the NRC Authorization Act for FY 1979 directs the NRC to prepare a report on means for improving opportunities for State participation for siting, licensing and developing nuclear waste storage or disposal facilities. The NRC Working Paper "Means For Improving State Participation in the Federal Nuclear Waste Management Programs" (NUREG-0513) was published on December 20, 1978 (NRC, 78). Although it is heavily oriented to the high level waste program, it notes that a key item of interest to the States in the low level waste area is the question of Federal control of the sites. However, other issues such as waste classification, chemical toxicity of wastes and volume reduction are also of high interest to the States.

It certainly seems that a prime need is to get a more formalized regulatory framework in place so that this sector of the industry can be regulated on a more consistent, uniform and effective basis. It is vitally important that the regulatory framework be one which can be implemented by either the States or the Federal Government. This would help assure that any regulatory or technical uncertainties in low-level waste management are minimized.

A closing word about the role of government. From time to time we have heard various people proclaim there should be less regulation. In recent months there have been a number of steps taken to deregulate the airlines and there have been some actions taken to reduce the extent of OSHA regulations.

Heclo (1977) in his book "A Government of Strangers" stated:

> "Americans have long expressed impatience with red tape and Washington bureaucrats, but few of the heavy demands they make on the federal government can be satisfied without some form of organized bureaucratic activity. Public opinion polls show, for example, that the overwhelming majority of Americans agree that the federal government should control inflation; avoid depression; assure international peace; regulate (but not run) private business; and see to it that the poor are taken care of, the hungry fed, and every person assured a minimum standard of living. But a comparably large majority also agree that the federal government is so big and bureaucratic that it should return more taxes to subnational governments and count mainly on the states to decide what programs should be started and continued."

The issue of low level radioactive waste disposal might be included in this list.

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Table 1. Volume of wastes and amount of activity buried at low level sites; cumulative through 1977.

Location	Year Licensed	Volume buried $X10^6 ft^3$	BPM (Ci)*	SM (lbs)	SNM (kg)	Pu (kgm)
Beatty, NV	1962	2.16	156,059	142,976	191.46	14.3**
Maxey Flats, KY	1962	4.77	2,400,691	532,988	431.8	63.76
Hanford, WA	1965	.58	434,358	14,354	93.74	30.63***
Sheffield, IL	1967	3.0	58,000	594,000	54.8	13.4
West Valley, NY	1963	2.36	700,000	1,026,000	56.0	4.0
Barnwell, SC	1971	5.04	558,000	732,000	653.0	0
Totals		17.91	4,307,108	3,042,318	1480.80	122.09

* BPM, byproduct material; SM, source material; SNM, special nuclear material, including plutonium.

** Cumulative through 1976.

*** 1970 through 1977 only.

REGULATORY ASPECTS OF LOW LEVEL WASTE DISPOSAL

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Abstract

Regulating a low-level waste disposal site reveals many interesting facets of how this nation's low-level waste is being managed. Incidents and occurrences that happen to radioactive waste prior to arriving at a burial site and after it is received at the burial site will be described. Regulating the site involves numerous disciplines of Engineering, Chemistry, Hydrology, Geology, Health Physics, and others. Regulatory involvement of these elements will be reviewed. Classification of low-level waste as presently being implemented at the Chem-Nuclear, Barnwell, South Carolina, site will be discussed.

Discussion

Before commencing the substance of my presentation, I would like to briefly discuss the State's role in the regulation of ionizing radiation. The State of South Carolina became involved in the regulation of ionizing radiation in 1967 when the South Carolina Legislature enacted the Atomic Energy and Radiation Control Act. On September 15, 1969, the State became an "Agreement State" pursuant to Section 274 of the Atomic Energy Act of 1954 as amended, thereby assuming regulatory responsibilities for certain radioactive materials. Such responsibility includes the licensing of low-level waste burial facilities; and, on April 13, 1971, one such license was issued to Chem-Nuclear Systems, Inc., authorizing the use of approximately 250 acres of property in Barnwell County, near the Savannah River Plant property, as a low-level radioactive waste burial facility. This information provides you with some understanding of the regulatory atmosphere within our State.

The Barnwell, S. C., Chem-Nuclear site is one of the three remaining low-level commercial burial sites in the U. S. that is presently receiving commercial low-level waste for land disposal. The other two sites are located on the west coast. The volume of low-level radioactive waste disposed of at the Barnwell site has increased significantly over the past several years. For example, in 1975 the total volume of waste buried was 638,137 cu. ft.; whereas, in 1978, this volume had increased to 2,173,955 cu. ft. Much of this volume increase was due to the Maxey Flats and West Valley shutdown. The recent termination of the Sheffield, Illinois operation has further complicated the disposal of commercial low-level radioactive waste.

As a result of closing the Sheffield site, the State of South Carolina deemed it necessary to impose a monthly restriction on the volume of waste that could be disposed of at the Barnwell site. This volume restriction was imposed on the operator of the site because of concerns over using available burial space much faster than was originally planned. Approximately one-half of the waste buried at the Barnwell site during 1978 originated outside the region of the U. S. that the site was intended to serve. The volume restriction has not been the first such action imposed on Chem-Nuclear's operation. The State of S. C. initially restricted burial of plutonium or any other transuranic elements. During 1976, Chem-Nuclear was not allowed to ship or receive additional bulk shipments of contaminated liquids from nuclear power reactor sites. Solidification at the point of origin was required rather than solidify the liquids at the burial site for disposal. Also, we have recently imposed a restriction on the site operator not to receive or bury radioactive waste containing more than 1% oil by volume.

The Chem-Nuclear site property is owned by the State of South Carolina, whereupon, the State leases the site for Chem-Nuclear to operate. Chem-Nuclear's operation is licensed by both the State of South Carolina and the U. S. Nuclear Regulatory Commission. Special nuclear material disposal authorization is licensed by NRC in quantities greater than is authorized under State jurisdiction. The joint licensing of the site operation has proven to be a satisfactory arrangement. Close communication between the NRC Office of State Programs and Region II Compliance Office has demonstrated a close Federal-State partership in this respect.

During 1978, 12 incidents occured with waste being shipped to the Chem-Nuclear site that required investigation. These include contamination of personnel, breach of package integrity, liquid waste shipments contaminated vehicles, and freight, and vehicle accidents. A total of 35 man-days was expended due to investigation and follow-up action for protection of the public health and safety. Many of these investigations could have been avoided had the waste been prepared and packaged properly for shipment and disposal.

Routine monitoring (spot checks) of packages received at the Chem-Nuclear site indicates to us that not enough attention is being given to the management of low-level waste at the point of origin. During a routine inspection of a shipment of institutional waste from one of our major educational institutions, the following was found. The shipment of 74 - 55 gallon steel drums contained glass jugs, coke bottles, wine bottles, etc. filled with liquid chemicals (organics) containing tracer amounts of radioactivity. In addition to violating Chem-Nuclear's license, the shipper was also in violation of DOT regulations.

Solidified waste that arrives at the burial site is also routinely inspected by personnel from the Bureau of Radiological Health. It is evident from our observations that quality assurance programs are inadequate with respect to preparation of liquid waste for shipment and

ultimate disposal. Solidification medias and the management of organics has been of major concern at the Chem-Nuclear burial site. Standards in these areas and areas I have not touched on are needed. We need these standards now.

An important aspect of perpetual maintenance of a low-level burial facility is a program of daily site upkeep and continuous environmental monitoring. The former is important to prevent the occurrence of problem situations and the latter to detect potential problems in their infancy. Chem-Nuclear Systems, Inc. presently undertakes all site maintenance being reviewed periodically by Health Physicists from the Bureau of Radiological Health. Both the State and Chem-Nuclear conduct extensive on and off site environmental monitoring programs.

In regulating the burial site operation, the staff of our Bureau of Radiological Health reviews and approves changes in site operation, inspects each trench prior to use for burial, physically inspects the site routinely for erosion control and trench conditions, and audits the entire burial operation. Based on our experience at the Chem-Nuclear site, we believe that properly engineered trenches and management of surface water is an important factor in the management of a low-level radioactive waste burial facility.

BURIAL OF SMALL QUANTITIES OF RADIONUCLIDES WITHOUT PRIOR NRC APPROVAL

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Abstract

The U.S. Nuclear Regulatory Commission has proposed to delete a regulation (10 CFR 20.304) which allows licensees to bury specified small quantities of radionuclides anywhere without notification or approval of NRC, subject to certain conditions. In developing this proposed deletion, the NRC staff contacted licensees, state officials, inspectors, and others to obtain information on disposal practices. The Commission and its staff tentatively concluded that burials pursuant to 10 CFR § 20.304 should stop, and that the impact of the proposed deletion would be small. If review of public comments does not change this evaluation, 10 CFR § 20.304 will be deleted, and burials will have to be specifically approved in advance by NRC.

Background

U.S. Nuclear Regulatory Commission (NRC or Commission) regulations currently allow licensees to bury small quantities of radionuclides anywhere without notification or specific approval of NRC. Specifically, 10 CFR § 20.304 provides that no licensee shall dispose of licensed material by burial in soil unless:

(a) The total quantity of material buried at any one location and time does not exceed 1,000 times the amount specified in Appendix C of 10 CFR Part 20,

(b) Burial is at least four feet deep,

(c) Burials are at least six feet apart, and

(d) Not more than 12 burials are made per year. Records of the burials must be maintained by the licensee as provided in § 20.401.

Burials which do not comply with § 20.304 must receive specific Commission approval as provided by 10 CFR § 20.302.

Requests for reevaluation of 10 CFR § 20.304

Section 20.304 has not been amended since its adoption in 1957. Over the last few years, NRC has received several requests for reevaluation of this regulation. For example, the National Conference of Radiation Control Program Directors and Officials from the majority of the States have requested such a reevaluation.

The reason given for the need to reevaluate § 20.304 is that people could be overexposed to radiation by inadvertently disturbing burials, since licenses may be terminated, burial records lost, land sold, etc.

The NRC agreed that § 20.304 should be reviewed, and the radiological risks and potential impacts of revising § 20.304 were assessed as described below.

Radiological risk associated with § 20.304

Section 20.304 allows burial of quantities up to 1,000 times the amounts listed in Appendix C of 10 CFR Part 20. The Appendix C values are approximately the lesser of two values: (1) the amount that a standard man would inhale when exposed for one year to the highest concentration allowed for unrestricted areas, or (2) the amount which produces 1 milliroentgen per hour of gamma radiation exposure at 10 centimeters.

There is a remote possibility that radionuclides buried pursuant to § 20.304 could deliver large doses to individuals disturbing a burial site. However, an individual would have to dig up the buried material, and remain near the site long enough to inhale a large fraction of the material or be exposed to a large direct radiation dose.

The NRC staff concluded that the risk associated with burials pursuant to § 20.304 is small. However, it was recognized that amendments to § 20.304 could improve public health protection by improving data and controls over burials of even small quantities of radionuclides. For the NRC staff to decide whether such amendments were justified, it was necessary to assess the potential impacts on licensees.

Potential impacts of amendments to § 20.304

Amendment or deletion of § 20.304 would not prohibit burials. Rather, licensees wishing to continue burials previously conducted pursuant to § 20.304 would have to apply to NRC for approval in advance, or send waste to a commercial burial ground. The staff estimated that obtaining prior approval would require a few man-days extra effort on the part of each licensee.

Because licensees are not required to inform NRC of burials made pursuant to § 20.304, it is difficult for the staff to estimate how many licensees might be affected by an amendment to the regulation. However, the staff made a rough estimate by contacting NRC inspectors and State officials. This survey covered 25 states, over 5,000 NRC licensees, and over 5,000 licensees under the NRC Agreement States. (There are over 16,000 NRC and Agreement State licensees in the U.S.)

A summary of the information collected is shown in Table I. It should be emphasized that these are rough estimates; exact data could only be obtained by the time-consuming process of contacting thousands of licensees for review of records. Also, it should be noted that some Agreement States have already prohibited burials which NRC would permit pursuant to § 20.304.

Based on this survey, the NRC staff estimated that less than 100 licensees are using § 20.304 to perform burials. Therefore, it was concluded that the impact of amendments to § 20.304 would be slight. The staff then considered the alternatives described below.

TABLE I

NRC AND AGREEMENT STATE LICENSEES PERFORMING § 20.304 BURIALS

States Surveyed	Total Material Licensees	Approx. Number Performing Burials
NRC Region I (11 states)	2300 (non-agreement)	less than 5%
NRC Region III (8 states)	2900 (non-agreement)	less than 25
California	1700	5
Texas	1310	10-15
New York	1000	none
Florida	700	none
South Carolina	151	none
Oregon	170	none
Arkansas	248	1
New Hampshire	59	2
Kansas	207	3

Notes:

1. There are 25 Agreement States regulating over 9000 materials licensees. NRC regulates about 7000 materials licensees.

2. Licensee totals for individual states are Agreement State licensees only.

3. There have been no unlicensed burials in New York, Florida, South Carolina, and Oregon for several years.

4. Source: NRC inspectors and Agreement State officials.

Alternatives for action on § 20.304

No Action - The "no action" alternative was rejected because it was not responsive to the expressed concerns of the public.

Notification - The staff considered amendments to § 20.304 which would merely require notification of NRC and the Agreement States after the burials were performed. This was rejected because burials could still be conducted without prior regulatory review for suitability of location, proper marking, etc.

Additional restrictions - The staff considered additional restrictions such as (1) confinement of burials to restricted areas, (2) removal of buried materials prior to termination of licenses, or (3) additional limitations on quantities and types of radionuclides to be buried. The staff concluded that generic decisions on these issues were less desirable than case-by-case decisions.

Deletion - The staff chose deletion of § 20.304 as the best alternative because it is responsive to the concerns of the public and would have little impact on licensees.

Conclusion

The NRC staff recommended deletion of § 20.304 to the Commission, and the deletion has been approved as a proposed amendment and published for public comment in the Federal Register (43 FR 56677, December 4, 1978). Copies of the Federal Register notice were mailed to all NRC licensees and Agreement State officials. The public comment period ended on February 2, 1979. However, I would encourage attendees who have additional comments to submit them within the next few weeks, and I will see that they are considered.

Although our review of the public comments is not complete, we have already identified the following issues which will have to be addressed:

1. The public comments contain data identifying licensees using § 20.304 and assessing the potential impact of deletion of § 20.304. This data will have to be reviewed so that the impact analysis of the proposed regulation can be updated and improved.

2. If § 20.304 is deleted, some licensees using § 20.304 may apply to NRC for approval to continue burials. The staff will need to provide guidance to licensees on how these applications will be handled.

3. If NRC deletes § 20.304, appropriate actions on the part of the Agreement States will have to be determined.

4. Some licenses contain conditions which allow burials in compliance with § 20.304; that is, specific NRC approval has been obtained for the burials. The impact of deletion of § 20.304 on these licenses, if any, needs to be determined.

After our review of the public comments is complete, we will make a recommendation to the Commission for final action on § 20.304. This process usually takes several months. I should point out that none of the alternatives described previously have been ruled out. After public comments have been considered, any alternative could be chosen.

WASTE MANAGEMENT PRACTICES IN DECOMMISSIONING NUCLEAR FACILITIES*

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ABSTRACT

Several thousand sites exist in the United States where nuclear activities have been conducted over the past 30 to 40 years. Questions regarding potential public health hazards due to residual radioactivity and radiation fields at abandoned and inactive sites have prompted careful ongoing review of these sites by federal agencies including the Department of Energy (DOE) and the Nuclear Regulatory Commission (NRC). In some instances, these reviews are serving to point out poor low-level waste management practices of the past. Many of the sites in question lack adequate documentation on the radiological conditions at the time of release for unrestricted use or were released without appropriate restrictions. Recent investigations have identified residual contamination and radiation levels on some sites which exceed present-day standards and guidelines. The NRC, DOE, and Environmental Protection Agency are all involved in developing decontamination and decommissioning (D&D) procedures and guidelines which will assure that nuclear facilities are decommissioned in a manner that will be acceptable to the nuclear industry, various regulatory agencies, other stakeholders, and the general public.

INTRODUCTION

Decontamination and decommissioning (D&D) of nuclear facilities is playing an increasingly greater role in demonstrating the credibility of

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the nuclear industry. Several thousand sites exist in the United States where nuclear activities have been conducted over the past 30 to 40 years. Many hundreds of these sites either have been abandoned or have become inactive. A few have been totally decommissioned and released for unrestricted use. Recently, questions regarding potential public health hazards due to residual radioactivity and radiation fields at the abandoned and inactive sites have prompted careful review of these sites by federal agencies including the Department of Energy (DOE) and the Nuclear Regulatory Commission (NRC).

The DOE is responsible for the radioactivity in facilities it owns or controls. Also, DOE has assumed the responsibility for abandoned or inactive sites which were under the control of its predecessors, the Manhattan Engineer District (MED), the Atomic Energy Commission (AEC), and the Energy Research and Development Administration (ERDA). Of immediate concern to DOE are 22 inactive uranium mill sites in the western part of the United States. In addition, DOE has the responsibility for reviewing more than one hundred excess MED and AEC sites that played a role in the early development of the atomic energy program.

Decommissioning criteria applied to NRC licensees prior to 1965 were not as stringent as present guidelines (Di78). Documentation of the final radiological status of the properties involved may be inadequate. As a consequence, NRC has initiated a systematic program to review all of its docket files of licenses terminated prior to 1965. In addition, formal radiological surveys are being conducted at a few selected sites with a known potential for residual contamination.

While individual states have not as yet undertaken extensive review programs, it is well-known that a few problem areas exist. For example, a former nuclear facility in Tennessee and another in New York (DOE78), both with significant levels of residual contamination, have become inactive and essentially abandoned. The site in Tennessee was partially decontaminated at federal expense by the Oak Ridge National Laboratory (ORNL), and at least a portion of the New York site may become a ward of the state for cleanup.

OBSERVATIONS

Some nuclear sites have been decontaminated successfully and decommissioned. Former AEC reactors, including the Piqua, Elk River, and BONUS reactors, have been decommissioned. In one of the most ambitious decommissioning actions ever undertaken, the Elk River Reactor in Minnesota was completely dismantled and removed from the site. The NRC also has decommissioned a large number of formerly licensed sites with documentation verifying that the sites met the established decommissioning criteria (Di78). However, it has been pointed out that a number of sites have either been abandoned or allowed to become inactive without adequate documentation of radiological conditions at the time of release or without imposing appropriate restrictions. As a consequence, recent investigations (Ha77, Di77, Le78b, Pe78) have identified residual contamination and radiation levels on these sites which exceed applicable standards and guidelines (ANSI78, Di78).

Of the 22 inactive uranium mill sites, 16 are accessible to the general public, seven have had no significant stabilization against erosion, and 16 show evidence of off-site contamination (Go76). While some of

these locations are remote, at least four of the sites are within a 16-km radius of a population exceeding 10,000 persons (Go76). Many of these sites have existed without active surveillance for 10 to 15 years. From these observations, it is apparent that some early waste management practices were less than adequate by today's standards even though waste management was judged to be adequate at many of these sites under the then existing standards.

In the case of the excess MED/AEC sites, properties with significant levels of residual contamination and/or radiation levels have been identified in or near major metropolitan areas (Di77, Le78a, Le78b). While the total quantities of residual radioactive materials may be less than those quantities at inactive uranium mills, there are more people who potentially could be exposed. Most of these sites were contaminated in the 1940's and 1950's and have been inactive for 10, 20, or even 30 years.

The NRC also has discovered previously licensed sites that have been decommissioned without adequate verification of the radiological status (Pe78) It is difficult to assess the possible extent of this problem. The NRC estimates that as many as 8,000 source material and special nuclear material licenses have been terminated over the years prior to 1965. Again, because waste management practices in the past were not as thorough as present practices, many of these sites could not be decommissioned using present-day decommissioning criteria without substantial decontamination.

FACTORS CONTRIBUTING TO RESIDUAL CONTAMINATION PROBLEMS

It should be pointed out that many of the current radiological problem sites had their beginning long before the days of the AEC and other regulatory authorities. For example, Lindsay and Company began operation

in West Chicago in 1931 (Fr78). Another example is the former Vitro Rare Metals Plant in Canonsburg, Pennsylvania, which was used as early as 1911 for the commercial extraction of radium from carnotite ore (Le78a). Consequently, at least a portion of the present problems can be blamed on a total lack of regulation. Although advisory groups had been formed as early as 1929, no regulatory authority existed until the Atomic Energy Act of 1946 when Congress established the AEC. The AEC and its successors had no regulatory authority over naturally occurring, non-source material (e.g., ^{226}Ra) until Congress passed the Uranium Mill Tailings Radiation Control Act of 1978 (PL 95-604) which defines tailings as "byproduct material," thus, giving NRC authority over such materials.

Users or handlers of large quantities of radioactive materials (e.g., uranium mills) have tended to use large scale industrial processing techniques which have a few percent loss and/or spillage. As a consequence, the facility involved became generally contaminated with low-level radioactive waste. Much of the feed material contained "natural" radioactivity which was considered rather innocuous. Efforts to prevent the spread of materials which had been extracted recently from the earth received little attention. Even to this day, several uranium mill tailings piles have had no deliberate surface stabilization (Go76) to prevent erosion or security measures to prevent casual access by the public.

In some cases, the large user would contract for waste disposal via conventional industrial means. As a result, radioactive waste has been placed in muncipal or industrial landfills or other such accessible

locations. The examples of this are numerous and include Middlesex, New Jersey, and Burrell Township, Pennsylvania. These specific examples are covered in greater detail by Goldsmith (Go79).

In other cases, the large users possessed a property which was unused, and perhaps unusable, for other purposes which became the collecting place for nuclear waste. Although the site was not strictly considered a waste burial site, radioactive material accumulated there over the years awaiting ultimate disposal. Specific cases are represented by the Kerr McGee site (Fr78) (old Lindsay Light and Chemical Company) in West Chicago, Illinois, Canonsburg, Pennsylvania, (Le78a), and the Haist property (Le78b) in Tonawanda, New York, which was used by Union Carbide under a lease arrangement with property owners and MED.

Many licensees who used small quantities of radioactivity took advantage of the on-site burial provisions of 10 CFR 20. While this provides expedient removal of radioactive waste from sight, the problem of ultimate disposal was simply deferred to license termination. It is uncertain as to whether many sites can be decommissioned and released for unrestricted use when substantial quantities of radioactive materials are known to be buried on the site, even if the material is below licensable concentrations (e.g., ores containing by weight 0.05% or more of uranium).

The pressures of commercial competition and governmental regulation caused the termination of many nuclear activities. In the case of uranium mills, antiquated equipment and a low profit margin caused by a depression in the price of uranium were responsible for the premature closing of several mills. Some firms with marginal operations tended to short-cut on waste management procedures to maintain a favorable economic picture. Such

was the case with the American Nuclear Corporation in Oak Ridge, Tennessee. Government (AEC or state) inspections were too infrequent to detect items of noncompliance on a timely basis. As a result, a facility could experience significant degradation in general housekeeping in the period between inspections, which in some cases might be as long as several years.

In a few cases, sites have been virtually abandoned. One can speculate that the reasons for this abandonment range from ignorance of decommissioning requirements to the more likely case of financial insolvency. Since it has not been regulatory practice (NRC78) to require decommissioning funding arrangements (e.g., posting of bond) in advance of decommissioning for small users, the licensee frequently does not have the financial resources to cope with the cleanup and decontamination required to be able to obtain consent for unrestricted release.

There have been numerous cases where radioactive waste materials have been misused. The removal of tailings and their subsequent use as fill around homes, schools, and other buildings in Grand Junction, Colorado is one noteworthy case. In fact, many of the inactive uranium mill tailings sites are accessible to the general public (Go76). Consequently, the tailings materials easily could be misused at these sites. The misapplication of radioactive materials extends to other source material as confirmed by a review of NRC records (Cr78). Another example concerns the unauthorized removal of contaminated tools and equipment from the commercial burial site of the Nuclear Engineering Company at Beatty, Nevada.

Along similar lines, radioactive material has been transferred to clean sites without specific application, probably in ignorance of the radiological hazards involved. Examples of this include the spread of contamination from the Kerr McGee site in West Chicago to at least 75 other locations in in the Chicago area (Fr78) and the relocation of a major portion of the radioactive residues from the Haist property (Le78b) to the nearby Seaway Industrial Park in Tonawanda, New York (Le78c). Numerous small areas of radioactive contamination can also be found in residental areas of Canonsburg, Pennsylvania, presumably spread there from the early operations at the Vitro site (Le78a).

Another problem has been the lack of a comprehensive, internally consistent set of decommissioning criteria and numerical guidelines. Many contamination limit proposals have been adopted for use at specific sites, apparently with marginal scientific justification. The Grand Junction Remedial Action Criteria (CFR76) were written specifically to resolve the dilemma at Grand Junction but may have applicability to other sites contaminated with radium. The Environmental Protection Agency (EPA) is the federal agency responsible for providing federal guidance on radiation exposure related to the release of contaminated property. As an example, EPA is considering interim recommendations for radiation levels at new structures located on Florida phosphate lands (FR76). While these fragmentary guidelines are of value for specific applications, a master set of decommissioning criteria with general applicability does not exist. Surely it is not practical for the nuclear industry to develop a new and different set of criteria for each D&D action.

CORRECTIVE ACTION

While waste management practices involved in the decommissioning of nuclear facilities in the past have resulted in unacceptably high levels of residual contamination at many sites, a number of steps have been taken recently to correct this situation. Several federal agencies are actively pursuing programs to correct past D&D deficiencies and to provide improved D&D processes in the future.

For many of the sites formerly utilized by MED and AEC, available records before the recent resurveys were not adequate to identify the radiological condition at the time government controls were relinquished (Cr78). Records for some formerly licensed sites are similarly lacking in pertinent radiological information (Cr78). Both DOE and NRC have programs to determine the adequacy of documentation and to make new surveys if warranted. The DOE program is known as the Formerly Utilized Sites—Remedial Action Program.

In addition to the review of terminated licenses which is being conducted by the NRC, the whole decommissioning policy of that agency is being reevaluated (NRC78). The NRC has sponsored considerable research to determine the technology, safety, and costs associated with decommissioning reactors (Sm78) and fuel reprocessing plants (Sc77). The DOE was instrumental in the passage of Public Law 95-604, Uranium Mill Tailings Radiation Control Act of 1978. This law provides the legal basis for remedial action at the inactive mill tailings sites and at the former Vitro Rare Metals Plant in Canonsburg, Pennsylvania. The EPA continues to work on the development of appropriate criteria and guidelines (FR77, FR78); however, a comprehensive set of decommissioning criteria is in an embryonic state.

Interest has been shown by a number of technical societies such as the American Nuclear Society and the Health Physics Society, especially with respect to their standards committees. Other interest groups such as the Atomic Industrial Forum (Ro78) and the American National Standards Institute (ANSI78) continue to make contributions in the D&D field.

While all of the problems related to waste management in D&D activities have not been solved, it is encouraging to see so much interest and effort. One concern is that all of this effort is not well coordinated. An inter-agency task force much like the one organized in Canada (AECB77) to provide D&D criteria might be the answer to a more efficient production of the much needed guidance in this country. For example, DOE could take a leading role in this activity since it is encumbent upon DOE to implement D&D at a large number of facilities including the excess MED/AEC sites, inactive uranium mill sites, and 300 to 400 excess contractor facilities. In all likelihood, Congress would have to act to set up the machinery for such a broad scope effort. Other participants in this undertaking should include, but not be limited to, NRC, EPA, and state regulatory agencies.

SUMMARY

A number of factors have contributed to the marginal waste management practices observed in decommissioning of nuclear facilities. Some of the more pertinent factors include:

1. lack of regulation--particularly with respect to naturally occurring radioactivity;
2. poor control measures on large scale industrial processes;

3. radioactive waste disposal by conventional methods such as dumps, landfills, and on-site burial;
4. misapplication of waste products containing radioactive material;
5. short-cutting of waste management procedures to increase the profit margin;
6. abandonment of sites;
7. lack of continuing surveillance over inactive sites; and
8. lack of a comprehensive set of D&D criteria.

As a consequence, recent investigations have revealed residual contamination and radiation levels on some sites which exceed present-day standards and guidelines. Efforts by major federal agencies including DOE, NRC, and EPA are serving to correct these deficiencies. An interagency task force could be the most expedient approach to arrive at the D&D guidance which is urgently needed by the nuclear industry.

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Decommissioning Standards - the Radioactive Waste Impact

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Introductions and Conclusions

Several considerations are important in establishing standards for decommissioning nuclear facilities, sites and materials. This review includes discussions of some of these considerations and attempts to evaluate their relative importance. Items covered include the form of the standards, timing for decommissioning, occupational radiation protection, costs and financial provisions, and low-level radioactive waste.

Decommissioning appears more closely related to radiation protection than to waste management, although it is often carried under waste management programs or activities. Basically, decommissioning is the removal of radioactive contamination from facilities, sites and materials so that they can be returned to unrestricted use or other actions designed to minimize radiation exposure of the public. It is the removed material that is the waste and, as such, it must be managed and disposed of in an environmentally safe manner. It is important to make this distinction even though, for programmatic purposes, decommissioning may be carried under waste management activities.

It was concluded that the waste disposal problem from decommissioning activities is significant in that it may produce volumes comparable to volumes produced during the total operating life of a reactor. However, this volume does not appear to place an inordinate demand on shallow land burial capacity. It appears that the greater problems will be associated with occupational exposures and costs, both of which are sensitive to the timing of decommissioning actions.

Other areas which are not addressed in this paper but which may become important in considerations of decommissioning standards include: differences between existing facilities and planned facilities regarding decommissioning, concepts related to dedicated sites and facilities, issues of the relative costs and benefits of various methods such as removal/disposal or fixation in place, and the period for which impact assessments must be conducted. All of these topics must be examined during the development of decommissioning standards but their possible influence on the standard itself is currently not clear.

REVIEW OF EXISTING STANDARDS, REGULATIONS AND CRITERIA

The most difficult problem in establishing radiation protection standards for decommissioning is the choice of the rationale or basis of the standard. Once the rationale is chosen, the approach to setting the standard (or the form of the standard) is defined and the standard effort can proceed in a straightforward fashion. For example, if the existing FRC guidance is to provide the basis for decommissioning standards, the principal task becomes the ALAP approach, the familiar cost-effectiveness assessment, and the standard is established at a level where further expenditures provide little additional health protection. Other rationale are available, however, and must be examined in light of identifying which rationale would potentially provide the greatest assurance of maximizing public health and environmental protection.

As a initial step in identifying rationale which may be suitable for decommissioning, a thorough review is planned of existing standards, regulations and criteria which deal with areas such as exposure rate limits in uncontrolled areas, decontamination criteria and standards, and others. The objective of the review is more than a listing of applicable standards, however it is the rationale for the standards which requires identification. This identification will require a thorough search of the history of the applicable standards and will include such items as:

the reason the standard was established,
the contribution and opinion of expert advisory groups to the development of the standard,
the use or interpretation made of existing authoritative guidance, such as that of the Federal Radiation Council, and
the input by the public and governmental agencies during the public comment period, both written and oral.

This information can be useful in determining the basis for decommissioning standards in that it will make available the logic previously followed by those who addressed the decommissioning issue, or one closely related. Too often the thinking performed in the past is neglected because it is believed new or unique issues are being confronted. While there is no great assurance that past thinking will provide a sound rationale for the current effort, it is believed it will offer sufficient insight to be helpful.

Previous efforts that have been chosen for examination thus far include:
-10 CFR Part 20, Paragraph 105, U.S. NRC
-EPA's proposed Federal Radiation Protection Guidance on Dose Limits to Persons Exposed to Transuranium Elements in the Environments

-the Surgeon General's Recommendations to the State of Colorado for Radiation Protection for Individuals Living in Residence Contaminated with Uranium Mill Tailings
-Regulatory Guide 1.86, Termination of Operating Licenses for Nuclear Reactors, U.S. NRC.
-ANSI Standard N328, Control of Radioactive Surface Contamination on Materials, Equipment and Facilities to be Released for Uncontrolled Use

TIMING

At the end of its operating life, a nuclear power plant itself or other nuclear facilities become a form of radioactive waste. As such, these facilities and sites must be restored to some level of radioactive exposure that can be found acceptable (or not unacceptable) for their unrestricted use in the future. An important part of the decommissioning qustion is the time at which facilities and sites must meet dose level requirements. This is especially pertinent to the commercial reactors because at the time of reactor retirement radiation levels will be quite high, primarily due to activitation products. Since most of the activation products have relatively short lifetimes, less than 5 years half-lives, there are potentially significant benefits to be gained by delaying decommissioning until there is appreciable decay of the activation products. However, delay of decommissioning is undesireable from the view of both the possibility of institutional failures and the responsibilities of the generation which received the benefits to reduce the impact (or cost) to future generations. Thus, the challenge is to select the timing for decommissioning to optimize the benefits to be obtained through radioactive decay and to minimize the remoteness of the generation conducting the decommissioning from the generation receiving the benefits.

An important consideration here is the ultimate use to which the site or facility is to be put. Obviously, a contaminated laboratory that is planned for future use must be decommissioned (or decontaminated) to a level that will protect laboratory workers during this future use. However, the case is not as clear-cut for a nuclear power and centralized station power is unclear. A nuclear power plant site represents a large investment for the owner-utility in terms of transmission lines, switch yards and site improvements, especially cooling water provisions. This results in a strong incentive for continued use of the site for centralized power, nuclear or not. Thus,

central power sites may eventually become mixtures of nuclear and fossile fuel generation. In addition, utilities may choose to recycle certain structures or other parts of a site, such as containment or cooling water structures, if nuclear remains a viable option. These considerations serve to greatly complicate the timing question for decommissioning. However, timing requirements must be established to assure public health and environmental protection for future generations. Thus, such uncertainties should be considered to the extent possible at this time with the concept that future activities may require a reevaluation of the timing requirements.

As an initial estimate of what the timing for decommissioning should be, it is suggested that EPA's proposed Radiation Protection Criteria (EP) for the Disposal of Radioactive Waste be used. Criterion No. 2 of this proposal states, "Controls which are based on institutional functions should not be relied upon for longer than 100 years to assure continued isolation from the biosphere." While this criterion addresses radioactive waste disposal rather than decommissioning, these two issues share a common theme in the timing area, that of the acceptance of continuing reliance on institutions for health and environmental protection. Applied here the result is the scenario:

Nuclear power plant licensing and construction	10 years
Nuclear power plant operations	40 years
Maximum decommissioning time	50 years
Total	100 years

This scenario provides a miximum 50 year period for decommissioning while meeting the intent and spirit of the criterion. A 50 year period following reactor shutdown also provides a most substantial reduction in radiation exposure levels from activation products.

OCCUPATIONAL RADIATION PROTECTION

The major health related issue in determining timing requirements for decommissioning is occupational radiation exposure. In most cases and especially in the reactor situation, appreciable reductions in occupational exposures incurred during decommissioning can be realized by postponing decommissioning activities until short-lived radionuclides have significantly decayed. This is based on the assumption, or even possibly the fact, that most of the residual radioactivity at a reactor will be due to activation products which are predominantly short-lived materials. The situation at nuclear facilites other than reactors is not as clear because the contaminating materials will have longer lived characteristics.

Decommissioning evaluations for reactors normally follow a scenario which includes removal and offsite shipment of all spent fuel, packaging and offsite shipment of the low-level radioactive waste, and mothballing the reactor for a period of time. This time period is largely controlled by the decay rate of cobalt-60 a significant activation product in terms of production and external exposure with a half life about 5 years. Longer lived activation products are also present, such as Ni-59 and Nb-94 but not in the same magnitude as cobalt and other short-lived radionuclides. Since cobalt-60 is a predominant radiation source and has longest half life of the shorter-lived materials, occupational exposure reductions to be gained by delayng decommissioning can be directly related to the half life of cobalt-60. For example, a 20 year delay would produce a reduction of 16 and a 50 year delay the maximum as discussed above results in a factor of 1,000 reduction. Therefore, it appears highly that a maximum timing limit of 50 years for reactors will provide an optimum point for occupational exposures balanced with economic and institutional considerations.

Facilities providing the fuel for and managing the waste from reactors, along with most of the defense and research oriented facilities pose an additional problem in decommissioning since the contamination is a result of longer-lived materials. First, the front end of the fuel cycle is dominated by uranium and its daughters thorium-230 and radium-226 which lead to the radon-222 decay chain. Because of the long-lived nature of these products, there is nothing to be gained through a delay and decommissioning activities should be conducted immediately upon plant closings. As for the tail end of the fuel cycle, regardless of decisions concerning recycle, facilities most likely will be highly contaminated with longer-lived materials and little will be gained through delays in decommissioning such facilities. A similar situation can be expected at most of the defense related and research oriented facilities.

DECOMMISSIONING COSTS AND FINANCIAL PROVISIONS

Current estimates indicate decommissioning costs may be great and, what is worse yet, the current estimates have not been substantiated by actual proactice. Thus, no evidence exists to prove that the costs may not be significantly greater than current estimates. There is no way to avoid some costs since prudent public health policies dictate that nuclear facilities and sites be returned to a condition of insignificant radiation threat to the public. A logical policy

regarding such costs would be that the beneficiaries of the activity be responsible for the decommissioning of the activity-related sites and facilities. In addition since the decommissioning costs appear great, it seems logical that financial provisions should be made before the activity is initiated, or at the very least during the early stages of the activity, to assure the existence of sufficient funding when decommissioning takes place.

The diverse nature of nuclear facilities will lead to numerous different solutions to the funding problem. For instance, nuclear power plants, as regulated utilities, have a relatively secure economic future, especially when compared to such facilities as uranium mills. The situation for reactors appears relatively simple, because the electric utility that can afford to build nuclear power plants is presumed to have a continuing role. If the utility could demonstrate that the decommissioning cost was a small fraction of the power production cost, say less than 5% to 10%, this cost could be included in the operating costs and regulated by the controlling Public Utility Commission (PUC). Perhaps PUCs could even drop the decommissioning cost from the operating costs, if sufficient emphasis was placed on the relatively small fraction the decommissioning cost would be and on a commitment by the utility that decommissioning would occur and costs would be borne by the utility. The major obstacle to this approach is the lack of firm cost data for decommissioning. It should be recognized that any funding requirements would require approval of the regulating PUC.

The situatiuon is not as simple for other nuclear facilites, however. First the question of financial responsibility and a continuing role must be addressed since most of these facilities are owned by companies in a competitive market with no oversight provided for the common good, in contrast to utility regulations by PUCs. A case in point here is the company which owns the NFS facility in West Valley, New York. According to a recent study the company can terminate its' lease for the site at the end of 1980 with the question as to their financial responsibility unclear, but apparently with little likelihood that the company will be liable. The cost for decommissioning this site is estimated at 536 (DE) million, or much greater than the initial cost of the plant, constructed during the early and middle 60's. This illustrates the second point to be made in decommissioning non-reactor sites and facilities - the cost of decommissioning may be far greater than the initial capital investment. In addition to the West Valley situation, the abandoned uranium mill tailings sites in the western part of the U.S. further amplify this financial problem.

The only logical conclusion that can be drawn here is that arrangements must be made before licensing such activities to assure both responsibility for decommissioning and adequate funding.

LOW-LEVEL RADIOACTIVE WASTE FROM DECOMMISSIONING

Estimated volumes of low-level radioactive waste from a reference PWR and from a generic fuel reprocessing plant were taken from the work performed by Battelle. (Sm)(Sc). The volume of waste from the PWR was 17,924 cubic meters, all of which would be suitable for disposal by methods currently practiced. Volumes of waste from the reprocessing plant were separated into TRU and non-TRU contaminated. The TRU-contaminated waste estimate was 4,600 cubic meters and was further divided into high, intermediate, and low-level categories. The non-TRU contaminated waste estimate was 3,100 cubic meters presumably all suitable for disposal by current practice.

To gain insight into what impact this waste volume will have on waste disposal capacity, a review of waste generated annually by reactors is made. Phillips (Ph) summarized this data through 1975 as:

	Average	High	Low
	(All in cubic meters)		
BWR	1,000 to 2,000	1,780	178
PWR	200 to 500	810	10

The average represents an assessment and projection of what volumes could be expected. It was also noted the volumes increased as the reactors aged, at least for the first few years of operation.

A comparison of annual volumes versus decommissioning volumes indicates that over its operating life, a reactor is expected to generate anywhere from about the same volume to as much as four times the volume that will be generated during decommissioning. This assumes that volumes from decommissioning BWR's will be about the same as for PWR's. Thus, while decommissioning will result in a significant increase in demand for waste disposal capacity, it appears that such demand will not be inordinately excessive. The non-TRU waste from reprocessing plant decommissioning is a factor of four lower and represents an insignificant additional burden. TRU waste from reprocessing represents a much greater problem in that suitable disposal methods do not currently exist.

A second approach at assessing the waste volume problem in decommissioning is to determine the trench volume that would be required if the waste is disposed of by shallow land burial methods. The estimated PWR waste volume would fill a trench of dimensions 9m wide by 9m deep by 220m long or 15m wide by 9m deep by 130m long. Given the spacing required between trenches, the decommissioning volume thus represents a land use commitment of about one acre per reactor. This would not appear to be an inordinate commitment of land, especially when compared to fossil fuel power demands. Thus, it is concluded that the low-level waste volume from decommissioning is not a serious problem nor a limiting consideration.

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MODELING AND ENVIRONMENTAL ASSESSMENT OF LAND DISPOSAL METHODS FOR LOW-LEVEL RADIOACTIVE WASTES

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ABSTRACT

The Environmental Protection Agency (EPA) is evaluating the potential environmental impact and risk to man from disposing of low-level radioactive waste (LLW) in the ground. A generic model is being developed to simulate the interaction of a variety of LLW waste types and land disposal methods and operations with the natural characteristics of the site and their potential impact on the environment. Shallow land burial will be used as a base case. This model will be an important tool in EPA's work to develop environmental standards for LLW. This paper presents the general objectives, information requirements, components, and pathways considered in the environmental assessment model and some considerations in developing the model.

INTRODUCTION

Since 1945, low-level radioactive wastes (LLW) have been disposed of principally by shallow land burial and, until 1975, it was believed that there would be "zero" activity released from the burial sites. (1,2) Studies by the Environmental Protection Agency (EPA) and Department of Energy (DOE) at several burial grounds have shown, however, that radionuclides can escape from the burial trenches to the uncontrolled environment. (3,4,5) In 1975, it was determined that, "the activity detected in the environment around the site does not create a public health hazard at this time". (3,5)

In 1979, we still can not realistically estimate what the environmental impact of LLW disposal will be in 50, 100, or 1,000 years, if temporary remedial actions at the burial site are halted. Nor can we advise others whether present and proposed disposal methods are environmentally acceptable.

The Environmental Protection Agency has issued draft environmental protection criteria for storage and disposal of all forms of radioactive waste. (6) EPA is now preparing environmental radiation protection standards for high-level radioactive waste (HLW) and spent fuel management (7) and will prepare a radioactive waste standards rationale document by mid-1979. (8)

Presuming the recommendations of the Interagency Review Group to the President are approved, EPA will develop a proposed standard for LLW by the

The environmental standard for LLW should comply with the environmental protection criteria. For compliance, (1) the health risk from disposing of LLW should not be unacceptable, (2) future generations should not be subjected to risks greater than present generations, and (3) the health risk should be further lowered as much as reasonable, taking into account economic and social factors. If the environmental criteria are met, it follows that the needs of NEPA will also be satisfied.

Benefit-cost analyses, estimated health risks, and comparison of alternative methods will all be considered during the development of the LLW standard. The focus of this paper, however, is on development of an environmental assessment model to simulate the health risks from disposing of LLW by a shallow land burial (SLB) method.

GENERAL APPROACH

EPA's Criteria for Radioactive Wastes (6) recommend that the environmental standard should be based on predetermined models and should examine the projected effectiveness of alternative methods of control. Since a site specific model will not satisfy all these requirements, a generic environmental assessment model is required to estimate the health risk from LLW disposal methods to support development of an environmental standard for disposal of LLW. The proposed generic model will be a dynamic simulation model which will include release transport, pathways and dose and health effect submodels. The release and transport models will consider a range of geological, hydrological, meteorological, and waste properties and characteristics which may exist at present and potentially acceptable future sites. We are currently developing the model and collecting relevant data. (Details of these procedures follow.)

A. Data Base

There is already an extensive data base on the transport of radionuclides in the environment. The data required for our generic environmental assessment model are summarized as follows:

1. Waste form and inventory
2. Hydrological and meterological data of the site
3. Hydrogeological and geochemical characteristics of the disposal site and waste
4. Hydrological, demographic, and land-use distribution of site
5. Transport of non-volatile radionuclides by surface and ground water and wind
6. Transport of gaseous radionuclides through trench cover and their consequent transport by wind
7. Hydrogeological characteristics of trench cover
8. Macro-porosity of buried waste
9. Environmental impact assessment of leachate evaporator
10. Summary of available and current environmental study results
11. Summary of current engineering and operations practises for disposal and their associated costs.
12. Potential biological pathway

In some cases, information needed for these categories is available from previous studies. Other data which characterize simple physical and chemical processes can be obtained from simplified laboratory models or experiments. However, some data can only be obtained from field observations at existing burial sites. The data available will be reviewed and where the information is not sufficient, supplemental data will be obtained from ongoing and proposed field studies by DOE, NRC and EPA at a number of existing shallow land burial sites. At the present time, it appears that additional information is needed on: gaseous and surface transport of radionuclides, trench cover characteristics, biological pathways, waste forms, and current engineering practices and costs.

The parameters required for the generic environmental assessment model will be obtained from the results of the statistical analysis of the site specific and general data described above.

B. Description of Model

The environmental assessment model consists of four submodels: a release model; a primary transport model; an environmental transport model; and a dose and health effects model. Figure 1 shows the general flow of radionuclide transport simulation.

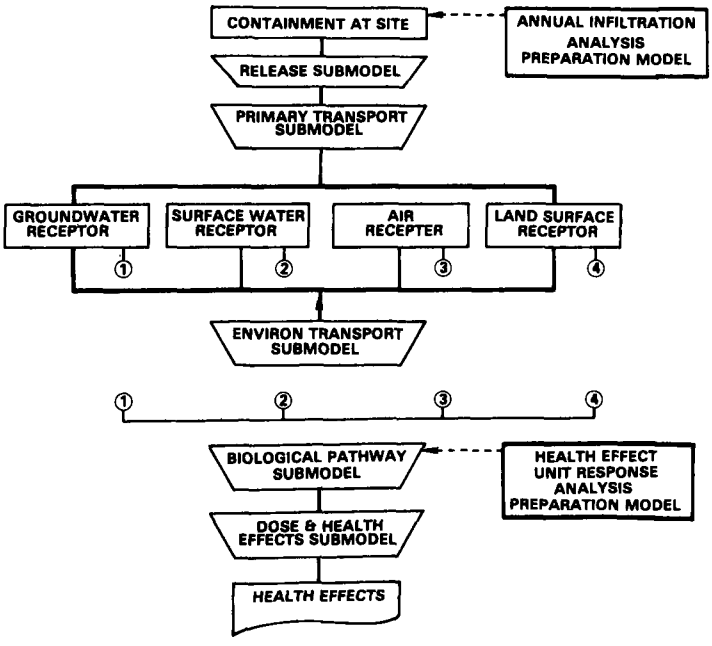

FIGURE 1 FLOW DIAGRAM FOR A GENERIC ENVIRONMENTAL ASSESSMENT MODEL FOR THE LAND DISPOSAL OF LOW-LEVEL RADIOACTIVE WASTES

The release model simulates the release of the radionuclides from the waste by a driving force, due to either natural processes or human activities. Water is considered to be the most important driving force to release radionuclides from the waste. The predominant release modes are expected to be leaching, erosion, and gas generation. The radionuclides released are then available for transport to environmental receptors through the primary transport model. The environmental receptors considered include groundwater, surface water, air, and land surface. The environmental transport model simulates the transport of radionuclides among the receptors. The resultant accumulation of radionuclides in the environmental receptors are then available for biological uptake by human beings through food chains, drinking water, inhalation, and direct irradiation pathways. The results of the pathway simulations then serve as input to the biological pathway model. The main environmental assessment model then integrates the total health effects for the total radionuclide uptake into humans through all critical pathways, both for individuals and for population.

C. Model Development and Analysis

The analyses will be accomplished by a system model which includes a main model and two independent preparation models. The main model is designed to accept inputs from the direct interpretation of collected data and from the simulation results obtained from the preparation models and to simulate the health effects therefrom.

The two preparation models are designed to simulate respectively (1) the synthesized hydrological conditions for a normalized driving force (infiltration) analysis and (2) the total number of health effects per unit intake of each radionuclide from each environmental receptor. The relationship between the main program and the preparation models is also shown in Figure 1.

The environmental assessment analysis will be conducted for SLB and for alternative land disposal systems. The scenarios described in the following section will be used as input to the established system model to simulate the health effects resulting from a disposal system implemented at a postulated generic site as a function of time. Use of these health effects in benefit-cost analyses are beyond the scope of this paper and are, therefore, not covered.

VIABLE POTENTIAL DISPOSAL SYSTEMS

A. Shallow Land Burial - The Base Case

Shallow land burial (SLB) is the method currently used for disposing of most LLW in the United States and, it is believed, SLB will continue to be used for disposal of LLW in the future. Since there are extensive data and practical experience available with SLB, it will be used in EPA's environmental assessment and standards development studies as a base case from which to compare other disposal alternatives.

In its most basic form, disposal of LLW by SLB consists of excavation of a large open-cut trench in the ground, placing wastes therein, and covering the wastes with the earth excavated thereform. As such, SLB can be treated and analyzed as a disposal system in which (1) the form of the

waste, (2) the hydrogeology, meterology, and environment of the site, and (3) special emplacement and protective engineering practices all perform and interact together to provide varying degrees of radionuclide retention. The degree of retention (i.e., performance) by the disposal system can be improved by changing one or more components of the system. Ideally, the SLB site, or system, would retain all the radionuclides until they have decayed to innocuous levels.

At present, there appear to be three viable variations of the SLB method: conventional SLB; improved SLB; and intermediate-depth SLB. Each method has its own unique engineering practices which result in its own associated costs; it may also, have different retention capabilities. A brief description of the three SLB methods follows.

Conventional SLB: This is the most basic and commonly used disposal method to date. Largely untreated wastes are placed in the trench and are covered with about one meter of earth. It is the least expensive disposal method and, in most cases, offers the least protection to the biosphere. In humid climates, precipitation can penetrate the earthen trench cover, leach radionuclides from the waste, and potentially release radionuclides to the environment.

Improved SLB: In this method, one or more components of the disposal system are modified to improve the performance (retention) of the system, or site. For example, converting a soluble waste into a solid with low leachability would retard the leaching of radionuclides from the wastes and greatly reduce the potential release of radionuclides to the environment. In another example the wastes could be covered with 2-3 meters of earth instead of one meter to reduce infiltration of precipitation. This method, naturally, is more expensive than conventional SLB but, in return, offers better retention of radionuclides and consequent environmental protection.

Intermediate-Depth SLB: In this method, wastes would be placed in trenches at least 10 meters below land surface and the trench would be backfilled to slightly above ground level. Other improvements in waste form and in site engineering such as suggested for improved SLB could be made, also. However, the main intent of this method is to reduce infiltration of precipitation and accessability of the wastes from human intrusion. This method is, also, more expensive than the other two SLB methods but, in return, it offers much more environmental protection. Shallow ground-water tables such as found in the eastern U.S. and the availability of geologic formations in which the open-cut trenches will remain open to depths of 18 meters or more without collapsing will limit the application this method.

B. Pathways and Scenarios for Shallow Land Disposal

In applying a generic approach to our environmental assessment modelling to SLB, it appears that three basic scenarios will cover the most sensitive parameters for radionuclide retention. These are: (1) an arid zone site for which no particular regard is given to the permeability of the disposal medium (because of a lack driving force or water); (2) a humid zone site with low-permeability disposal medium (which would fill like a "bathtub" in the event the trench cap leaked); and (3) a humid zone site with a moderately permeable disposal medium (which would allow water infiltrating through the trench cap to leak out the bottom like a "sieve"). [illegible] will be developed by altering the input parameters.

It is anticipated that all of the components identified earlier as making up the generic land "disposal system" will be present for SLB and all of the pathways involved in radionuclide transport will be considered in each of these three scenarios. The water pathway would certainly be less important for conventional SLB and improved SLB in the arid climates, whereas, all of the pathways would be important for these methods in the two humid climate scenarios. In intermediate-depth SLB (10-18m deep), the ground-water pathways would be considered most important.

A summary table of the transport and biological pathways to man which will be considered is given in Table 1.

TABLE 1. TRANSPORT AND BIOLOGICAL PATHWAYS TO BE CONSIDERED

TRANSPORT PATHWAY	TRANSPORT MEDIUM	REVEISING RECEPTORS	BIOLOGICAL PATHWAYS	INDIVIDUAL DOSE ASSESSMENT	POPULATION DOSE ASSESSMENT
GROUNDWATER TRANSPORT	GROUNDWATER	GROUNDWATER & SURFACEWATER	DIRECT & INDIRECT INGESTION	YES	YES
LEACHATE OVERFLOW	SURFACEWATER	SURFACEWATER	DIRECT & INDIRECT INGESTION	YES	YES
WASTE EROSION	SURFACEWATER	SURFACEWATER	DIRECT & INDIRECT INGESTION	YES	YES
GASSEOUS EMISSION	ATMOSPHERE	AIR	INHALATION	YES	YES
RESUSPENSION	ATMOSPHERE	AIR	INHALATION	YES	YES
DESORPTION	SURFACE AND/ OR GROUNDWATER	SURFACE AND/ OR GROUNDWATER	DIRECT & INDIRECT INGESTION	YES	YES
DIRECT EXPOSURE	---	---	DIRECT EXPOSURE	YES	NO
HUMAN INTRUSION	ATMOSPHERE	AIR	INHALATION	YES	NO

C. Viable Alternative Disposal Methods

As noted earlier, SLB is being analyzed as the base case for the disposal of LLW. However, some wastes may not be suited for disposal by the SLB method by virtue their specific activity, half-life, or chemical or physical character, or cost of preparation. In such cases, alternative disposal methods may be required or more cost-effective. An environmental assessment of these alternatives will be required to simulate and compare the alternatives.

On the basis of present information, much of which is of a preliminary nature, the following seven disposal methods appear to be viable and suitable for the disposal of one or more major types of LLW; deep geological disposal; deep well injection; hydrofracturing; engineered surface storage; ocean disposal-on-sea-floor; and ocean disposal-beneath-sea-floor. Brief descriptions of these potential alternatives follow.

Deep Geological Disposal: LLW can be emplaced in mined, solution, or explosion cavaties in geologic formations more than 100 meters deep(8). This method has already been studied in some detail for the disposal of non-radioactive hazardous wastes, transuranic wastes and HLW. It is suitable for solids or solidified liquids and for a wide range of specific activities and half-lifes which require a higher degree of containment.

Deep-Well Injection: The disposal of liquid wastes by injection into deep geological formations has been used widely for more than 40 years (more than 200 hazardous waste wells, 40,000 brine injection wells, and several LLW wells) (9,10). Liquids are injected through specially constructed wells under controlled pressure into permeable formations or saline aquifers at considerable depths. This method appears to be a viable alternative to certain liquid LLW presently being solidified for disposal in shallow land burial sites.

Hydrofracture: Hydrofracturing is widely used in the petroleum industry to stimulate the recovery of oil. This technique has been adapted for LLW disposal at the Oak Ridge National Laboratory(11). In a demonstration at ORNL: (a) fractures were opened along planes essentially parallel to the bedding in the Conasauga Shale by hydraulic pressure applied through an injection well at selected zones; (b) pumping a propping agent of sand and a gelling agent under pressure into the open fractures; and (c) then breaking the gel by injection of a special compound to open fractures. A grout slurry, containing cement, radioactive waste, and clay was then injected into the open fractures and solidified as grout sheets parallel to bedding in the impervious shale. Limitations of this method include separation of the liquid radioactive waste from the grout during emplacement and determining whether the grout is emplaced horizonally.

Engineered Surface Structures: Engineered surface structures, such as those described by Morisawa et al (12), would be capable of containing as much LLW as would be produced in the next 20 years in Japan. France already uses another form of surface storage for disposal of certain LLW (13). The Atomic Industrial Forum has also recently completed extensive studies on the on-site storage of LLW in engineered structures at power stations (14) and has found it to be a feasible but expensive management method. This method would normally be limited to very short-lived radionuclides unless removal was anticipated.

Ocean Disposal-On-Seafloor and Ocean Disposal-Beneath-Seafloor: LLW could be deposited (a) directly on the ocean floor or (b) in open-cut trenches in the sediment of the ocean floor and then covered. The seafloor sediments have relatively high cation exchange capacities and provide radionuclide retention capabilities as an additional barrier to the migration of radionuclides. Seafloor disposal is an alternative disposal method in which additional advantages may exist in the presence of a water barrier and more remote isolation between the waste and the biosphere than exists in SLB.

It can be seen from an examination of the gross characteristics of the various alternatives that the wastes are disposed of into deeper geologic formations at depths greater than 100 meters except for the engineered surface structure and ocean disposal methods.

Those disposal methods in which wastes are disposed of at depths greater than 100 meters include: deep geological disposal, deep well injection, and hydrofracturing. It is believed that the environmental assessment model developed for disposal of high-level wastes in a deep geological formation would, to some extent, be useful and adaptable to simulate the impact of LLW disposal by these alternatives. With the exception of operational spillage, the near-surface pathways such as surface water, atmosphere, soil, biological, and erosion, would be significantily less important. The ground water and intruder pathways would assume primary importance.

Environmental assessment of ocean disposal will require a considerably different model.

CONSIDERATIONS IN IMPLEMENTING THE MODEL

Development of an environmental assessment model to support a LLW standard will not be simple. There are a number of factors such as timing, philosophy, national affairs, and the state of technology development which will affect the depth and accuracy of analysis. These are discussed briefly below.

Time Constraint: EPA must develop a proposed environmental standard for LLW by the end of FY 1982. To be useful, the environmental assessment modeling and analysis must be completed by early FY 1982.

Data Available: In some cases, the data required for simulating important processes are not readily available (i.e., leaching of wastes, radionuclide transport through unsaturated soil, etc.). In other cases, it will be necessary to use best estimates of combined field and laboratory data.

Unusual Period of Analysis: The model will attempt to analyze hydrogeological and radiological processes which cover 100's and 1,000's of years. The scientific and engineering professions involved are not experienced in making predictions over such long periods of time.

Complexity of the Model Limited: The model must be simplified to some degree because (1) it must analyze both the fast and the slow responses of very complex hydrogeological, ecological, and radiological processes and (2) the costs of computer time would be prohibitive if a sophisticated model is used to make an environmental analysis of a disposal method such as SLB when many possible variations in parameters and consequent changes to performance must be considered.

Caution Required in Using Output: It must be clearly realized that the model may produce estimates which may be in error by orders of magnitude because of the long periods of time involved, the many unknown parameters, and uncertainty in the reliability of certain data. Therefore, output from the model must be used with caution and good judgment.

Benefits and Necessity of Model: Although the environmental assessment model will have limitations, it's use is both benefical and necessary. It forces us to organize our thoughts to consider the total disposal system and it helps us to identify our real data needs. Also, modeling is, to our knowledge, the best tool to compare and estimate the impact of a large complex system such as a SLB facility.

In summary, it is believed that development of an environmental assessment model is necessary; that the model must be relatively simple; and that we must use caution and common sense to successfully apply it.

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THE USE OF A RISK LIMIT AS AN ENVIRONMENTAL SAFETY STANDARD FOR RADIOACTIVE WASTE DISPOSAL SITES*

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Abstract

This paper demonstrates how the recommendations of ICRP Publication 26 may be applied to setting environmental radiation standards for radioactive waste disposal sites. Traditionally, such standards prescribe the allowable radiation dose to the maximally exposed offsite individual. Dose limits are usually established for the total body and for individual organs. In this paper, the risk factors recommended by ICRP for individual organs and the doses to those organs are combined to calculate the total risk per unit of ingested radioactivity. The allowable ingestion of radioactivity is then calculated from ICRP's individual risk limit. When these data are compared to normally derived ingestion limits, significant differences appear whenever a relatively insensitive organ receives the majority of the dose. Maximum allowable concentrations of radionuclides in water are derived from the ingestion limits and the concept of an effective water consumption rate. Using risk assessments in environmental standards for waste disposal sites would allow 1) a rapid and conservative (cautious) assessment of the potential health impact of a waste disposal facility, and 2) a simpler evaluation of the impact of ingesting several radionuclides even if each radionuclide affects different human organs.

Introduction

The International Commission on Radiological Protection (ICRP; ICRP 77) and the U.S. Environmental Protection Agency (EPA; EPA 78) have recently proposed the incorporation of risk assessments into the process of setting environmental health and safety standards. In this context, risk is a combined measure of the probability and severity of health effects which may result from doses of radiation. At low doses and dose rates, a linear, zero-intercept regression of observed incidence rates of health effects versus dose is the basis for calculations of probabilities. Although the observed health effects occur only at high doses and dose rates, the probabilities are assumed to be accurate at low doses and dose rates. The resulting model is not, strictly speaking, a predictor of environmental health impact, but rather an analytical aid for the comparison of alternative regulatory strategies. The term health effect normally refers to a mortality which results from neoplastic disease or to a mortality or severe morbidity arising from genetic defects. The risk factor is defined to be the

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number of health effects per unit of radiation dose and per person-year at risk.

ICRP Publication 26, (ICRP 77) recommends an upper limit of 10^{-5} to 10^{-6} health effects per person-year at risk for the hazard associated with non-occupational doses of radiation. The risk limit applies to the sum of risks from annual external exposures and 50 yr internal dose commitments; the risk which results from nonuniform exposures is the sum of the risks to each sensitive organ or tissue. This system of risk assessment provides an appropriate framework for calculating the potential effects of radionuclide ingestion. The determination of acceptable levels for this risk, however, requires a political value judgment rather than a scientific evaluation of facts. The former is beyond the scope of this paper.

The EPA (EPA 78) acknowledges that acceptable levels of risk are not as dependent upon statistical considerations as upon society's perceptions of the benefits and risks of radiation exposures. The proposed radioactive waste criteria hold that assessments of risk are key elements in the selection of waste management alternatives. EPA has not proposed numerical standards for environmental and public health risks associated with radioactive waste disposal.

This paper demonstrates how secondary standards for environmental radioactivity in the vicinity of waste disposal facilities might include analyses of risk, discusses practical difficulties encountered in implementing ICRP's method of risk assessment, and proposes a simple scheme for deriving operating limits useful in environmental assessments of radioactive waste disposal facilities. Using an arbitrary nonoccupational dose limit for radioactive waste disposal facilities, the risk factor for each radionuclide may be converted to a maximum allowable intake (secondary standard), which is in turn proportional to a maximum concentration in ground or surface waters (operating limit). These operating limits may be used to evaluate sites and disposal methods or to compare radioactive waste management alternatives.

Due to the state of knowledge concerning several key parameters, the contents of this paper should be considered as an example of a methodology rather than a recommendation for standards or limits.

Method

We selected 12 radionuclides (^{3}H, ^{51}Cr, ^{54}Mn, ^{59}Fe, ^{57}Co, ^{59}Co, ^{60}Co, ^{65}Zn, ^{131}I, ^{134}Cs, ^{137}Cs, ^{239}Pu) for consideration on the basis of their significance in low-level and high-level waste or due to inhomogeneities of internal dose distribution that enhanced the value of this demonstration. A compilation of radionuclides which are significant for waste management might also include ^{14}C, ^{90}Sr, ^{99}Tc, ^{129}I, ^{226}Ra, ^{230}Th, ^{232}Th, ^{237}Np, ^{240}Pu, ^{241}Am, and ^{243}Am. However, our purpose is not to perform impact assessments of specific disposal facilities but to suggest a method of integrating risk assessments into the derivation of secondary standards and operating limits and to discuss practical aspects of implementing ICRP's recommendations (ICRP 77). For similar reasons, we only treat ingestion of radionuclides. We calculated the average 50 yr dose equivalent commitment (hereafter called "dose") per pCi of ingested radioactivity to 11 organs (Table 1) of significance in risk analysis (ICRP 77). Each dose calculation consisted of two parts: the dose to a target organ from radioactivit

in that organ ($D_{t\to t}$) and the dose to a target organ contributed by penetrating radiations whose source is a different organ ($D_{s\to t}$). The former calculations generally employed the method of ICRP Publication 2 (ICRP 57), the latter are based on Monte Carlo calculations (MIRD 75). All bone marrow calculations, however, are based upon Monte Carlo calculations (MIRD 75). The quality factor for ^{3}H beta particles in these calculations is 1.7; this prevents a potential conflict in comparing these results to models used by NRC (NRC 77). Some parameters for biological uptake and removal rate in organs were not available to us. In these cases we assumed an organ or tissue uptake proportional to the uptake by the soft tissues of standard man (ICRP 75). Biological removal rates that were unknown were conservatively estimated to yield high ratios of dose per pCi ingested.

Monte Carlo calculations of doses to an organ due to penetrating radiation in another organ were calculated from the relationship

$$D_{s\to t} = D_{s\to s} \quad (S_{s\to t}/S_{s\to s})$$

where $D_{s\to t}$ and $D_{s\to s}$ are the doses from radioactivity which is external or internal to the organ and $S_{s\to t}$ and $S_{s\to s}$ are the appropriate ratios of absorbed dose per unit cumulated activity. The S factors have been tabulated for many radionuclides and combinations of source and target organs (MIRD 75). Since S is defined in units of dose per unit cumulated activity, the ratio $S_{s\to t}/S_{s\to s}$ is simply the ratio of dose to the target organ from a different source organ divided by the dose to the source organ from radioactivity in the source organ. Multiplying this ratio by $D_{s\to s}$ gives the dose to the target organ from radioactivity in the source organ.

Since we only considered ingested radioactivity, and since the lower large intestine (LLI) has the greatest cumulated activity, it was the only organ treated as a source of external exposure to other organs. The upper large intestine (ULI) and the small intestine (SI) may also have appreciable cumulative activities. These organs were not considered as sources of penetrating radiation for other organs because we achieved the purpose of our demonstration - to show that $D_{s\to t}$ may be significant - using only dose contributions from the LLI. Additional doses from the ULI and SI would not qualitatively change our results. For relatively insoluble radionuclides, this method will approximate the correct external organ dose since a large fraction of the cumulated activity will occur in the LLI. For soluble radionuclides, the cumulated activity is divided among all target organs and the doses from radioactivity within the organ will be considerably larger than doses from penetrating radiation which is emitted in other organs. Hence in the case of soluble radionuclides, the ratio $D_{s\to t}/D_{t\to t}$ will be low and the inclusion of the effects of cumulated radioactivity in the ULI and SI would not significantly alter the results.

Since the ratio $S_{s\to t}/S_{s\to s}$ will obviously vary according to the mean path or range of the radiation produced by each nuclear transition, we corrected this ratio whenever the error would otherwise exceed 10%.

While these approximations are adequate for the present demonstration, the process of standard setting or impact evaluation would require more rigorous derivations of the dose conversion constants.

The organ specific risk parameters (Table 1) are proportionality factors which relate the sensitivity of specific organs for radiation induced stochastic effects to the total risk that results from whole body exposures (ICRP 77). The risk factors specific to the gonads and breast are averages for both sexes; cancer cure rates are included in the calculation of risk to breast and thyroid. The product of the organ specific risk factor and the organ specific dose factor may be summed for all 11 organs to yield the total stochastic risk per pCi ingested. The details of this calculation are presented in the case of ^{58}Co (Table 1). The effective whole body dose equivalent is the ratio of the risk/μCi ingested divided by the risk/mrem under conditions of uniform irradiation. In the case of insoluble ^{58}Co, this is 2.6 mrem/μCi (Table 2). That is, ingesting one μCi of insoluble ^{58}Co is equivalent in terms of risk to a uniform whole body dose of 2.6 mrem.

Risk assessments will therefore be systematically incorporated into standards and criteria for waste disposal if the primary standards are given in units of effective whole body dose equivalent. ICRP's recommendation of a maximum nonoccupational risk due to radiation exposures (10^{-5} to 10^{-6} yr^{-1}) is equivalent to an effective annual whole body dose of 7 to 70 mrem. Assuming for the sake of these calculations that 7 mrem/yr is the primary standard, the maximum allowable intake of ^{58}Co is 2.7 μCi/yr (Table 2). The use of 7 mrem/yr in this paper does not constitute advocacy of such a standard, but merely the adoption of a convenient bench mark.

Operating Limits

For a given radionuclide, the maximum allowable intake for nonoccupationally exposed individuals is inversely proportional to the dose per μCi ingested and proportional to the maximum allowable dose. This intake, expressed in μCi/yr, may then be related to an operating limit, or maximum concentration in ground or surface waters. Assuming we are concerned with intrusion of radionuclides into potable or irrigation water and subsequent ingestion by humans (Figure 1), the yearly radionuclide intake from any pathway will be proportional to the concentration of radioactivity in the source of contaminated water. Hence the maximum allowable concentration in water of radionuclide i (W_i) is

$$W_i \ (\mu Ci/\ell) = \frac{7 \ (mrem/yr)}{R_i \ (mrem/\mu Ci) \ \Sigma_j \ f_{ij} \ U_j \ (\ell/yr)}$$

where f_{ij} is the ratio of the concentration of radionuclide i in the material which is ingested in pathway j to the concentration of radionuclide i in the drinking or irrigation water (μCi/kg ingested per μCi/ℓ of water); R is the effective whole body dose equivalent per ingested μCi of radionuclide i and U_j is the rate at which material j is consumed (kg/yr). The term $\Sigma_j \ f_{ij} \ U_j$ may be viewed as an effective rate of water consumption; f_{ij} is then a weighting factor specific to each combination of radionuclide and pathway.

Assuming that a human population has a single source of drinking and irrigation water, that all vegetables and fruit are cultivated by irrigation, that drinking water is unfiltered, and that meat and milk are produced from herds raised on irrigated pastures, we selected values of f_{ij} and U_j that are appropriate for an average adult (NRC 77, Ba 76, Ei 73, Table 3). For ^{137}Cs and ^{239}Pu, this model yields the following relationships between allowable radionuclide concentrations in water and the effective whole body dose per unit ingestion to an average member of the nonoccupational population.

$$W\ (^{137}Cs) = 4.1 \times 10^{-4}\ R_{Cs}^{-1} \quad (\mu Ci/\ell)$$

$$W\ (^{239}Pu) = 2.5 \times 10^{-3}\ R_{Pu}^{-1} \quad (\mu Ci/\ell)$$

Results

Table 2 lists the effective whole body dose equivalent per unit ingestion for the 12 radionuclides considered here and compares the secondary standards derived according to limits on effective whole body dose with secondary standards calculated according to the dose commitment to critical organs. The ratio of the latter to the former indicates the degree to which uniform limits for whole body and critical organ doses result in unequal assessment of risk.

The limiting concentrations of ^{137}Cs and ^{239}Pu in water supplies are

^{137}Cs: 1×10^{-5} $\mu Ci/\ell$

^{239}Pu: 1.8×10^{-5} $\mu Ci/\ell$

under the assumptions of our scenario.

Discussion

The risk to each organ which was calculated in the case of ^{58}Co (Table 1) indicates that the critical organ does not necessarily bear the majority of the stochastic risk. For ^{58}Co, the total risk is 3.7×10^{-7} μCi^{-1} and the risk to the LLI is 1.5×10^{-7} μCi^{-1}. In particular, the risk to the gonads is approximately 0.8×10^{-7} μCi^{-1}, in spite of the fact that ^{58}Co is not readily absorbed by the small intestine. The reason: irradiation of the gonads by ^{58}CO in the LLI produces approximately 80% of the total dose equivalent which is committed to the gonads by ingestion of ^{58}Co. This phenomenon also occurs when other relatively insoluble radionuclides which emit penetrating radiation are ingested.

The preliminary nature of these results must be emphasized. We did not calculate external organ dose to the gonads from ^{58}Co in the SI or ULI. Moreover, we acknowledge that refinements in uptake, distribution, and "S" factors, the ratios of dose commitment to cumulated activity in an organ (MIRD 75), may change these results. Nevertheless, we feel that these data indicate a more thorough understanding of external or "between-organ" doses may be necessary before a complete risk assessment is possible in the case of environmental impact assessments associated with radioactive waste disposal. These results also indicate that placing equal limits on whole body doses and organ dose commitments will not produce standards which equalize the risks from each radionuclide. For ^{60}Co, the allowable intake calculated from considerations of the risk to specific organs is 6 times greater than the allowable intake calculated according to the critical organ concept.

Although accurate internal dosimetry calculations are crucial to the environmental impact appraisals, the final results cannot be more accurate than the accuracy of the intake data. Table 3 indicates that the consumption of fish is the most important route for ingestion of ^{137}Cs. However, the bioaccumulation factor and the yearly ingestion rate are, in general, poorly known. Moreover,

organ doses and hence risk, may vary significantly according to age. Usage factors may vary according to geographical or socioeconomic factors. Agricultural methods may affect the values of f_{ij}.

The calculation of an effective water consumption rate and the weighting factors, f_{ij}, allows a rapid assessment of the relative importance of the ingestion pathways and provides a focal point in the environmental pathway analysis where an operating limit may be set. Although the primary limits will be based on dose to humans, the derived concentration limit for water provides a convenient design basis for radiological engineers charged with selecting the operating characteristics of a repository or shallow burial site. The present MPCs for continuous occupational exposure to water do not serve this purpose adequately. If only human drinking water were to be considered in the case of ^{137}Cs, the term $\Sigma_j f_{ij} U_j$ would be equal to 370 ℓ/yr. By including the other ingestion pathways, the sum is 17,000 ℓ/yr, a 47-fold increase. Doubtless these results would vary widely from site to site. The precise chemical and physical form of radionuclides which are leached or dissolved from waste and enter ground waters may also cause significant changes in the estimated values of the parameters in these environmental pathways. Nevertheless, the methodology demonstrated in this report serves as a basis for performing environmental risk assessments and setting site-specific environmental limits for waste repository designs.

Summary

Risk assessments related to waste disposal require relatively thorough analyses of the environmental behavior of relatively few radionuclides. Present models of the impact of ingested environmental radioactivity provide calculations of internal organ doses. These dose assessments may be inadequate for thorough assessments of risk, in part, because penetrating radiation emitted in one organ is not considered in doses to other organs; age and uptake rate may also be factors. The number of organs and tissues included in the present models may also need to be increased.

Environmental standards based on effective whole body doses and critical organ dose commitments do not result in equivalent risks for all radionuclides.

The concept of an effective water consumption rate simplifies the process of deriving operating limits based upon dose limits and risk assessments. These operating limits may serve as design guidelines for radioactive waste repository engineers or they may serve to assist regulators in distinguishing between acceptable and nonacceptable repository designs.

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TABLE 1

Risk Parameters

Organ	Weighting Factor (W_T)	Risk x 10^{-9} ($mrem^{-1}$)	Risk x 10^{-8} from ^{58}Co ingestion (μCi^{-1})
Gonads	0.25	35*	7.8
Bone	0.03	4.2	4.5
Red Marrow	0.12	17	1.4
Breast	0.15	21	2.3
Lung	0.12	17	1.7
Thyroid	0.03	4.2	0.17
Lower Large Intestine	0.06	8.4	15.
Stomach, Small Intestine Upper Large Intestine, Liver	0.24	34	8.4
TOTAL	1.00	140	37

*Risk of genetic effect in first two generations

TABLE 2

Total Risk Coefficients and Allowable Intakes

(1) Radionuclide	(2) Effective Whole Body Dose Equivalent per Unit Ingestion (mrem/μCi)	Allowable Intake (μCi) (3) Standard: 7 mrem/yr Effective Whole Body Dose Equivalent	Allowable Intake (μCi) (4) Standard: 7 mrem/yr Whole Body Dose Equivalent or Critical Organ Dose Equivalent Commitment*	Ratio (3)(4)
H-3	0.10	69.	69.	1
Cr-51	0.19	37.	10.	4
Mn-54	2.6	2.8	0.50	6
Fe-59	5.7	1.3	0.21	6
Co-57	0.61	11.	1.6	7
Co-58	2.6	2.7	0.48	5
Co-60	7.0	1.0	0.17	6
Zn-65	6.7	1.1	1.0	1
I-131	71.	0.10	0.01	10
Cs-134	40.	0.18	0.06	3
Cs-137	40.	0.18	0.10	2
Pu-239	140.	0.05	0.008	6

*I-131 intake calculated at 2 mrem/yr

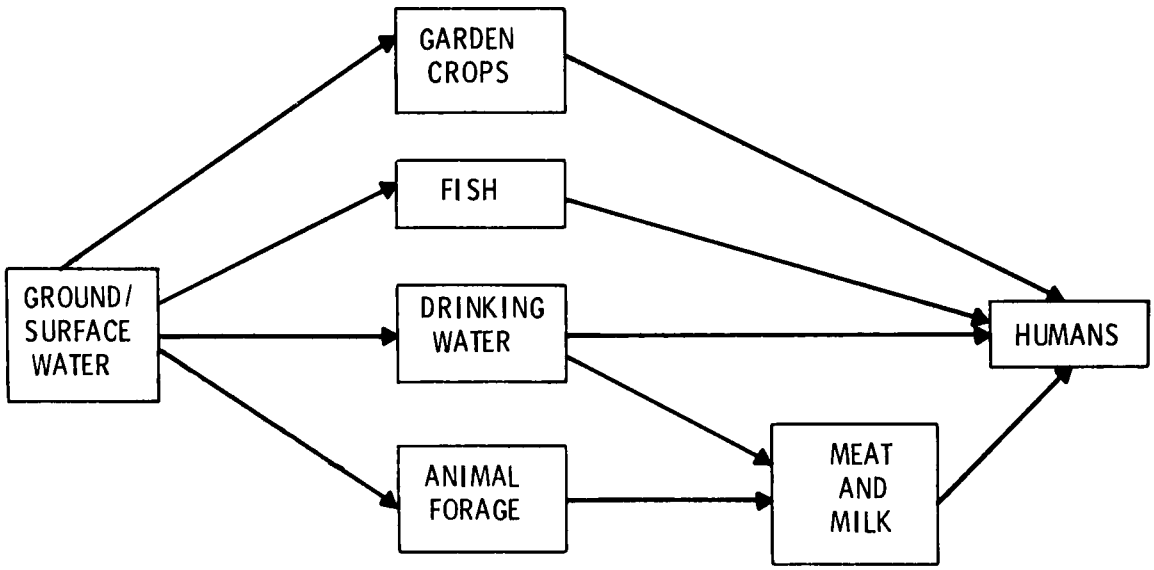

FIGURE 1

ENVIRONMENTAL PATHWAYS RELEVANT TO RADIOACTIVE WASTE DISPOSAL

PATHWAY	f_{ij} (ℓ/kg)		U_j (kg/yr
	^{137}Cs	^{239}Pu	
POTABLE WATER	1	1	370
FISH	2000	4	7
GARDEN CROPS	11	12	190
MEAT	4	0.9	95
MILK	3	< 0.01	110

ASSUMES 0.017 ℓ/m^2 hr OF IRRIGATION WATER; 9 MONTH GROWING SEASON; 100 yr ACCUMULATION OF ^{137}Cs; 1000 yr ACCUMULATION OF ^{239}Pu

TABLE 3

VALUES OF f_{ij} AND U_j FOR AN AVERAGE ADULT

RADON -- AN ENVIRONMENTAL POLLUTANT?

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Abstract

Radiological concerns with the disposal and use of mining and milling residues have heightened to the point that Federal agencies are asking or being asked to formulate new regulations for controlling radon daughters from a variety of sources -- radioactivity previously considered to be part of our natural environment. Based on information derived from epidemiologic studies of underground miners, particularly uranium miners, the health impact on the general public is being projected. Depending on the assumptions made, these projections vary widely. Because of these variations in health risks, decisions on control measures have even wider implications on economic and social considerations. Thus the question: Is radon an environmental pollutant? While not fully answering the question, recognizing the uncertainties in assessing and controlling radon daughters can put the question in better perspective.

Discussion

Sources of radon have become very much a part of the radiation protection scene. Up until a few years ago, most health physicists probably thought of exposures to radon and its daughters only in terms of exposures to underground uranium miners and the episode of Grand Junction with the permitted use of uranium mill tailings in occupied structures. Today, however, such a peripheral view is not possible and we must view this natural occurring -- human enhanced -- potential exposure source with more direct vision.

Evidence of this current direction is recorded in the passage of the Uranium Mill Tailings Radiation Control Act of 1978, the lengthy deliberations of the Nuclear Regulatory Commission regarding the inclusion of guidelines for radon daughter exposures in its "S-3" table for the uranium fuel cycle, and EPA's proposed actions under the Resource Conservation and Recovery Act related to "hazardous wastes." Radon daughter exposures play a predominant role in the decisions to be made in implementing regulations for these and other responsibilities. Yet, in reality, our knowledge of the biological effects associated with radon, of general environmental levels, of sources, of proper instrumentation and measurement procedures, and of the appropriate dosimetry to be used, is among the most lacking in all of health physics. The keystone to health physics has been dosimetry -- this allows us to do marvelous things with units and facilitates understanding problems on a common ground. However, for radon and its daughters, this common ground is so complex that many of us now prefer to go directly from how we describe

exposures, working levels, to risk of lung cancer, bypassing the rad/rem calculations.

I titled this brief discussion "Radon -- An Environmental Pollutant?", leaving it as a question so that each of you, after some thought, can provide your answers. What I intend to do now is to give you a brief on the problem so that your thoughts on an answer will be more than a "knee-jerk" response. In my opinion, too often the response has been that radon exposures are part of natural background and why attempt to do anything about it -- after all, we evolved in a natural radiation environment. I trust that you will at least give more thought than this reaction to the question.

On a risk basis, and I will quantitate this later, radon daughters represent the highest health risk of all radionuclides present in the atmosphere. The so-called natural background ambient outdoor levels of radon range from 40-1000 pCi/m^3, according to NCRP Publication No. 45 (NCRP75), with an average of about 100-200 pCi/m^3. The outside ambient background is about 0.001 WL, but highly variable. However, since most of us spend about 3/4 of our time indoors and the concentration can build up, the exposure levels we experience are greater than 0.001 WL and average about 0.004 WL (Ge78). The exposure level variations in residential structures can be considerable, perhaps even by as much as a factor of ± 10. Actually we have very limited information on "natural background levels" of radon and radon daughters, making any decisions using background as a reference point a very tenuous one at best.

But the determination of whether or not radon is an environmental problem cannot be answered using solely the range in natural background levels. The basis for this determination has to lie in how we project the risks from a given exposure to radon and its daughters. For this determination we must turn to a limited health effects base which exists in epidemiologic studies of miners, primarily uranium miners; studies which have been made in several countries. The most often quoted studies are of U.S., Swedish, Czechoslovakian, and Canadian miners.

While any of these studies can be criticized for one reason or another, it is rather accepted that the risk of lung cancer induction is a function of cumulative exposure (i.e., CWLM) and that linearity of this relationship cannot be totally rejected. It can even be generally agreed that, regardless of whether one uses the absolute model for risk estimates or the relative risk model, the risk estimates are within factors of two to three when such estimates are limited to miners. The absolute risk takes the <u>difference</u> between the risk in the irradiated population and the risk in the nonirradiated population, e.g., number of excess cases per 10^6 person years per WLM. Relative risk takes the <u>ratio</u> of risk between the risk in the irradiated population and the risk in the nonirradiated population, e.g., percent increase in cases per WLM.

The more serious problem arises when we take the observations for these populations and <u>have</u> to extrapolate to exposures of the general public. Here we run into a multitude of complexing factors of differences -- differences in sex, age distribution, breathing rates, smoking habits, in degree of radionuclide equilibria, other environmental factors, etc. These factors do not

act in one direction, e.g., slower breathing rates decrease the exposure, and younger ages have longer periods at risk; smoking goes both ways, increasing the promotion of lung cancer but perhaps decreasing the dose because of thickening of the bronchial epithelium.

Thus, in the end we are cautious on making such extrapolations, but they must be made because, regardless of how uncertain our health risk may be, the magnitude of the risk is large enough that the public health implications cannot be ignored. Drs. Ellett and Nelson of our program, in an analysis to appear in connection with our efforts to evaluate potential health impact of radon levels in Florida, have estimated this risk of increased fatal lung cancer to be between 1-5% per WLM on a relative risk model. This corresponds to lifetime risks of about 10^{-3}/WLM. Using an absolute risk model, an upper bound value of about 10^{-4}/WLM has been proposed by an ad hoc group of the Nuclear Energy Agency. I will not attempt here to evaluate the pros and cons of these estimates because in the end the differences derived are not terribly important to the regulatory process and, in my opinion, differences in risk of a factor of 10 are not major determinates for this problem.

Assuming a mortality rate, i.e., lung cancer deaths, of 3% per WLM, it can be estimated that the risk of lifetime exposure at 0.02 WL is about 1 in 50. More properly stated, this risk is about 2000 excess cancer deaths in a population of 100,000 persons exposed. The 0.02 WL value is given because this is a proposed recommendation being considered for the phosphate situation in Florida. Taking the 0.004 WL value given earlier for indoor background, we have about 400 cancer deaths attributable to radon daughters, in a total of about 2900 ($\sim$42/yr for 70 yrs), or about 10% of the total lung cancer deaths. If we take the NEA's absolute model derived value, we can account for about 1% of the "natural" incidence of lung cancer. In either case, the apparent contribution of radon daughters exposure to the current number of lung cancers is not trival. For perspective, consider that for a continuous lifetime dose equivalent rate of 25 mrem/yr, whole body external exposure, the risk estimate is 20-100 cancer deaths per 100,000 persons exposed.

While I recognize that in many instances emanations from a given source cannot be discriminated from the noise of background surrounding that source, a true public health protection role requires that we give this some attention and act prudently, especially for those who reside on or near the source and for which the practice of "as low as reasonably achievable" has obvious public health meaning. It is also true that, for such sources, we must put ourselves in a preventative frame of mind and not be in a corrective mode. Reasonable preventative measures are socially more acceptable and less costly than mandated corrective measures; and as an aside, a preventative frame of mind helps our creditability. In my own mind, I have answered the question -- YES!

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PROPOSED FEDERAL RADIATION GUIDANCE FOR RADIOACTIVE WASTES

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Abstract: The Administrator of EPA, in his role of providing Federal radiation guidance, will soon forward radiation protection guidance for radioactive wastes to the President for approval. The recommended guidance addresses: (1) the types of materials to be categorized as radioactive wastes and considered for control; (2) the use of institutional functions, engineered controls, and natural barriers to isolate wastes; (3) the potential health risks associated with wastes, and the factors involved in determining them; (4) factors which determine the unacceptability of various levels of risk; and (5) other considerations such as retrievability, site locations, and communication to succeeding generations to assure continued isolation. When approved by the President, the recommendations will guide Federal agencies in making policy, programmatic, operational, and standard-setting decisions in providing radiation protection for radioactive wastes.

Introduction

Federal radiation protection guidance is being developed to establish the basic radiation protection principles which Federal agencies should apply in the formulation of policies, plans, programs, standards, and other decisions involving the storage and disposal of all forms of radioactive wastes. The guidance would not apply to non-Federal sectors of our society except through regulatory or other actions by an affected Federal agency.

The recommended guides were developed in accordance with Executive Order 10831 and Public Law 86-373 (U.S.C. 2021(h)), which charge the Administrator to "...advise the President with respect to radiation matters, directly or indirectly affecting health, including guidance to all Federal agencies in the formulation of radiation standards and in the establishment and execution of programs of cooperation with States." The Department of Energy will provide and operate disposal facilities for several types of radioactive wastes. The Nuclear Regulatory Commission is preparing specific regulations for licensing such facilities in accordance with environmental protection standards which the Environmental Protection Agency will establish for the various types of radioactive wastes. Each of these Federal activities will be guided by the recommended guidance.

The Agency received considerable assistance from the public in developing this guidance through participation in two Public Workshops and a Public Forum. The bases for the guidance are being developed in a Federal Guidance Report No. 10 which reflects the comments received from

the Public Forum participants and from public review of the proposed criteria, which were published for comment in the Federal Register on November 15, 1978.

Radioactive Waste Materials

The National Environmental Policy Act of 1969 states that each generation has a responsibility "as trustee of the environment for succeeding generations." Some radioactive waste materials are both long-lived and highly toxic, and thus may pose substantial health risks to both present and future generations.

The primary factor in deciding that a material is a "waste" is the material's residual value. If it has no designated net value as a resource or a product or is impractical to control as such, the material should be considered a waste. Because these recommended guides are addressed to the treatment of radioactive wastes, as opposed to other radioactive materials with current or possible future uses, materials which may have such uses would not be considered wastes subject to the guidance. Depending on the circumstances, the judgment of the material's remaining value will be made by the person or organization possessing or controlling it, or by a government agency on behalf of the society as a whole. Once a material has been designated as a waste, whether or not it should be disposed of under this guidance depends on its radiological hazard and the capability and costs of controls.

The Agency stated in its proposed criteria that the requirement to keep radiation exposure as low as practicable precluded the establishment of a general "de minimus" level below which waste materials that are also radioactive would not be considered radioactive waste. Considerable public comment was received on this statement with the bulk of opinion being that such a level was required, and the application of the balance of the criteria as proposed would result in de facto levels of de minimus. This guidance is designed, as were the proposed criteria, to allow for various radioactive materials to be excluded for justifiable causes from additional control because of their radioactivity content, but these exclusions are to be determined on the basis of the particular circumstances involved using the principles established in the guidance. It is proper for Federal agencies to designate the materials that would and would not be subject to types of controls; in the Agency's judgment this approach should be used rather than attempting to define an arbitrary, general level of de minimus which may be overly restrictive for some situations or non-conservative for others.

Three broad classes of materials should be considered radioactive wastes for disposal purposes. The first class consists of those materials produced artifically from nuclear reactions or by fabrication from naturally radioactive materials into concentrated sources. These materials are directly controllable and their circumstances are readily identified and described.

A second class of substances contains diffuse radioactive materials of natural origin. However, these materials would be subject to control only if they could result in exposures greater than those which would have

occurred, through any pathway, prior to the disposal of the material. The rationale underlying this distinction is that requiring the material to be treated as radioactive waste implies that its potential radiological risk is a decisive characteristic in deciding its control. Yet if failure to treat the material as radioactive waste would not increase exposures over pre-existing levels, requiring disposal of the material as a radioactive waste seems unreasonable. If such an improvement is desirable, it should be for reasons other than that the material is a waste containing some natural radioactivity.

The final class of wastes which should be considered radioactive wastes for purposes of disposal under this guidance includes any retained waste materials which are prohibited by government regulatory action from unrestricted discharge into the general environment due to their radioactivity. While this category of substances will obviously contain many wastes covered by the two previous classes, certain materials fall into neither category. Regardless, the Agency believes the restriction on discharge to the general environment is in itself sufficient basis for requiring any such material to be considered for disposal in accordance with this guidance. Examples of such materials could include those removed from effluent streams at nuclear reactors or from the processing of ores which contain uranium or thorium.

Recommended Guide Number One designates broad types of materials in these three classes that should be regarded as wastes whose radioactivity content requires consideration in their disposal. Future study and information may lead to the designation of other materials in accordance with the principles discussed above. Usual methods of control of a material solely as waste may be used if they provide a degree of control consistent with this guidance.

Control of Radioactive Waste

Federal control of any radioactive waste should attempt to meet a fundamental goal of complete isolation over the period it would represent unacceptable risks to humans. Controls for radioactive wastes are of three general types: engineered barriers, natural barriers, and institutional mechanisms. Engineered barriers such as containers or structures generally can be considered only as interim measures for containment, despite the fact that some structures have survived intact through the ages. Stable geologic media with high retention characteristics are an example of natural barriers. Institutional controls are those which depend on some social order to prevent humans from coming in contact with wastes, such as controlling site boundaries, guarding a structure, land use policies, record keeping, monitoring, etc.

It generally is accepted that long-term isolation should depend on stable natural barriers. Institutional mechanisms can be very effective and are essential in the early stages of management of any waste, but for practical reasons they can be relied on for only limited periods. The appropriate time period for relying on institutional controls was discussed extensively during the developmental stages of this guidance since the issue is a matter of judgment. A maximum time period of 100 years was recommended for such controls to be depended upon with any degree of

assurance. Public comments recommended shorter times, longer times, and substitution of general goal statements; however, the Agency has not been persuaded that a different time period would constitute a better recommendation for reliance on institutional controls.

For wastes that represent unacceptable risks for longer than 100 years, Federal decision makers should, therefore, not approve radiation protection systems that are based primarily on restricting customary uses of land and of ground or surface waters for longer than 100 years. This does not mean that institutional controls are required for 100 years, or that they must stop at that point if society can still maintain them; only that people making the initial disposal decision should not plan on their use to maintain protection beyond 100 years.

Risk Assessment

The risk a waste poses over time is the pivotal part of any environmental and public health protection policy for radioactive wastes, and is the key consideration in deciding whether and how to store or dispose of the wastes. The term risk expresses a general concept encompassing both the probability of occurrence of adverse effects and their severity. Both aspects are basic to decisions on the acceptability of controls for radioactive wastes; however, there is no generally accepted methodology for determining the risk that society would accept in a given set of circumstances.

Risk determinations rest on a number of factors, especially the total amount of waste material at a particular location, its persistence due to form and concentration, its potential to enter the biosphere and produce adverse effects on individuals and populations, the effectiveness of various controls, and the inherent uncertainties of many of the parameters.

Determinations of risk are most useful in making public policy decisions if they estimate effects on health; however, projections of population size, land use, and human factors become very uncertain beyond a few hundred years. Because of the persistence of many wastes, however, it is recommended that health effects estimates be performed for at least 1000 years to consider both acute exposure risks and chronic exposure risks represented in many types of wastes. Some very significant waste materials persist well beyond 1000 years; thus, it is important that these be considered in choosing the best control alternative. Such comparisons can be made with health effects estimates based on very general assumptions beyond 1000 years; they can also be reasonably made by other parameters such as curies released, calculated doses to assumed individuals, etc. Public comments divided over this recommended degree of emphasis before and after 1000 years, principally, we believe, over perceptions that resulting risk estimates would be over- or under-conservative. The intent was, rather, that risk estimates be sufficient to recognize the potential level of risk and its persistence and yet be of a reasonable form for responsible decision-making. Reducing the rigor required for estimates beyond 1000 years was an attempt to consider both circumstances.

Examination of risks for waste disposal systems in terms of the consequences of exposure and their probability of occurrence will result in values greater than zero which vary with the types of waste, the controls available, and the kinds and severity of events that can be postulated to disrupt isolation. Consequences may range over chronic and acute exposures of a few individuals or large groups, with probabilities ranging from very remote to near certainty over the long term for some waste types and controls.

Risk Acceptability Considerations

All societies experience risks and have developed patterns of acceptance of various types of risks. For high-technology societies these patterns are difficult to discern, and they often change in relatively short time frames because such societies are continually creating new sources of risks. As a starting point, it appears that any present or future risks due to radioactive wastes would not be acceptable to society if reasonable controls that are available and economically feasible have not been used. Also, because of the responsibility each generation has to succeeding ones, a key social consideration is that at a minimum the current generation should not pose larger risks to a future generation than it would be willing to accept for itself. This does not mean that the risk has to be the same in future generations, only that it would not be unacceptable to the current generation if imposed on it -- even though the projected impact on the first few generations may be well below levels determined to be acceptable.

In general, risks are more readily acceptable to society if the consequences are common ones that are not highly dreaded, and are well understood situations which can be related to other common risks. On the other hand, generally beneficial circumstances which have high potential for harm appear to be acceptable to people if occurrence of the harm is virtually impossible; that is, the probability is very low. These interrelationships between probability of occurrence and consequence are not linear functions, and thus an unacceptable risk value can not be chosen as a product of the two. Since a broad range of circumstances is possible for wastes, a continuum of probability/consequence is possible; thus, it is only possible to give general guidelines for deciding when various levels of risk would be unacceptable. These are rooted in the basic concepts discussed above: risks due to events likely to occur will be acceptable only if adverse consequences are low and are of a common type; events with high adverse consequence potential must be virtually certain not to occur.

High consequence events resulting from waste disposal can be compared to those associated with productive, shorter term technologies such as dams, dikes, and large stores of toxic or explosive chemicals. High consequence events for radioactive wastes should be considered unacceptable unless their probability is only a small fraction of that deemed acceptable for shorter term productive technologies.

Exposure situations which are likely to occur for certain kinds of wastes would be unacceptable unless predicted effects would be of questionable significance, even on a statistical basis. This means that if

acute exposure is projected as a likely result of waste disposal, it should affect only a few individuals randomly. If large groups are likely to be chronically exposed, the projected risks should be small and no greater than comparable risks that society has already willingly accepted.

Each Federal agency that is involved in establishing policy for radioactive waste disposal will be responsible for estimating the consequences of likely radiation exposure circumstances for wastes, and for determining whether they are unacceptable based on an examination of their consistency with other established social values that have evolved for comparable circumstances.

Supplementary Protection Goals

A number of other subjects such as provisions for retrievability, choosing site locations, and passive communication to future generations may provide positive aspects for control of radiological hazards; however, their application may, in some instances, undermine the goal of providing permanent isolation for wastes. For example, it is difficult to maintain retrievability or conduct a monitoring program without compromising the ability to provide isolation. It is not appropriate to depend upon methods and techniques such as these or other similar ones for long-term control; nonetheless, when such methods could be reasonably applied to enhance overall protection from wastes, it is prudent to use them.

Provisions for retrievability of radioactive waste is desirable, if isolation is not compromised, in order to provide a mechanism for correcting losses in protection due to unforeseen circumstances. Wastes may also become future resources which has led some to argue that retrievability should be required for this possibility; however, it appears that such materials ought not be disposed of in the first place.

It is desirable to select sites if they are reasonably available where the action of natural forces over time such as erosion, sedimentation, and crystallization can be projected to improve, rather than reduce, isolation of the wastes over the time they may represent potentially unacceptable risks if they were to enter the biosphere. Such sites may not be practicably available, but if they exist among available alternatives they should be considered for use. Similarly, if geological media are used, they should reduce the effect of potential adverse interaction of the wastes with water to the greatest extent possible. Public comment on the proposed criteria discussed the desirability of choosing waste disposal locations away from valuable natural resources to reduce the likelihood of inadvertant destruction of the isolation capability of the site. Federal agencies should consider this aspect of siting when it is practicable to include it, all other features being equal.

In many disposal situations, the residual risk will mainly be attributable to potential failure mechanisms involving eventual intrusion by humans. Passive methods of communicating the hazard, such as markers which call attention to the waste, may sometimes be judged to provide a net reduction of risk. Other passive methods, such as creating records

describing the waste, or setting aside of the land title to the disposal site, have value in reducing the likelihood of intrusion for some limited time.

An example of a circumstance where land title transfer is reasonable is a current site that has been in use for some time where optimal environmental isolation is no longer a practicable alternative, such as an abandoned mill tailings site, a nuclear test facility site, etc. In these cases, Federal ownership of the land beyond the normal period of institutional control would be reasonable to minimize potential intrusion. Such decisions should be made on a case-by-case basis and provision for specifically treating such exceptions should be addressed in standards and regulations which are promulgated for these types of wastes.

Recommendations

Recommended guides have been developed which address each of the considerations presented above. It is intended that upon adoption, each of the following recommended guides would be applied, unless a specific circumstance is excepted in a guide, by Federal agencies in providing environmental and public health protection for radioactive wastes:

Recommended Guide No. 1: Radioactive materials should be considered by Federal agencies for control as radioactive wastes if they have no designated net product or resource value and: 1) are human-produced by nuclear reactions, fabricated from naturally radioactive materials into discrete sources, or as a result of regulatory activities are prohibited from uncontrolled discharge to the environment; or 2) contain diffuse naturally-occurring radioactive materials that, if disposed into the biosphere, would increase exposure to humans above that which would occur normally in pathways due to the pre-existing natural state of the area. Examples * of radioactive waste materials that should be subject to environmental protection requirements are:

a. All radioactive materials associated with the operation and decommissioning of nuclear reactors for commercial, military, research, or other purposes and the supporting fuel cycles, including spent fuel if discarded, fuel reprocessing wastes, and radionuclides removed from process streams or effluents.

b. Artificially produced radioisotopes, including concentrated radium sources, for medical, industrial, and research use and waste materials contaminated with them.

c. The naturally-radioactive residues of mining, milling, and processing of uranium and phosphate ores.

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The materials listed should be subject to this Federal radiation protection guidance even though some such materials may not upon examination require any control above that they would receive as ordinary wastes; Federal agencies may also designate other radioactive materials for consideration if they are found to satisfy this guide.

Recommended Guide No. 2: Radioactive wastes should be controlled to meet a fundamental goal of complete isolation over the period they represent unacceptable risk. Controls which are based on institutional functions should not be relied upon for longer than 100 years to provide such isolation; radioactive wastes that represent unacceptable risk in excess of 100 years should be controlled by engineered and natural barriers.

Recommended Guide No. 3: Radiation protection requirements for radioactive wastes should be based primarily on an assessment of risk to individuals and populations; such assessments should examine at least the following factors:

a. The amount and concentration of radioactive waste in a location and its physical, chemical, and radiological properties;

b. the projected effectiveness of proposed alternative methods of control;

c. the potential adverse health effects on human individuals and populations for a reasonable range of future population sizes and distributions, and of uses of land, air, water, and mineral resources for one thousand years, or any shorter period of hazard persistence;

d. estimates of environmental effects using general parameters or of health effects based on generalized assumptions for as long as the wastes pose a hazard to humans when such estimates could influence the choice of a control option;

e. the probabilities of releases of radioactive materials to the general environment due to failures of natural or engineered barriers, loss of institutional controls, or intrusion; and

f. the uncertainties in the risk assessments and the models used for determining them.

Recommended Guide No. 4: Any risks due to radioactive waste storage or disposal activities should be deemed unacceptable unless it has been justified that the further reduction in risk that could be achieved by more complete isolation is impracticable on the basis of technical, economic, and social considerations; in addition, any method of control should be considered unacceptable if:

a. radiation risks to a future generation are greater than those acceptable to the current generation;

b. probable events could result in adverse consequences greater than those of a comparable nature generally accepted by society; or

c. the probabilities of highly adverse consequences are more than a small fraction of the probabilities of high consequence events associated with productive technologies which are accepted by society.

Recommended Guide No. 5: Certain additional procedures and techniques should also be applied to waste disposal systems which otherwise satisfy these guides if their use provides a reasonable net improvement in environmental and public health protection. Among these are:

a. procedures or techniques designed to enhance the retrievability of the waste;

b. selection of waste disposal locations which take advantage of natural processes to enhance isolation over time and minimize unintentional disruption due to resource recovery; and

c. passive methods of communicating to future people the potential hazards which could result from an accidental or intentional disturbance of disposed radioactive wastes.

Implementation and Follow-Up

It is expected that each Federal agency will use the recommended Federal radiation protection guides as a basis for developing detailed radiation protection requirements for radioactive wastes which are consistent with its particular responsibilities. The Agency, in cooperation with other Federal agencies, will follow the implementation of these recommended guides, and will promote the coordination necessary to achieve an effective Federal radiation protection program for radioactive wastes. Periodically, the Agency will interpret and expand upon each of the recommendations as required to assure effective implementation by Federal agencies in accordance with new and changing information.

SESSION E

ENVIRONMENTAL - PUBLIC HEALTH ASPECTS I

Session Chairperson

S. V. Kaye

Oak Ridge National Laboratory

PREVIOUS MANAGEMENT PRACTICES FOR NATURALLY OCCURRING RADIONUCLIDE WASTES: CURRENT RADIOLOGICAL STATUS

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Abstract

Many installations used during the early days of the United States' atomic energy program have been released in recent years for unrestricted private uses. These installations include lands and buildings used for the storage of radioactive wastes resulting from refining and processing of uranium and thorium. Waste management practices at these sites in the 1940's and 1950's were not conducted with today's emphasis on as-low-as-reasonably-achievable (ALARA) principles. Consequently, many of these older waste storage areas are contaminated with naturally occurring radionuclides in concentrations which are orders of magnitude greater than those found ordinarily in the earth's crust. Current and potential elevated human exposures at fifteen of these sites are due primarily to radon daughters and external-gamma radiation. A wide variety of exposure conditions may be found at these sites — ranging from slightly above background to more than thirty times the guidelines recommended for the public. Remedial actions are contemplated for a number of these sites where contamination levels or radiation exposures exceed current guidelines.

Introduction

Early in this nation's development of atomic energy, extensive efforts were made to utilize all available resources for a program to demonstrate controlled nuclear fission. Initially, this program was administered by the Department of the Army under the Manhattan Engineer District (MED). Although conducted in wartime secrecy, the MED program encompassed a wide variety of materials research and development activities as well as various commercial source material handling operations. Contracts for needed services were entered and terminated as required.

Administration of the MED program was taken over by the Atomic Energy Commission (AEC) after the conclusion of World War II. Although the initial MED/AEC contractor facilities processed uranium, increased interest in the thorium fuel cycle resulted in a corresponding increase in the number of facilities involved in thorium processing. Services provided by private commercial firms under MED/AEC contracts covered a

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wide variety of activities such as ore transport and storage; dissolution and leaching of ores; production of mill concentrate (yellow-cake); refining of mill concentrate; conversion of this refined product to other compounds and/or metal; smelting, rolling, extrusion, cutting, and packaging uranium and thorium metal products for distribution to other institutions such as national laboratories; and the recovery of uranium from scrap and salvaged material.

Disposal of radioactive residues frequently consisted of shallow-land burial on-site or at some federally owned or leased land in the vicinity of the site. At the termination of contract operations, efforts were made to decontaminate buildings, land, and equipment to levels consistent with guidelines which existed at that time. However, no consideration was given to cleanup in accordance with ALARA objectives.

In many cases, present records are insufficient to document the radiological condition of these sites, most of which are now in the public domain. The overall program to determine the current status of these sites has been described elsewhere (e.g., Ha78, DOE78). This paper will present results found at three sites involved with waste disposal during MED/AEC activities; these results represent the range of findings obtained during the current resurvey program.

Early Solid Waste Management

The handling and processing of ores and ore concentrates produced large volumes of low-level solid residues at MED/AEC contractor facilities. Furthermore, solid wastes were generated when equipment and building surfaces were decontaminated or discarded. These wastes, particularly the residues, contained most of the radioactive material present in the original ore or ore concentrate. Consequently, these wastes are the source of present day radiation exposures at MED/AEC sites.

The highest priority items at MED/AEC sites, particularly during World War II, were the development of processing technology and the production of source material. Typically, a portion of the contractor's land would be dedicated to surface storage or shallow burial of process residues. If this proved to be impractical, a contract for these purposes would be established with a nearby property owner. Residues were generally regarded as wastes when the source material content was no longer recoverable by the contractor's processes. Thus, qualititative and quantitative aspects of the radionuclide inventory of residues were quite dependent on the processing history of the material.

Previous waste management practices are intimately associated with the present radiological status of fifteen former MED/AEC sites surveyed to date by the Oak Ridge National Laboratory (ORNL). The contamination of these sites by radionuclides such as ^{230}Th, ^{232}Th, ^{227}Ac, ^{228}Ra, ^{226}Ra, and ^{210}Pb indicates that problems could be associated with inhalation and ingestion of these long-lived materials. On-site measurements indicate that external gamma radiation exposures and, where

structures are present, indoor radon daughter concentrations constitute the principal radiation exposure problems at these formerly utilized sites (e.g., Le78a).

Radiological survey results indicate that present exposures range from those which cannot be distinguished from background to more than thirty times the guideline values recommended for the general public (Cr78, Le78a). These results are due to a wide range both in exposure rates and in site occupancy. Radiological survey results from three sites will be used to demonstrate the relationship between previous solid waste management practices and present radiation exposures at formerly utilized sites.

Former Vitro Rare Metals Plant, Canonsburg, Pennsylvania

The 8-ha (18-acre) site at Canonsburg, Pennsylvania, was used for the commercial extraction of ^{226}Ra from 1911 to 1922. From 1930 to 1942, radium and uranium salts were extracted for commercial purposes. From 1942 to 1957, uranium was recovered from ores, concentrates, and scrap materials under MED/AEC contracts. The site remained under the control of various AEC licenses until 1966. Since 1967, the property has been developed by the present owner. Various light industries currently occupy the twelve buildings in what is now an industrial park. Approximately 125 persons are employed at this site. None of these employees is performing any work which is related to the nuclear industry.

The site is divided into three separate parcels, designated A, B, and C as shown in Fig. 1. Extraction of radium began on the western side of parcel A. Residues were dumped on parcels A and B. As the plant expanded toward the east of parcel A, new buildings were constructed over the residues. Liquid slurry wastes were impounded in a swampy area located in parcel C; this area was later filled with site residues and covered with uncontaminated dirt.

Results of the radiological survey (Le78a) indicate that a layer of contaminated soil can be found within 1 m of the surface at almost any point on the site. Apparently, all buildings on the site are built over or directly adjacent to contaminated soils. A "typical" boring on parcel A would indicate contamination to a depth of almost 3 m; the average ^{226}Ra content of the core would be about 200 pCi/g. In parcel B, about the same results could be expected except for an area in the center which has up to 2 m of fill material over the contamination. A 150-cm layer of highly contaminated muck (up to 17,000 pCi/g of ^{226}Ra) would be encountered within 1 m of the surface of parcel C. The ratio of ^{226}Ra activity to ^{238}U and ^{227}Ac activities varied widely from sample to sample because of the wide variations in processes and in feed material used to generate the residues.

A summary of radiation exposures being received by the industrial park employees is given in Table 1. These elevated exposures can be attributed directly to the contaminated residues which cover and underlie practically the entire site. Variations in external gamma radiation

levels can be correlated fairly well with variations in subsurface contamination (Le78a). Radon and radon daughter concentrations in buildings can be attributed to ^{226}Ra contamination in surface and subsurface soils, in floor and former process drains, and on interior surfaces of the buildings. Daughters of ^{219}Rn, attributable to ^{227}Ac contamination in surface soils, were also detected inside structures (Le78a).

The ranges of average exposures inside structures shown in Table 1 represent the lowest and highest average obtained in any building on site. Each of these individual building averages represents numerous individual measurements. However, the range for airborne ^{230}Th represents the range observed in individual spot samples; these should not be construed as average values.

A summary of current exposure guidelines for an individual in the general public is given in Table 2. These guidelines, except for radon daughters in commercial structures, assume that the exposure is continuous. Comparisons between Table 2 and Table 1 show that employees at the Canonsburg site are exposed to average radon and radon daughter concentrations which are far in excess of the guidelines. In fact, only a portion of one of the twelve buildings had average radon and radon daughter concentrations below the guidelines. Remedial measures are obviously required to reduce on-site exposures to radon and its daughters. Results of spot samples and the presence of alpha contamination on practically all building surfaces indicate that occupants of some of the buildings on this site may be exposed to average concentrations of long-lived airborne radionuclides (particularly ^{230}Th) which exceed guideline values.

Exposures at the Canonsburg site are far higher than those associated with the other MED/AEC sites surveyed by ORNL. The Canonsburg site is the only MED/AEC site which was specifically included in the "Uranium Mill Tailings Radiation Control Act of 1978" (H.R. 13650, 95th Congress). Under the provisions of this act, the Secretary of Energy is authorized to conduct remedial measures at this site.

Pennsylvania Railroad Landfill Site, Burrell Township, Pennsylvania

The Pennsylvania Railroad Landfill Site is located approximately one mile southeast of Blairsville, Pennsylvania, in Burrell Township. This property, owned by the Properties Division of the Penn Central Transportation Company, lies between the Conemaugh River and the mainline tracks of ConRail (see Fig. 2) and consists of approximately 25 ha (60 acres).

During the period of October 1956 through January 1957, an estimated 10,500 metric tons of radioactive material were shipped by rail from the uranium processing plant in Canonsburg, Pennsylvania, and were dumped on the site. The material contained approximately 5000 metric tons (dry weight) of waste residues containing an average 0.097% U_3O_8 by weight. The uranium (approximately 5 metric tons of U_3O_8) was classified as "unrecoverable material-measured." The wet weight of the residues was

estimated to be 9000 metric tons, and since the material was shipped wet, it appears that approximately 1500 metric tons of possibly nonradioactive materials were mixed with the radioactive materials during loading. The waste residues were generated under an AEC contract at the Canonsburg plant.

The area where the residues were dumped was previously the river bed of the Conemaugh River, which had been diverted approximately 150 m to the south several years earlier. Apparently, this site was chosen, in part, because the dumping site and scattering technique used in unloading from the railroad cars would cause the material to be widely dispersed and intermixed with large volumes of nonradioactive wastes. Furthermore, it was thought that the material would be confined in a large chasm approximately 10 m deep. There were, and still are, no public thoroughfare passes through the site or in its immediate vicinity, other than the railroad. The nearest dwelling is approximately 150 m from the site. There are probably a few persons, for example, railroad workers or hunters, who may occasionally be on the site.

Results of the radiological survey (Le78b) indicated more than 75% of the residues lie at least 3 m beneath the surface. It was estimated on the basis of historic records that about 1.5 Ci ^{238}U was dumped on this site. Auger and core hole analyses performed during the recent survey could account for approximately 1.3 Ci (Le78b). Most of this activity was in an area of less than 2 ha (4 acres). Thus, it appears that most of the dumped residue has been accounted for by the recent survey.

Areas of surface contamination appear to coincide to a large extent with those of subsurface contamination shown in Fig. 2. Beta-gamma dose rates as high as 5.4 mrad/hr were measured at 1 cm above the surface; however, most values were below 0.1 mrad/hr. The average ^{226}Ra content of numerous surface soil samples taken from the shaded area of Fig. 2 was 10 pCi/g; ^{238}U average was 3.9 pCi/g. Background concentrations in the Burrell Township core soils are 1.9 and 0.9 pCi/g, respectively.

A summary of current exposure conditions at the Burrell Township site is given in Table 3. The average outdoor ^{222}Rn concentration on the site was 0.52 pCi/ℓ. However, instantaneous measurements as high as 9.7 pCi/ℓ were observed. Radon daughter concentrations measured on the site were reasonably typical of outdoor radon daughter measurements in that area of Pennsylvania. Average background gamma radiation in the Burrell Township area was found to be 8 μR/hr. Thus, the general average of external gamma radiation on site is slightly above background. Furthermore, the concentrations of radionuclides in all water samples were below the concentration guide for water in 10 CFR 20.

In summary, the Pennsylvania Railroad Landfill Site is contaminated by about 4 Ci of ^{226}Ra and 1.5 Ci of ^{238}U spread over an area of about 2 ha (4 acres). Although most of the contamination is presently a few meters below the surface, the random dumping of materials has resulted in some areas of significant surface contamination. Radiation

exposures on this site are slightly above regional background. Occupancy of the site is very infrequent. Scenarios can be offered, such as building structures over contaminated soils on the site, which could result in overexposures to site occupants.

This site is much more typical of the MED/AEC sites surveyed by ORNL than is the Canonsburg site. The Burrell Township site has a relatively small amount of measurable contamination. However, some type of remedial action, consistent with ALARA principles, could be taken to reduce potential radiation exposures to potential site occupants.

Middlesex Landfill Site, Middlesex, New Jersey

In 1948, about 6000 m^3 of dirt contaminated with pitchblende ores were brought to the 10-ha (23-acre) landfill site from the former Middlesex Sampling Plant. In 1960, elevated gamma radiation levels were detected on this site by civil defense monitors during a local civil defense exercise. A radiological survey of the site was made at that time by the AEC, and it was found that external gamma radiation levels over an area of approximately 2000 m^2 were 20 to 50 times background levels for the surrounding area. The AEC subsequently removed approximately 600 m^3 of the contaminated material nearest the surface and covered the area with about 1 m of uncontaminated dirt. This action reportedly lowered the external gamma radiation levels to no more than 50 μR/hr.

In 1974, a second survey of the site was performed to reevaluate the radiological conditions. During the time between the 1960 and 1974 surveys, an approximately 2-ha parcel of the landfill site (originally owned by the Borough of Middlesex) had been sold to the Middlesex Presbyterian Church, and a church had been constructed on the parcel. During weekdays, part of the church building and grounds is used as a day care center for local children. The church and the Middlesex Municipal Building are located on the western edge of the site. It appears from discussions with local people that both the church and the Middlesex Municipal Building were constructed on "non-fill" or solid ground. The landfill site is surrounded by residences which approach to within 0.5 km of the south and west and to Bound Brook on the eastern and northern edges. Results of the 1974 AEC survey indicate that the remaining contamination on the property was in an area (see Fig. 3) bounded by the baseline and by the lines designated as 300R, 2+0, and 6+0 (Cr78).

Radiological survey results showed that average surface contamination of ^{226}Ra and ^{238}U in soil were indistinguishable from background activity of about 1 pCi/g of each. A few subsurface samples contained detectable activity caused by small pieces of material, presumed to be pitchblende ore. These contaminated samples were found in the general area referred to in the previous paragraph, generally at depths of less than 4 m.

A summary of present radiation exposures is shown in Table 4. Background gamma radiation measurements in this area ranged from 5 to 10 μR/hr, with an average of 8 μR/hr. Hence, the average external gamma

radiation at this site is within the range of area background. The maximum external gamma radiation was associated with surface contamination in an area of about 50 m^2. Construction of a building on this contaminated area, particularly a building with a basement, could lead to elevated human exposures. Furthermore, the underground contamination poses the potential for producing elevated exposures if future activity at the site were to uncover pieces of uranium ore at or near the surface.

This site represents the radiological condition at those MED/AEC sites where present radiation exposures cannot be distinguished from background over almost the entire site. Minor remedial measures at these sites are expected to involve minimal expenditures.

Summary and conclusions

The three sites summarized in this paper represent the range of results found in ORNL surveys of waste storage areas at former MED/AEC sites. Most of the sites would be typified by the Burrell Township site—current radiation exposures averaged over the site are slightly, but demonstrably, greater than background; small portions of the site contain highly contaminated material in close proximity to the surface of the ground. Exposures to ^{222}Rn and its daughters in structures built on these contaminated areas would probably exceed guidelines. Exposures to external gamma radiation will probably exceed guidelines at several points within the contaminated areas. However, the Burrell Township site does not demonstrate appreciable radionuclide migration due to surface runoff. This is an appreciable problem at three other MED/AEC sites. Remedial actions are required at sites such as Burrell Township.

The Middlesex Landfill Site represents the lower end of the spectrum of present exposures found at MED/AEC sites. Exposures can be differentiated from background only at small portions of these sites. Minor remedial action would be required at these sites.

The Canonsburg site represents the upper end of the exposure spectrum found at MED/AEC sites surveyed by ORNL. Practically all of the present exposures at Canonsburg can be traced to previous waste management practices at that site. Extensive remedial measures are required to reduce radon and daughter concentrations in buildings as soon as possible at Canonsburg.

Waste management practices employed during the time that MED/AEC contracted activities were performed have a major bearing on the extent of current radiation exposures at these sites. The magnitude of these exposures is directly related to the amount of contamination still present at a site. Hence, sites with the best planned waste management practices generally have the lowest present day exposures.

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Table 1. Summary of current on-site exposures at the Canonsburg, Pennsylvania, site

Exposure source	Range of average values observed	Maximum value observed
Radon in air inside structures	2.6 to 107 pCi/ℓ average daytime concentration	227 pCi/ℓ
Radon daughters in air inside structures	0.01 to 0.43 WL* average daytime concentrations	0.5 WL†
External gamma radiation inside structures	20 to 80 μR/hr averaged over buildings	310 μR/hr
Airborne ^{230}Th inside structures	0.003 to 0.2 pCi/m^3 range of spot samples	0.2 pCi/m^3
Radon in air outside structures	2.5 to 17 pCi/ℓ average 24-hr concentration	69 pCi/ℓ
External gamma radiation outside structures	110 to 210 μR/hr averaged over parcels A, B, and C	1600 μR/hr

*The WL (working level) is defined as any combination of short-lived radon progeny per liter of air which will result in the ultimate emission of 1.3×10^5 MeV of alpha energy by decay to ^{210}Pb.

†Measured during good ventilation conditions. Under poor ventilation conditions (cold weather) maximum is estimated to be 1.9 WL.

Table 2. Summary of current guidelines for exposure to a member of the general public

Exposure source	Guideline value	Documentation
Radon in air	3 pCi/ℓ	10 CFR 20
Radon daughters in air	0.03 WL	10 CFR 20
commercial structure	0.03 WL	10 CFR 712
residential structure	0.01 WL	10 CFR 712
Airborne ^{230}Th (insoluble)	0.08 pCi/m^3	10 CFR 20
External gamma radiation (whole body)	500 mrem/yr	10 CFR 20

Table 3. Summary of current on-site exposures at the Burrell Township, Pennsylvania, site*

Exposure source	Average values observed	Maximum values observed
Radon in air	0.52 pCi/ℓ	9.7 pCi/ℓ
Radon daughters in air	0.0009 WL	0.001 WL
External gamma radiation (at 1 m)	11 μR/hr	630 μR/hr
Beta-gamma radiation (at 1 cm)	<0.1 mrad/hr	5.4 mrad/hr

*Includes component of exposure due to background.

Table 4. Summary of current on-site exposures at the Middlesex, New Jersey, landfill site

Exposure source	Average values observed*	Maximum observed value
Radon in air	0.04 pCi/ℓ	Calculated to be 0.01 pCi/ℓ above background
External gamma radiation	5 μR/hr	32 μR/hr

*Includes component of exposure due to background.

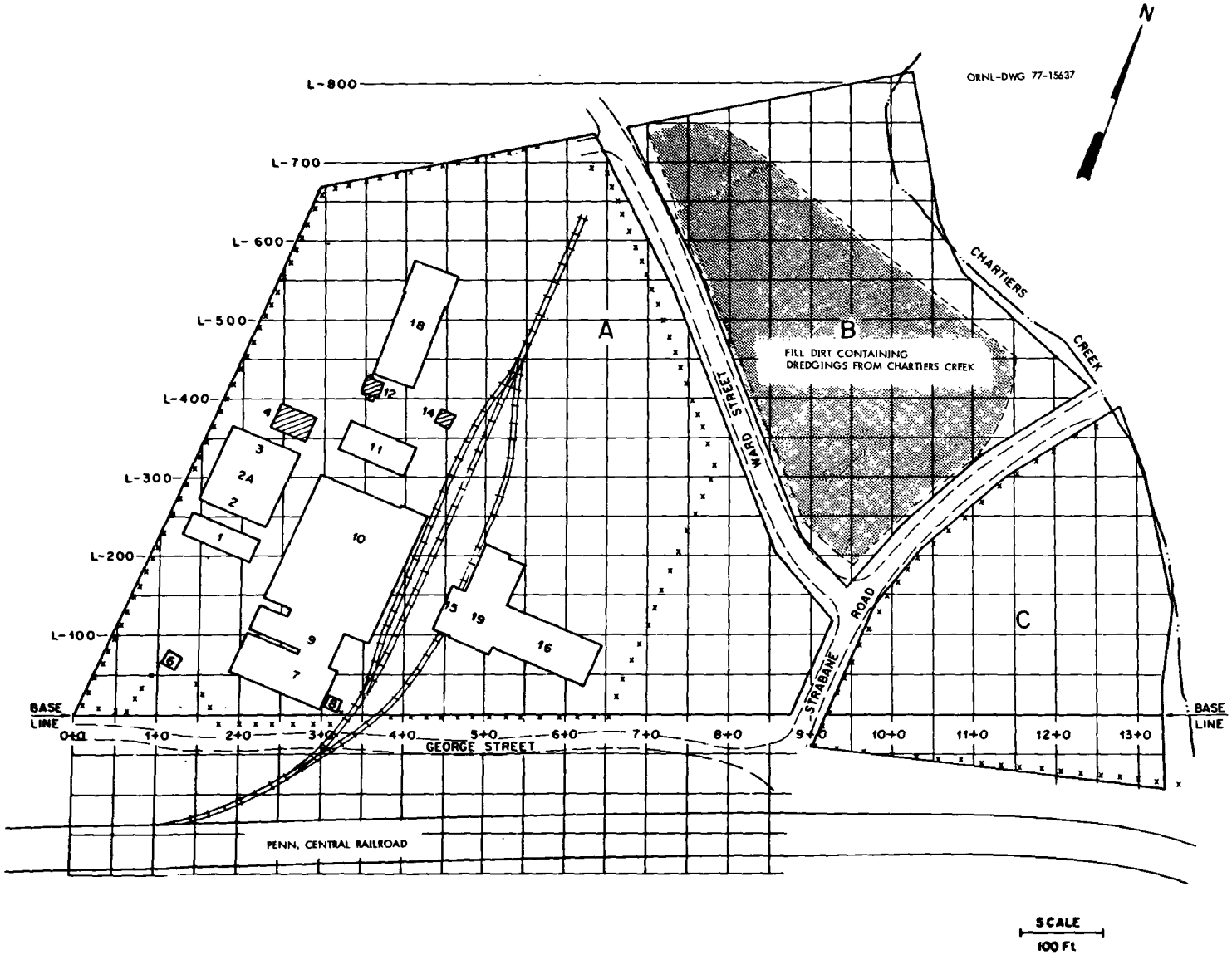

Fig. 1. Layout of the present setting of the site at Canonsburg, Pennsylvania.

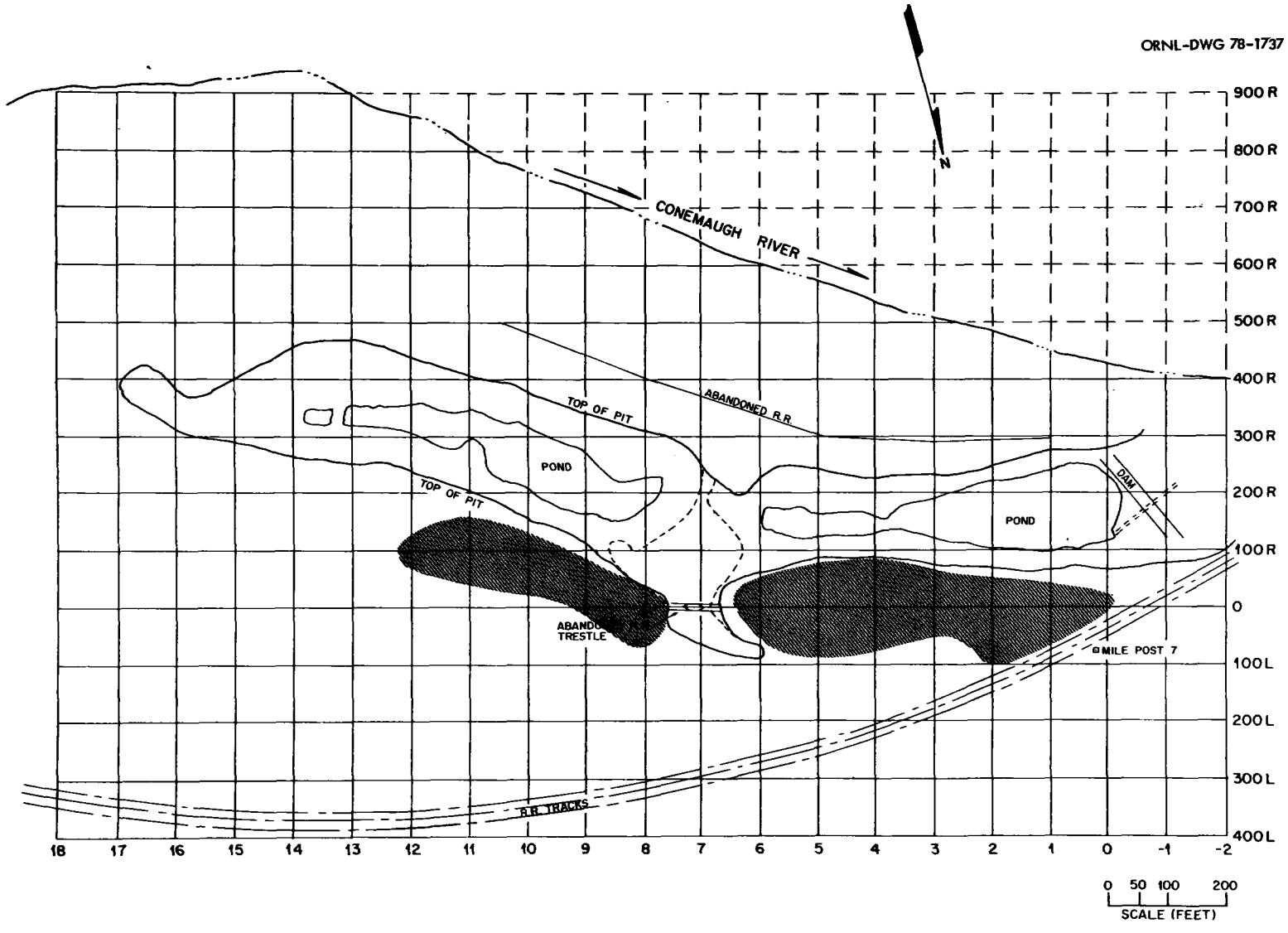

Fig. 2. Layout of landfill site at Burrell Township, Pennsylvania. Shaded areas are those where subsurface contamination has been found.

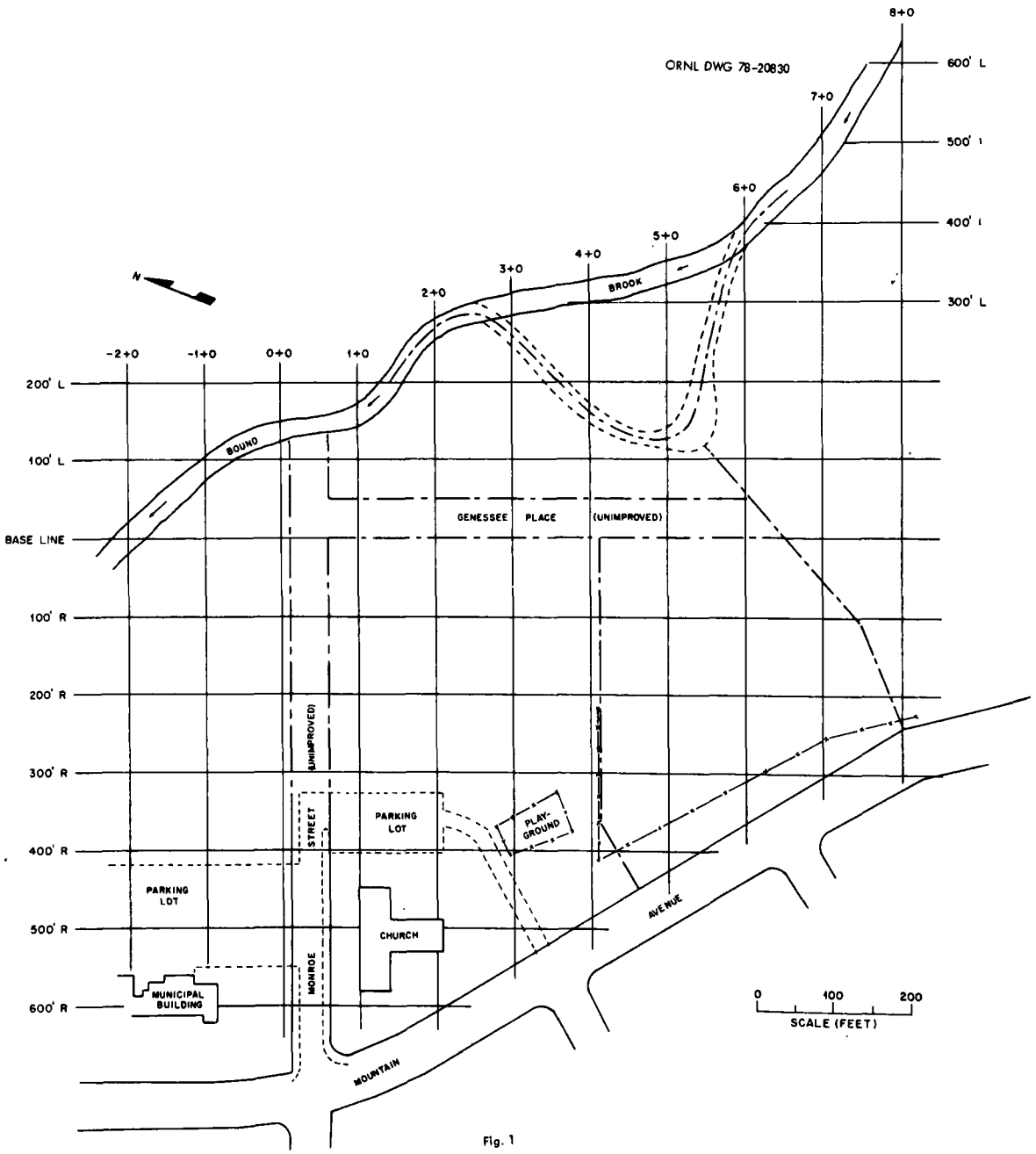

Fig. 3. Present setting of the Middlesex, New Jersey, landfill site.

Radiological Impact of Uranium Tailings and Alternatives for Their Management

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ABSTRACT

The radiological hazards associated with uranium tailings arises from inhalation of airborne particulates and radon daughters, ingestion of food grown in contaminated ground, and from external exposures to pollutants in the vicinity. Uncontrolled tailings piles are mobile sources of fugitive dust that may produce a practically uncleanable adjacent environment. A practical procedure for managing solid tailings is addition of surface moisture, mechanical and gravitational separation of slimes, and storage of slimes below solution tailings. Presently practical alternatives for tailings management are variations of two basic methods--surface and below-ground disposal. Protocol for tailing management should be based on both reduction of exposure pathways and containment throughout the predictable future. Isolation of tailings by natural materials such as clay lenses and combinations of overburden, top soil, vegetation and rip-rap may provide both minimization of exposure and stability. Experimental measurement of radon flux over two inactive tailings, acid and carbonate leached tailings resulted in average specific flux values of $\phi_\infty \simeq 0.64$ *and* $\phi_\infty \simeq 0.30$ *(pCi Rn-222/m^2·sec) / (pCi Ra-226/g), respectively. The average diffusion coefficient for these tailings were, respectively,* 2.4×10^{-3} *and* 5.7×10^{-4} *cm^2/sec. Tailings covered with native soil of clay-silt-sand mixture to a depth of 225 cm resulted in attenuation of flux with diffusion coefficients of* 3.69×10^{-3} *and* 3.60×10^{-3} *cm^2/sec for ACID and ALKO sites, respectively. By means of the UDAD code dose commitments were estimated for inhalation of particulates and radon and for external exposure under three degrees of surface moisture on the tailings. Based on these analysis and assumption that the dose contribution from ingestion pathway is comparable in magnitude to that of inhalation, both compliance with the 25 mrem/year limit and reduction of flux to background level is feasible. Stability of alternative decommissioned tailings over the predictable future is discussed.*

INTRODUCTION

Uranium mills process large quantities of ore containing uranium and its daughters (Th-230, Ra-226, Rn-222, Pb-210, Po-210) in concentrations exceeding several hundred times the average natural concentration. Thus the mills are sources of contamination of large areas of land (Do75) and surface waters traversing watersheds contaminated with tailings. The main hazards of uranium tailings are associated with release of radon-222 and fugitive dust from dried

tailings pond beaches and eroded tailings piles. These releases result in inhalation of particulates and radon progeny, ingestion of food produced from contaminated ground and water, and external exposure to beta-gamma radiation. Generally, liquid-borne mill wastes (tailings) are impounded in tailings ponds. Acid tailings are about 50% solids by weight. In many older mills the peripheral dikes are composed of the solid tailings themselves. These mills generally separate coarse sands in covering the exterior surface of the dam and discharge the finer particles (slimes) into the interior surface of the dam. Because of their mobility, the dusts and sands from these tailings contaminate the land and surface waters collected from the watersheds. In the following sections a selected alternative for tailings management to mitigate the potential releases are analyzed and a selection of the experimental data are reported.

FIELD INVESTIGATION

Experimental Procedures

The field investigations were made in support of the Uranium Milling Operation Generic Environmental Impact Statement for United States Nuclear Regulatory Commission. A major part of the field study has been conducted since June, 1977, at the Anaconda Uranium Mill, Bluewater, New Mexico, in cooperation with William E. Gray, Director of Environmental Affairs, Uranium Mining and Refining. Between 1955 and 1978 this mill extracted uranium using an acid leaching process with a throughput of 3500 tonnes per day of about 0.25% uranium ore. At present, the mill processes 5400 tonnes of ore per day. The tailings are pumped to a retention area of about 8×10^5 m^2. Before reconstruction of the present dikes, tailings overflow were collected in catch basins. These deposits are about 125 cm in depth. Between 1953 and 1956 Anaconda also operated a carbonate leach process. The tailings from the carbonate process were separated from those of the acid leach process. Figure 1 shows the location of these tailings areas; they are designated ACID (inactive acid tailings), ALKO (inactive carbonate tailings) and MAIN (presently active tailings). These inactive tailings were covered by an average of about 85 cm of clay during the fall of 1977.

Radon concentration in the air was continuously measured at three stations (#102, #103 and #104) on the Anaconda mill site, (Figure 1) and at two stations (Elks and Bride) located about 25 km east and west of the mill. Design and calibration of the continuous radon and working level (CRWM) monitors were previously reported (Mo78, Mo79c). Radon concentration was also measured based on integrating air sampling techniques (Si69).

Radon flux, i.e. the amount of radon-222 that is transported across a unit area of the surface per unit time, was measured using the accumulation method, charcoal cannister, and ANL continuous radon flux monitor (Mo79c). In this report, flux measured only by the accumulation method is reported. Accumulation method for measurement of flux has been used by Kraner et al. (1964), Wilkening (1972), Bernhardt (1975) and Clements (1978).

The accumulation of radon, Q, (pCi) in a collecting device placed directly over a surface area, A, is related to the radon flux, Φ, for the collection period, Δt:

$$Q = A \cdot \Phi \cdot \Delta t \quad (1)$$

assuming (a) that Δt << radon-222 half-life of 3.92 days, (b) that back diffusion of radon is low, and (c) that the accumulator does not disturb the exhalation process. In general, criteria (a) and (b) can be met for a Δt of less than one hour, but satisfaction of the criteria (c) is not yet proven. Radon flux is dependent on wind velocity over the surface and the accumulator will limit the surface wind over the area.

The accumulated radon, Q, was measured by determining the accumulator radon concentration, $\chi_g = Q/V$, where V is the accumulator volume of approximately 100 l. Unless Φ is very large, i.e., $\chi_g >> \chi_a$, the concentration of radon in air χ_a trapped within the accumulator must be measured. In this study two techniques for measuring Q were utilized. In the first technique, the radon concentration in air, shortly before placing of the device on the surface, and the radon concentration in the accumulator were measured. A collection time, Δt, of 10 and 20 minutes over tailings and soil, respectively, were used. In the second technique, air in the accumulator was sampled at 10, 20 and 30 minutes after placing the accumulator on the gound. The initial concentration of radon in air, χ_a, was estimated from extrapolation of the radon concentration in the accumulator to zero collection time. The flux was calculated from:

$$\Phi = \frac{(\chi_g - \chi_a) \cdot h}{\Delta t} \quad (2)$$

where, h and A are the height of the accumulator (65 cm) and the area of the collector surface (0.16 m^2), respectively (h = V/A). Statistical comparison of flux values showed that the two methods gave similar results.

Samples of radon in the ambient air and from inside the collector were obtained by means of an in-line filter and desicant using evaculated 0.4- and 1.4- liter ZnS scintillation cells (Eberline), respectively. After an elapsed time of about 4 hours, the radon concentrations were measured using an Eberline SAC-R5 alpha scintillation counter. Each cell was cross calibrated using 100-cm^3 Lucas cells (Johnston Laboratories) and an NBS radium solution (Lucas 1957, 1977).

Flux from the uncovered surface of the two inactive tailings (ALKO and ACID, Fig. 1) was measured between August, 1977, and October, 1978. On each pile region an area of about 1000 m^2 was selected for study. Within this area each tailings region was cored at several locations to a depth of approximately 1.5 m, and samples were collected at approximately 15 cm intervals. Selected samples were analyzed for Ra-226 concentration and particle size distribution.

Within each of the tailings region, test areas were randomly selected. Radon flux at the selected test areas was measured over several weeks. One area was selected for measurement of flux attenuation through native soil cover (ACID5 and ALKO5). Native soil near Anaconda is mostly weathered fine grained silty sandstone and mudstone with patchy presence of evaporities and limestone. The regional soil is composed of montmorillonite clay, kaolinite, quartz and unidentified amorphous material (Fu77). The test plots were

covered initially with soil to a depth of about 7.5 cm and compacted. Radon flux was measured after a waiting period of several days. Presently, each test tailings plot had been covered to a depth of about 225 cm.

Experimental Results

Average radon-222 background was 0.3 pCi/1, ranging from 0.2 to 2.5 pCi/1. Radon concentrations were measured using Continuous Radon Monitor (CRM) and integrated air sampling method (SI69). Net radon concentration (i.e. radon attributable to the mill operation) measured at stations #102, #103 and #104 were, respectively, 0.4, 1.2 and 0.4 pCi/1. The average of radon concentrations measured at six locations on the MAIN tailings was 8.6 pCi/1 (6 to 15 pCi/1 in range).

Radon flux is influenced by atmospheric pressure (C174), wind speed (Kr64), stability, and climatic factors such as soil moisture and temperature. Our flux measurements using Continuous Radon Flux Monitor (CRFM) will be reported elsewhere (Mo79d). In order to minimize diurnal effects, radon concentration in air on ACID and ALKO was measured between 10 a.m. and 1 p.m.

Normalized flux, i.e. the ratio (covered tailings/control tailings) was assumed to be independent of meteorological variables. Therefore, radon flux at both covered tailings (ACID5 and ALKO5) and control tailing sites (ACID2 and ALKO2) were measured at the same time, The average of 49 measurements of radon concentration in air during the measurements of radon flux on each area was 2.7 pCi/1 (0.4 to 12.2 pCi/1 in range) on ACID and 4.4 pCi/1 (0.1 to 12.7 pCi/1 in range) on ALKO.

Radon flux, Φ, measured at stations #100, #101 and #105 (Fig. 1) was between 0.4 and 1.5 (pCi/m^2•sec). Ra-226 concentration of soil was measured using high-resolution gamma spectroscopy (Ge detector). Radium concentration within the 10 cm of soil surface was in the range of 0.7 to 2.5 pCi Ra-226/g. Additional measurements of radon flux and radium concentration in the soil distant from the mill site are in progress.

The average radium concentration in cores through the ACID tailing was 616.6 pCi Ra-226/g with a range of 220 to 1800 pCi Ra-226/g. The average radium concentration in ALKO was 601.4 pCi Ra-226/g with a range of 70 to 1000 pCi Ra-226/g. Radium concentration did not indicate a distribution pattern with depth. Less than 50% of tailings by weight were 200 μm and 125 μm in diameter, respectively for ACID and ALKO. In comparison these sizes are smaller than those from the MAIN tailings ($\simeq$ 400 μm). Average core moisture in both test tailings was about 10%. The moisture in the MAIN tailings beach is from a dry 0.2% to a complete saturation.

The average of 49 radon flux measurements (Table 1) made over each control tailings (ACID2, ALKO2) between August, 1977 and October, 1978 was 4 15.6 and 174.2 (pCi Rn-222/m^2•sec). The average flux for ACID and ALKO was 376.8 and 190.0 (pCi Rn-222/m^2•sec), respectively. The range of flux was between 59.7 to 1103 (pCi Rn-222/m^2•sec) for ACID and 45.1 to 762 (pCi Rn-222/m^2•sec) for ALKO (Table 1).

Figures 2 and 3 show the measured radon flux through the soil cover. The particle size of the soil covering was 50% (by weight) less than 300 μm in diameter and with a moisture content of less than 5% during deposition. Soil moisture was higher at the point of contact with the tailings but in general it remained at about 5% near the surface.

Estimation of Diffusion Coefficients

Radon flux from the surface of a single, homogeneous layer of material is a function of radium concentration, R, emanating power of the tailings, ε, (fraction of the radon generated which is released into the diffusion space), and bulk diffusion coefficient, D_t, given by:

$$\phi_t(z) = \left(\varepsilon\rho R \sqrt{\frac{\lambda D_t}{P_t}}\right) \left[\tanh\left(z \sqrt{\frac{\lambda P_t}{D_t}}\right)\right] \times 10^4 \qquad (3)$$

where ρ is the tailings density, λ is the radioactive decay constant of radon, p_t is the porosity of the tailings, and z is the depth of the tailings deposit. Table 1 gives the average measured flux and diffusion coefficient D_t calculated by using the following parameters: ε = 0.25, P_t = 0.3, λ = 2.1 × 10^{-6} sec^{-1}, z = 125 cm, ρ = 1.6 g/cm^3 and R = 617 and 601 pCi Ra-226/g for ACID and ALKO, respectively. Emanation rates from domestic uranium ores vary only slightly with ore moisture between 10% and 80% of saturation. Emanating power for domestic uranium ores is between 0.01 - 0.9 with an average of about 0.25 (Au75, Me74). For uranium tailings ε = 0.23 (Cu73). In these measurements average specific flux, ϕ (pCi Rn-222/m^2·sec) / (pCi Ra-226/g) is 0.61 and 0.67 for ACID and 0.32 and 0.29 for ALKO. The lower specific flux and diffusion coefficient at the ALKO pile may be partially due to the smaller tailings size relative to the ACID pile. Based on these parameters, a 100-cm tailing depth is effectively an infinite depth (i.e. more than 95% of flux from infinite depth) for radon diffusion and the ϕ values given in Table 1 are effectively ϕ_∞. Specific flux ϕ measured (Si69) for loose sediments rich in clay is ϕ = 0.37, for sandy soils ϕ = 0.18, and for an average of clays and heavy loams ϕ = 0.28 (pCi Rn-222/m^2·sec) / (pCi Ra-226/g). The specific flux for abandoned Vitro tailings has been reported to be 1.6 (pCi Rn-222/m^2·sec) / (pCi Ra-226/g), (Sc74). In our radiological analysis of the Bear Creek Project (NRC77) an average specific flux of ϕ_∞ = 1 (pCi Rn-222/m^2·sec) / (pCi Ra-226/g) for tailings containing both dry and wet beaches was utilized.

The radon flux from tailings which escapes from the surface of soil cover, Φ_c, is dependent on the same parameters given for the Equation (3). The emanation factor ε measured for various soil conditions is reported to be 0.14 to 0.29 (Si69). In this study a value of ε = 0.25 for soil cover was used. Radon flux through a soil cover is given by:

$$\Phi_c(\xi) = \Phi(\xi = 0)\, f(\xi) \qquad (4)$$

where ξ is the depth of the soil cover over tailings which release flux Φ ($\xi = 0$) = Φ_t. The attenuation function $f(\xi)$ is:

$$f(\xi) = \left[2\, e^{-k\xi}\right] \left[\left(1+e^{-2k\xi}\right) + \left(\frac{D_t \cdot h}{D_c \cdot k}\right)\left(1-e^{-2k\xi}\right) T\right]^{-1} \quad (5)$$

where $k = \sqrt{\frac{\lambda P_c}{D_c}}$, $h = \sqrt{\frac{\lambda P_t}{D_t}}$, and $T = \text{Tanh}\ (h \cdot z)$

The above equation may be further simplified by substituting $\delta = \left(\frac{D_t}{D_c}\right)\left(\frac{h}{k}\right)$

into the Equation 5 which then reduces to:

$$f(\xi) = 2e^{-k\xi} \left[(1 + \delta kT) + (1 - \delta kT)\, e^{-2k\xi}\right]^{-1} \quad (6)$$

and assuming $P_t = P_c$. Equation (6) was fitted to the measured flux using the Marquardt (Ma63) method. Figures 2 and 3 as a function of soil cover ξ give the average measured flux and predicted flux using the Equation 6. The radon diffusion coefficients through the soil cover D_c is 3.69×10^{-3} and 3.60×10^{-3} (cm^2/sec) for the data shown in Figures 2 and 3. The number of radon measurements through the soil cover was 47 for each of the tailings; they were spread over 387 days.

Laboratory measurements of radon diffusion coefficients for soil covers obtained from the Powder River Basin, Shirley Basin, and Ambrosia Lake, under a porosity of 0.43 to 0.60; a moisture content of 1% to 11%; and a compaction of 65% to 89% gave values between 3.9×10^{-3} cm^2/sec and 3.2×10^{-2} cm^2/sec (Ro79). The effect of cover moisture is a reduction of the diffusion coefficient. Diffusion coefficients for soil obtained from Powder River Basin, similar to the soils from the Bear Creek Project, are 1.8×10^{-2}, 1.6×10^{-4} and 2.1×10^{-5} cm^2/sec, respectively, at 5%, 17% and 30% moisture (Ro79).

Since the soil cover contains Ra-226, the contribution to radon flux from thick soil covers should be considered. A composite soil sample had an average of 1.5 pCi Ra-226/g. Figures 2 and 3 show flux after application of this correction using a finite difference multilayer simulation code. Range of radon flux for background is also shown in Figures 2 and 3. The depth of cover needed to achieve a reduction of radon flux to twice the average background (0.75 pCi Rn-222/m^2·sec) is about 450 cm based on the flux corrected for the cover Ra-226 activity. But the depth of cover is only 400 cm using only the predicted flux, i.e. that not corrected for the Rn-226 in the soil.

TAILING MANAGEMENT

A major objective of tailings management during both milling operation and after decommissioning is the mitigation of radiation exposure by short-term impoundment and long-term containment. This objective may be accomplished by: (a) reduction or elimination of airborne radioactive releases, (b) reduction or elimination of contamination of surface and groundwater,

(c) insuring long-term stability and isolation of the tailings (Ma78, Sc77). In addition, in order to minimize the potential hazards associated with mill wastes, a mill site which is as remote as possible from both human population and local streams should be selected.

Practical alternatives for the management of tailings (NRC77) under conditions which satisfy the "as low as reasonably achievable" criteria, are variations of two methods, surface and below grade disposal. In each case disposal areas are designed to minimize both vertical and lateral seepage of tailings solution. An example of surface storage of tailings is the Bear Creek Project (NRC77) which was built using technologies available at the time. This mill has utilized the natural valley contour to impound the tailings. Seepage through the tailings dam foundation was controlled by excavating a cut-off trench to the top of the bedrock beneath the center of the dam and backfilling with impervious material. The dam itself contains a central impervious core. The surface of the tailings retention area (about 6×10^5 m^2) was covered with compacted clay to a minimum depth of 1 meter. The retention area thus created is capable of storing about 6×10^6 tonnes of solids.

Tailings can be disposed below grade in areas such as open pit mines or specially prepared pits. This type of disposal, when available and conditions permit, allows a deep cover to be applied without subjecting its surface to excessive erosive forces. An example of such disposal is the Morton Ranch facility (NRC78).

Active Tailings: Particulates

The tailings beach area (the surface which is not covered by tailings solution) may comprise from 10% to 90% of the retention area depending on the annual evaporation rate, rate of seepage and the recycling of the tailings solution. The quantity of tailings material released from the beach as fugitive dust is dependent on wind velocity, surface, moisture and the physical protection of the surface. Upon aging fine tailings tend to conglomerate on larger grains (Ca65). In this condition and otherwise undisturbed, they are relatively resistant to erosion by wind.

Based on the physical characteristics given in Table 2 and meteorological parameters, typical of the southwestern United States, listed in Table 3 the rate at which tailings particulates are transported by wind was estimated using the UDAD code (Mo79). The specific activity for particles less than or equal to 35 μm in diameter was assumed to be 62.5 pCi/g for U-238 and U-234, and 1250 pCi/g for Th-230, Ra-226, Pb-210 and Po-210. This is equivalent to assuming an average specific activity for the uranium ore of 500 pCi/g and a specific activity for the small tailings particles that is 2.5 times greater than the average. Figure 4 gives the total Ra-226 concentration of airborne radioactivity as a function of distance from the geometric center of the tailing beach (0.3 km^2 in area) in the direction of maximum transport (northeast). These concentrations include resuspension of Ra-226 from the ground after accumulation from 20 years of mill operation. Because the concentrations of Th-230, Pb-210, and Po-210 in the tailings are

about the same as for Ra-226, the concentrations given in Figure 4 are equally applicable to these radionuclides, except for Pb-210. As shown in Figure 5, the concentration of lead in the air begins to increase beyond about 1 km from the pile because of the contribution from decay of radon daughter products in the air. For example, at 10 km in the base case the total Pb-210 is 3. $\times 10^{-4}$ pCi/m^3 in comparison with about 1.5 $\times 10^{-6}$ pCi/m^3 released directly from the tailings surface. Since the specific activity of uranium in the tailings was assumed to be about 5% of that for Ra-226, the resulting airborne concentration of uranium is 5% of the values given in Figure 4.

The amount of dust released and, therefore, the concentration of airborne particulates is dependent on the moisture content of the tailings surface. As Figure 4 indicates, an increase in surface moisture (not necessarily the moisture content of the bulk tailing) from a dry, 0.2% moisture in the base case (a typical condition for a dry beach in a southwestern climate) to a moist (10%) and wet (30%) condition results in a marked decrease in airborne concentration. At 1 km from geometric center of the tailings beach, the predicted average concentration of Ra-226 in air is about 9 $\times 10^{-4}$, 3 $\times 10^{-6}$ and 1.3 $\times 10^{-6}$ pCi Ra-226/m^3, respectively, for the base case, moist and wet tailings surface. Concentration of radium in air for 1% and 5% moisture is 2.04 $\times 10^{-5}$ and 5.35 $\times 10^{-6}$ (pCi Ra-226/m^3), respectively. This results in a reduction by factors of about 4, 17, 30 and 70 times in airborne Ra-226 concentration with an increase in moisture from 0.2% to 1%, 5%, 10% and 30%, respectively. The 30% moisture is about the saturation condition of the tailings. The concentration predicted for 10% and 30% moistures are conservative and for 30% moisture the rate of tailings transported may be practically zero. The predicted concentrations for moistures exceeding 10% are only an order of magnitude estimates.

The accumulation of ground contamination as a result of the deposition of airborne radioactivity during the 20 years of mill operation also was estimated using the UDAD code and the data from Tables 2 and 3. The results of the calculation of Ra-226 surface soil concentration as a function of distance from the pile are shown in Figure 6 by the lower of the two curves for each case. The dependence of the total fallout on the surface moisture content of the beaches is clear from this figure. This dependence is not surprising since the deposition rate is directly proportional to the concentration of the radionuclide in the air, and the influence of surface moisture on the air concentration was shown in Figures 4 and 5. Even for the base case with dry beaches, the buildup of radium contamination on the ground, as predicted by the UDAD code and presented in Figure 6, is not large compared to background levels if the fallout is assumed to mix through the upper layer of soil. The average concentration of Ra-226 in soil is about 1.5 pCi/g, so a soil layer to plow depth of 25 cm contains 6 $\times 10^5$ pCi/m^2. Figure 6 shows that at 1 km from the center of the pile, the Ra-226 concentration in the plow layer would be increased about 0.3% by deposition of tailings particulates.

In addition to the vertical flux of dust particles which accounts for the ground concentrations calculated by the UDAD code, a horizontal flux of relatively large particles, similar to desert sand, can contribute significantly to soil contamination under conditions of high wind. With winds in

excess of 19 knots tailings particles as large as 1000 μm in diameter are transported. A total horizontal flux exceeding 5 kg/m.hr has been measured (mo79). Fortunately, winds of sufficient velocity to generate such a large horizontal flux are infrequent, but during a severe wind storm the Ra-226 concentration near a tailings pile could be increased by more than an order of magnitude in a short period. In Figure 6 the cross-hatched area between the two curves in each case is a qualitative estimate of surface contamination from the horizontal flux of particles.

Dose commitments to bone and lung from inhalation of Th-230, Ra-226, Pb-210 at a selected distance and in the direction of maximum transport of tailings from the dry beaches (the base case) were computed using UDAD code and are listed in Table 4. Dose commitments for the moist and wet surface conditions are about the same ratio as the ^{226}Ra concentrations in air for base case/moist/wet, viz. 1/30/70. External gamma exposures from the ground contamination due to fallout (not including horizontal flux) of the airborne tailings and from airborne radionuclides are given in Table 5. The dose commitment to any organ from inhalation plus external exposures from tailings particulates under base case conditions is less than the 25 mrem/year standard published by EPA (40CFR190), assuming a controlled boundary to exclude the general population at 1 km from the tailings center. In these analyses, the contribution of ingestion exposure was not included. The ingestion pathway may contribute equal to the combined dose commitment from inhalation and external gamma exposure.

Active Tailings: Radon

Radon flux, Φ, from the surface of the tailings was assumed to be 500 pCi $^{222}Rn/m^2.sec$ (dry tailings), 50 pCi Rn-222/m^2.sec (moist) and 10 pCi Rn-222/m^2.sec (wet). These correspond to specific fluxs of Φ = 1, 0.1 and 0.02 (pCi Rn-222/m^2.sec) / (pCi Ra-226/g). The diffusion coefficient of radon is exponentially dependent on the moisture content. An increase in moisture from a base case dry condition to saturation results in a decrease of about 10^{-4} in the diffusion coefficient and a decrease by about 10^{-2} in specific flux (Ro79).

Figure 7 gives radon concentrations in the direction of maximum tailings transport for the three surface moisture conditions. Concentrations of radon-222 at 0.1 km (Fig. 7), downwind from the geometric center of the tailings beaches are 10.3 pCi Rn-222/1 and at 1 km only 0.92 pCi Rn-222/1. At 10-km distance radon concentration decreases to 0.03 pCi/1. Corresponding working levels are estimated to be 7.4×10^{-3} at the center of the tailings, decreasing to 2.7×10^{-3} at 1 km and to 2.5×10^{-4} at 10 km. Measurements at several inactive tailings piles have yielded average airborne radon concentrations, three feet above the surface and directly over the tailings, ranging from 3.5 to 19 pCi/1 (3.5×10^3 - 1.9×10^4 pCi/m^3) (US-DH69). Our average of measurements at the six locations on the MAIN tailings was 8.6 pCi/1 (6 to 15 pCi/1 in range). Radon concentration predicted for the base case is 10.3 pCi/1 on the tailings (Fig. 7). A detailed comparison of theoretical predictions and experimental data has been previously reported (Mo78).

Dose commitments from inhalation of Rn-222 and its progeny are given in Table 6, for the base case, moist and wet tailings surfaces, assuming an average dose conversion factor of 0.625 (mrem/year) / (pCi Rn-222/m^3) for combined exposure to inside and outside radon concentrations. The dose from radon progeny exceeds by far the dose from the long-lived particulates. A comparison of the dose commitments given in Table 6 indicates that even under wet conditions and at a distance of 1 km, a dose commitment of 11.5 mrem/year to the bronchial epithelium is expected. But, at present, the only radiation limit for exposure of the general public is a maximum permissible radon concentration (MPC) of 3 pCi/l. Figure 8 gives the isopleths for the safety factor (SF) defined as follows:

$$SF = \log \left[\frac{\text{concentration of Rn-222 in air}}{\text{maximum permissible concentration}} \right].$$

At 2 km northeast, Figure 8 shows that the safety factor is about SF = -3, corresponding to a radon concentration of 1/1000 of the maximum permissible concentration. Under the base case condition a boundary 0.6 km from the tailings would satisfy the criteria that SF = 0, while with moist beaches the radon concentration right on the surface of tailings is less than the 3 pCi/l MPC.

Mill Tailings Decommissioning

The protocol for decommissioning uranium mill tailings should be based both on minimization of exposure from all pathways and on insuring the containment of the tailings throughout a predictable future. Tailings decommissioning protocol should consider the transition period from active tailings storage to completion of drying prior to isolation of the tailings. Because the rate of release of radioactivity from dry tailings is relatively high, the environmental impact produced during the interim drying-out period, if it is extended for a long time, could exceed the impacts of the operating phase when the tailings were kept moist. "Progressive decommissioning" of tailings pile by covering with overburden material more or less continuously in parallel with drying operations is both technically practical and financially feasible.

Viable decommissioning protocols under present technological and competitive conditions are isolation of tailings within a soil lens of clay material and burying beneath overburden from the mining operation and a combination of vegetation and rip-rap if required to reduce erosion. Examples of decommissioning plans are given in Chapter 10 of the Bear Creek Project environmental statement (NRC77). The suggested decommissioning protocol requires that the tailings solids be placed in a clay lined impoundment and covered with 25 cm of compacted montmorillonite clay thereby enclosing the tailing in an artificial clay lens. Further, the clay cap is to be covered with an additional 180 cm of overburden and top soil. Placement of the clay cap, compacted to engineering specifications of the Bear Creek model, requires initial covering of the tailings with overburden to support machinery. Thus, the total cover may exceed 300 cm at many locations. The minimum thickness specified by the Bear Creek model, will reduce the radon flux and gamma radiation to twice the normal background level.

Soil utilized for ACID and ALKO (Figs. 2 and 3) contains silt and sand in addition to clay and not compacted to Bear Creek specifications. Even so, 400 cm of this soil would reduce the radon flux to the background level. This depth of cover is explicitly for the measured conditions (Table 1) and may be less or more for other tailings depending on the radon flux from bare tailings and radium-226 activity of the soil cover.

In most of the southwestern United States, the rate of surface denudation is less than 15 cm per 1000 year, which is greater than the denudation rate for other parts of the continental United States. At this rate a soil cap of 300 cm protected from erosion by surface water run-off with both rip-rap and vegetation will last for about 20,000 years. Even so, the cover of these tailings will need occasional reconstruction to remedy the local effects of water erosion. In perspective, this period is long in comparison to predictable geological conditions and concern for the integrity of small scale features seems misplaced. For example, less than 10,000 years ago glaciers covered most the north-middle-western states and was responsible for creating lakes and changing the direction of rivers such as the Ohio River.

An alternative for tailings management (NRC78) after decommissioning is below-grade containment, such as planned for Morton Ranch. Below-grade tailings disposal, if it is hydrologically feasible, allows deeper burial of the tailings. Subsurface disposal of tailings results in a configuration which is stable and less prone to surface erosion than above-grade disposal. The choice of either model, Bear Creek or Morton Ranch, is dependent on the site-specific hydrological conditions. Choice of overburden containing substantial Ra-226 activity (> 5 pCi/g) for the top cover of tailings may itself generate a substantial radon flux. Adoption of either models for decommissioning of tailings results in mitigation of future exposures from inhalation, ingestion, and external irradiation.

Alternative methods for disposal of tailings have been discussed in Chapter 10 of the Bear Creek Project environment statement (NRC77) including removal of radioactivity during processing of the ore. At present, more advanced alternative disposal methods such as this are not technologically and financially feasible because the large volume of low level activity.

CONCLUSION

For some of the older mills, the dose commitment from inhalation of airborne tailings particulates, ingestion of contaminated food and external exposure to individuals living nearby may exceed the 25 mrem/year limit. Also, because of fallout of airborne tailings particulates and horizontal transport of tailings sand, a large area of uncontrolled land in the immediate vicinity may be contaminated to an extent which is unacceptable. Since expansion of the land controlled by the mill is often not practical or possible, consideration of alternative procedures for management of uranium tailings is necessary both to achieve compliance with the 25 mrem/year standard and to protect the environment, the mobility of the tailings sands

and fugitive dust should be reduced. A suggested approach is to minimize dried beach tailings surfaces and/or maintain a wet surface condition. The release rates from tailings completely submersed under solution is negligible, but for some mills, submersion is not possible because of the impact of the large surface head area on the groundwater quality. Fig. 9 depicts a suggested alternative procedure for maximizing evaporation and reducing the release rates of both radon and particulates. The multiple discharge of mill effluents minimizes the formation of large, dry deltas and maintains almost the entire surface of the tailings in a wet to moist condition. Recycling of solution over the surface in some cases may not result in a sufficient rate of volume reduction and may require the addition of auxilliary evaporation ponds as depicted in Fig. 9. In other cases, since essentially the entire surface of the main pond is utilized for evaporation, a pond of smaller area but greater depth may become practical. This in turn would reduce the cost of final stabilization of the tailings.

Gravitational settling encourages deposition of the larger sand particles of lower specific activity near the discharge points resulting in relatively greater deposition of finer particles (slimes) with higher specific activity at deeper depths below the tailings solution. This tends to reduce the radioactivity in the more exposed and drier tailings, and therefore, decreases the dose commitments which result from transport of this material. Mechanical separation of sands and slimes prior to discharge is suggested as a useful alternative procedure to augment this natural, differential settling process. Concentration of the slimes in the liquid discharge also reduces solution seepage by closing the channels for leakage.

Above ground storage of tailings after decommissioning in a configuration similar to that depicted in Fig. 9 will require periodic maintenance to insure that they remain isolated from the environment. For this reason, permanent disposal above grade is not recommended if any practical alternatives exist. Below ground level storage will provide more protection from erosion forces and reduce maintenance and, therefore, is the preferred method. In light of higher uranium market prices, reprocessing of some of the older tailings may provide an incentive for reconstruction of tailings retention areas in accord with the regulatory objectives listed by Martin and Miller (Ma78). Application of a minimum of 4 m of cover as a mixture of overburden, clay, soil and rip-rap over the tailings and protection of the slopes at dyke boundaries from erosion may provide an acceptable above ground alternative for storage of tailings if local conditions prevent disposal below grade. Stability of this configuration is dependent on the slope of the dykes, reduction of erosion and control of water run-off. The radiation dose commitment from the isolated tailings should be comparable to that of natural background. Decommissioning of the tailings should progress in parallel with drying of the ponds rather than being deferred until the beaches are completely dry. Deposition of overburden on the wet tailings beaches using presently available technology is both practical and financially feasible. Any protocol for decommissioning of tailings should include both minimization of exposures and assurance of predictable

long-term stability of containment. Management of uranium tailings at all new mills so that the hazard from radon and particulates is negligible during active milling and after decommissioning is feasible and practical. Further studies are needed, however, to provide reliable data for the analysis of reclamation plans in order to insure that they are cost effective.

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TABLE 1. Flux $\phi_t(z)$ (pCi Rn-222/m^2·sec), Specific Flux ϕ (pCi Rn-222/m^2·sec)/(pCi Ra-226/g), Bulk Diffusion Coefficient D_t (cm^2/sec) at Two Tailings Designated ACID and ALKO for Number of Measurements N

Tailing Area	N	$\phi_t(z)$	ϕ	D_t
ACID	55	376.8	0.61	2.4×10^{-3}
ACID.2	49	415.6	0.67	3.0×10^{-3}
ACID.5	6	169.0	____*	4.5×10^{-4}
ALKO	55	190.0	0.32	5.7×10^{-4}
ALKO.2	49	174.2	0.29	4.8×10^{-4}
ALKO.5	6	175.1	____*	5.0×10^{-4}

*ACID.5 and ALKO.5 are covered with soil and Ra-226 concentration has not been determined yet.

TABLE 2. Physical Characteristics of Tailings Solids

Particle Diameter (μm)	Deposition Velocity (m/sec)	Settling Velocity (m/sec)	Fractional Distribution
0.1	1.0×10^{-2}	7.2×10^{-7}	0.02
1.0	1.0×10^{-2}	7.2×10^{-5}	0.03
5.0	1.0×10^{-2}	1.80×10^{-3}	0.25
35.0	8.8×10^{-2}	8.82×10^{-2}	0.50
≥100.	7.2×10^{-1}	7.2×10^{-1}	0.20
Radon release rate:	4700 Ci/year		
Density of tailings:	2.4 g/cm^3		
Tailing area:*	3×10^{-1} km^2		

*This area was subdivided into 16 areas, each with an area of 1.88×10^{-2} km^2.

TABLE 3. Annual Relative Frequency of Occurrence (Sum of All Stability Classes) Metset Bluewater, New Mexico

Wind Direction	Wind Speed, knots						
	0-3	4-6	7-10	11-16	17-21	Over 21	Row To
N	0.01690	0.01560	0.00510	0.00150	0.00060	0.00040	0.040
NNE	0.01390	0.01260	0.00390	0.00100	0.00040	0.00030	0.032
NE	0.02130	0.01990	0.00630	0.00220	0.00070	0.00060	0.051
ENE	0.02100	0.02050	0.00620	0.00220	0.00050	0.00040	0.050
E	0.02060	0.02080	0.00620	0.00220	0.00050	0.00050	0.050
ESE	0.02780	0.02710	0.00880	0.00310	0.00070	0.00040	0.067
SE	0.03320	0.03430	0.00910	0.00250	0.00050	0.00040	0.080
SSE	0.02740	0.02640	0.00730	0.00300	0.00070	0.00050	0.065
S	0.02910	0.02970	0.00310	0.00310	0.00060	0.00060	0.071
SSW	0.03010	0.02910	0.00760	0.00310	0.00060	0.00060	0.071
SW	0.04950	0.04670	0.01280	0.00490	0.00070	0.00040	0.115
WSW	0.03150	0.02790	0.00710	0.00280	0.00030	0.00020	0.069
W	0.02860	0.04740	0.00610	0.00270	0.00080	0.00050	0.066
WNW	0.02850	0.02770	0.00610	0.00260	0.00070	0.00060	0.066
NW	0.02740	0.02880	0.00630	0.00260	0.00070	0.00060	0.066
NNW	0.01540	0.01410	0.00430	0.00140	0.00040	0.00040	0.036
Column totals:	0.42220	0.40860	0.11130	0.04090	0.00940	0.00740	0.999

TABLE 4. Total Inhalation Dose Commitment (mrem/year) from Particulates at the Direction of Maximum Dispersion and Base Case Condition

Distance (km)	Whole-body	Nasopharyngeal	Tracheobronchial	Pulmonary	Bone
0.3	2.26E-01	4.32E+01	9.17E-03	1.86E+00	6.07E+
1.0	1.33E-02	2.31E+00	5.56E-04	1.24E-01	3.64E-
5.0	2.61E-03	1.37E-01	9.11E-05	3.30E-02	7.82E-
10.0	2.46E-03	4.70E-02	7.99E-03	3.29E-02	7.51E-
50.0	2.18E-03	1.08E-02	6.86E-05	2.99E-02	6.73E-
80.0	2.06E-03	9.13E-03	6.48E-05	2.83E-02	6.37E-

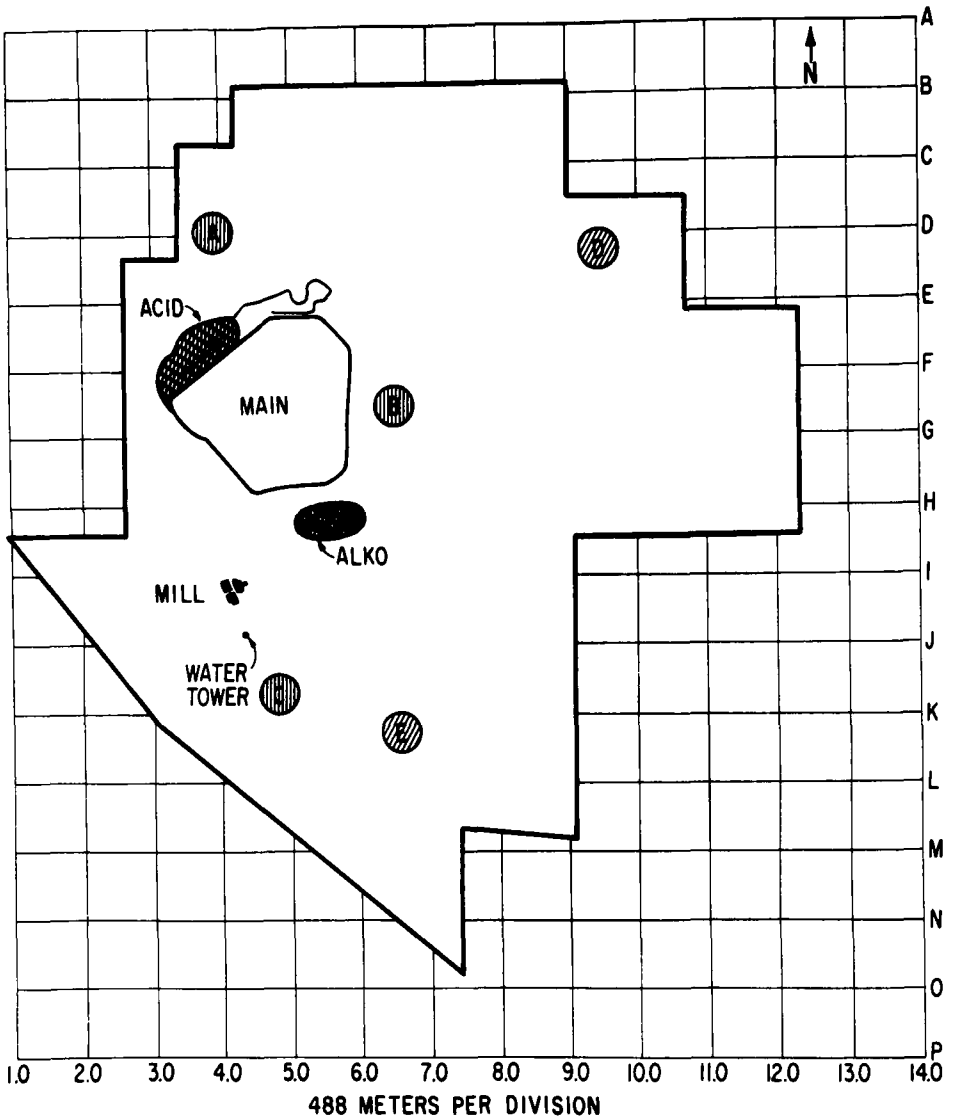
TABLE 5. Total External* Dose Commitment (mrem/year) from Ground Contamination and Airborne Radionuclides

Dose (mrem/year)	Distance (km)					
	0.1	1.0	5.0	10	50	80
Whole Body						
Ground	9.26E+00	2.74E-01	1.23E-02	3.65E-03	6.09E-04	5.05E-04
Airborne	6.58E-01	1.05E-00	5.00E-01	2.35E-01	3.53E-02	1.97E-02
Ovaries						
Ground	6.85E+00	2.03E-01	9.04E-03	2.60E-03	3.49E-04	2.78E-04
Airborne	3.64E-01	7.11E-01	3.69E-01	1.74E-01	2.63E-02	1.46E-02
Testes						
Ground	7.95E+00	2.36E-01	1.06E-02	3.13E-03	5.17E-04	4.28E-04
Airborne	6.87E-01	9.67E-01	4.27E-01	1.99E-01	2.99E-02	1.66E-02
Bone Marrow						
Ground	9.85E+00	2.92E-01	1.33E-02	4.13E-03	8.90E-04	7.67E-04
Airborne	7.85E-01	1.16E+00	5.25E-01	2.46E-01	3.69E-02	2.05E-02
Bone						
Ground	1.07E+01	3.18E-01	1.45E-02	4.51E-03	9.74E-04	8.40E-04
Airborne	8.38E-01	1.24E+00	5.64E-01	2.64E-01	3.96E-02	2.21E-02

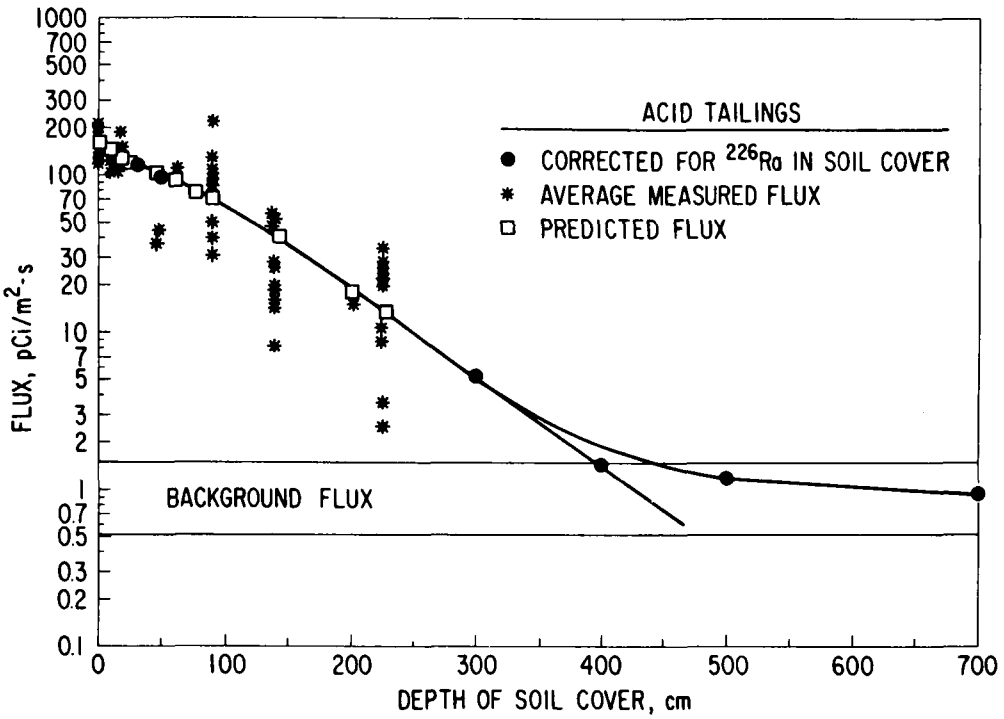
*Direct gamma and beta radiation from the tailings and contamination from creeping sand tailings are not included in these calculations. On tailings pile direct radiation dose to whole body is 1.0×10^4 to 1.5×10^4 mrem/year. But direct radiation at 0.5 km from tailings is small relative to background.

TABLE 6. Dose Commitments (mrem/year) from Inhalation of Rn-222 and Radon Daughters to Bronchial Epithelium under Conditions of Dry Tailings, Moist and Wet Beaches as a Function of Distance from the Tailings and in the Direction of Maximum Dispersion

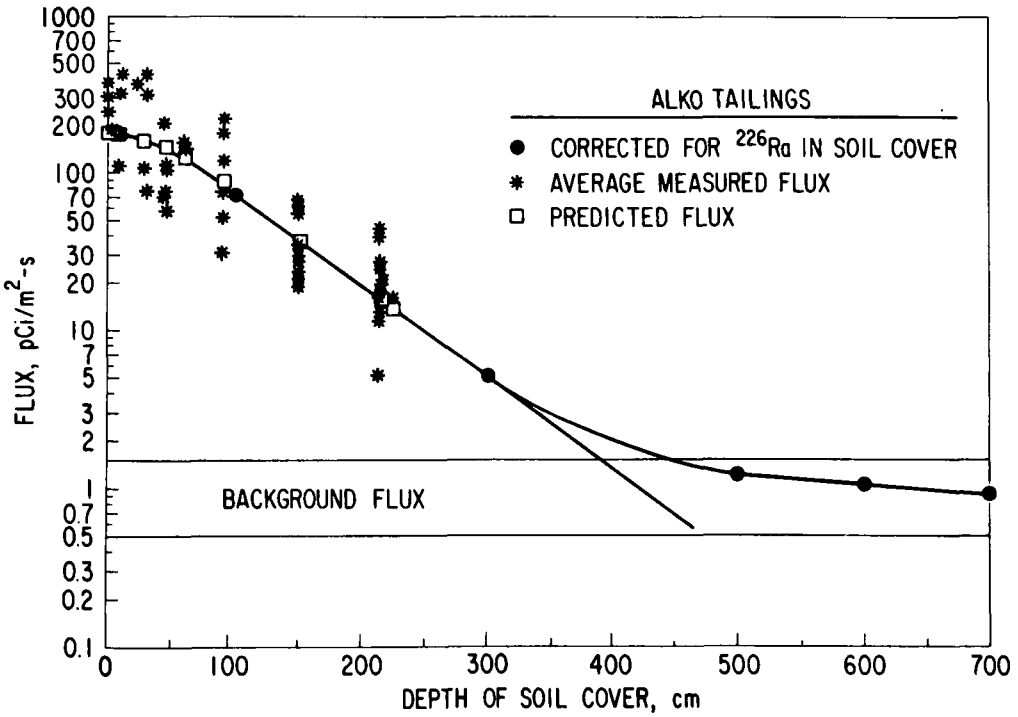
Distance (km)	Dry	Moist	Wet
0.1	6.41E+03	6.41E+02	1.28E+02
1.0	5.74E+02	5.74E+01	1.15E+01
5.0	4.49E+01	4.49E+00	8.98E-01
10.0	1.73E+01	1.73E+00	3.46E-01
20.0	7.13E+01	7.13E-01	1.43E-01
50.0	2.33E+00	2.33E-01	4.66E-02
80.0	1.30E+00	1.39E-01	2.60E-02



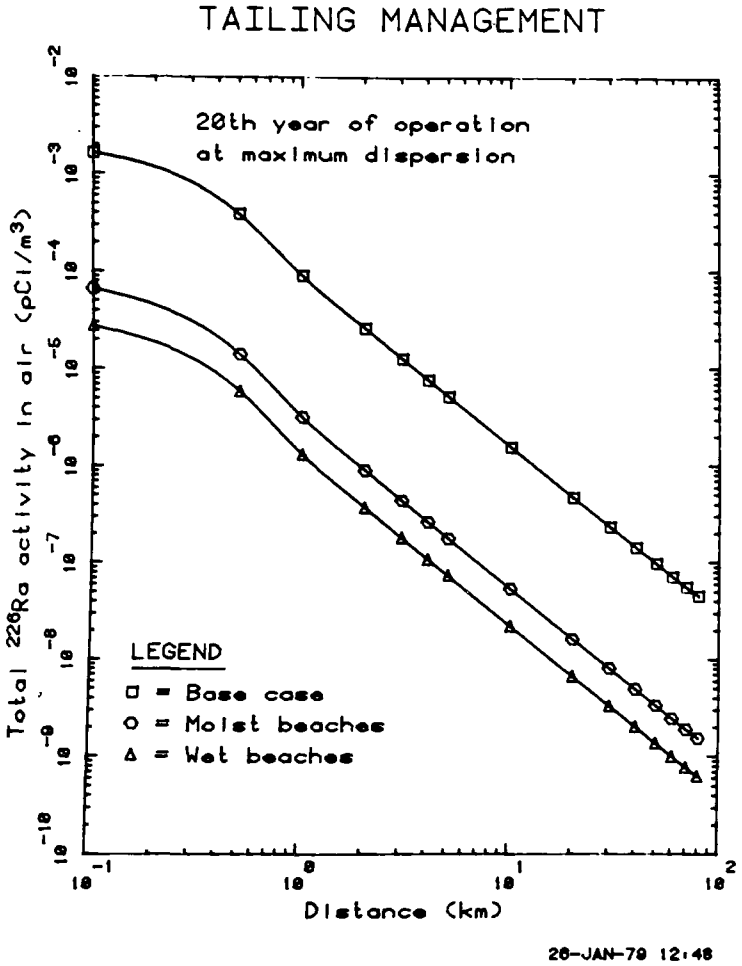
1. Experimental stations at Anaconda Uranium Mill, Bluewater, New Mexico. Areas designated by letters are A (Station #101, #102), B (Station #103), C (Station #104), D (Station #105), and E (Station #100). Inactive Tailings are ACID and ALKO. The tailings retention area still active is designated as MAIN.



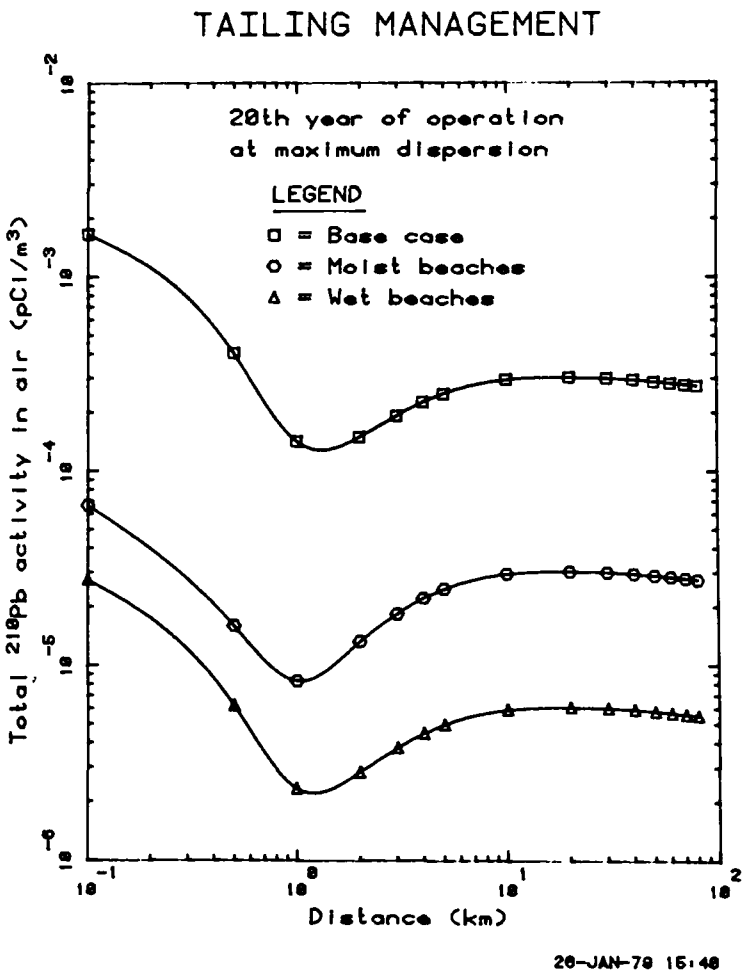
2. Measured flux over ACID tailings (see Figure 1), predicted flux, and flux corrected for the radioactivity content of soil cover, as a function of soil cover thickness.



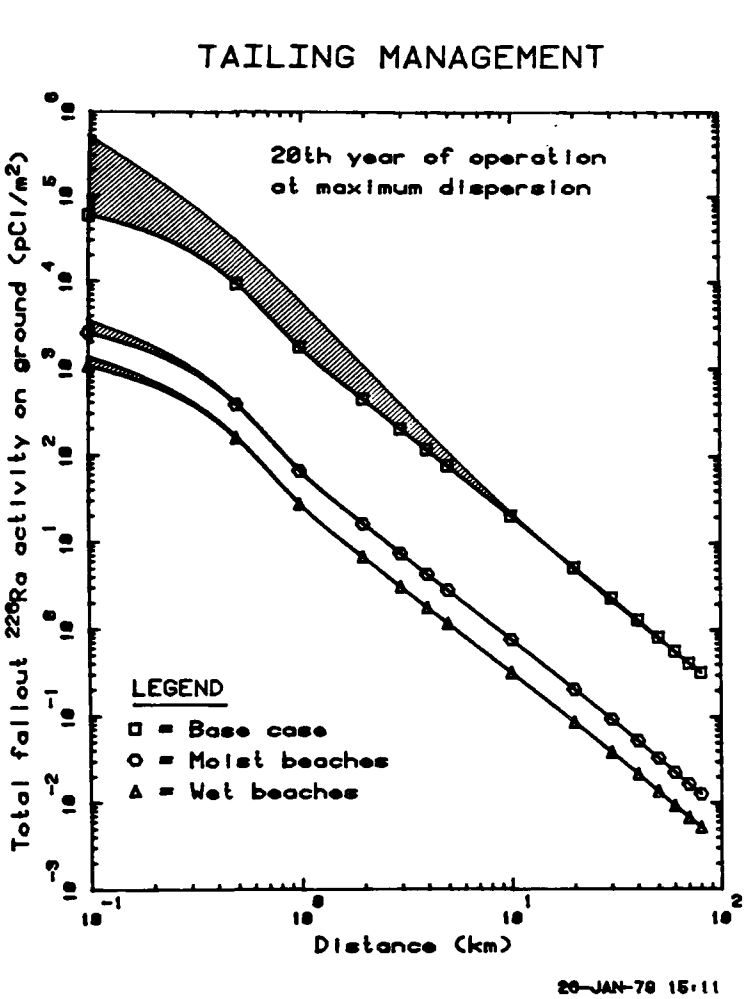
3. Measured flux over ALKO tailings (see Figure 1), predicted flux, and flux corrected for the radioactivity content of soil cover, as a function of soil cover thickness.



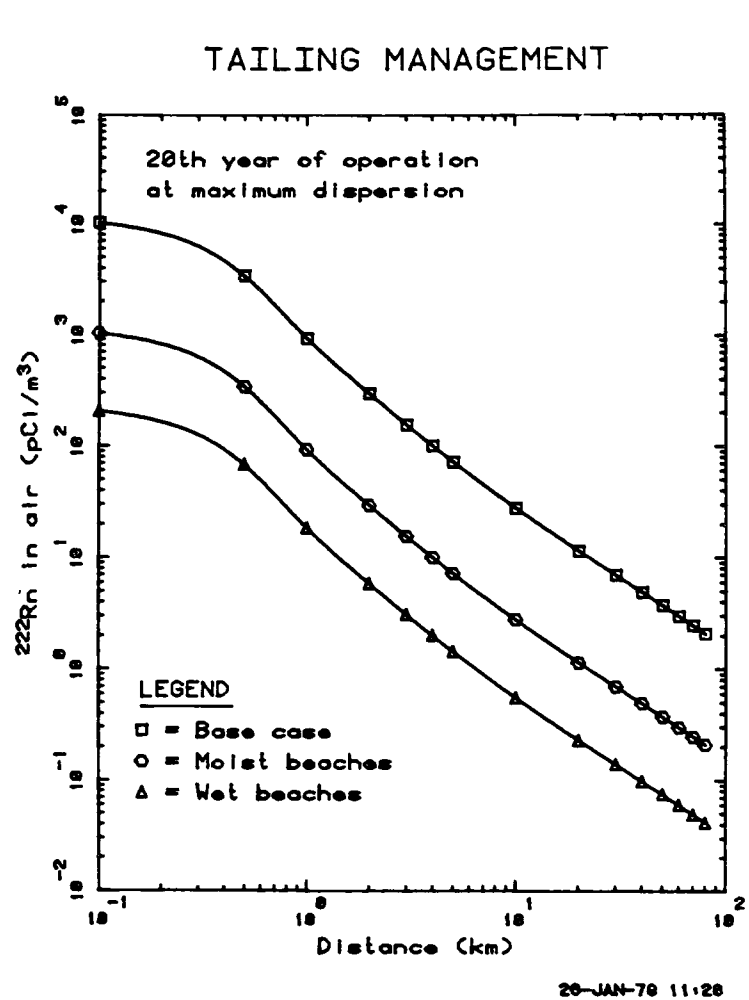
4. Concentration of Ra-226 in air as a function of distance from the geometric center of beach tailings predicted for the direction of maximum transport and for three surface moisture conditions: base case (dry), moist and wet beaches. The symbols superimposed on the curves do not represent the experimental data.



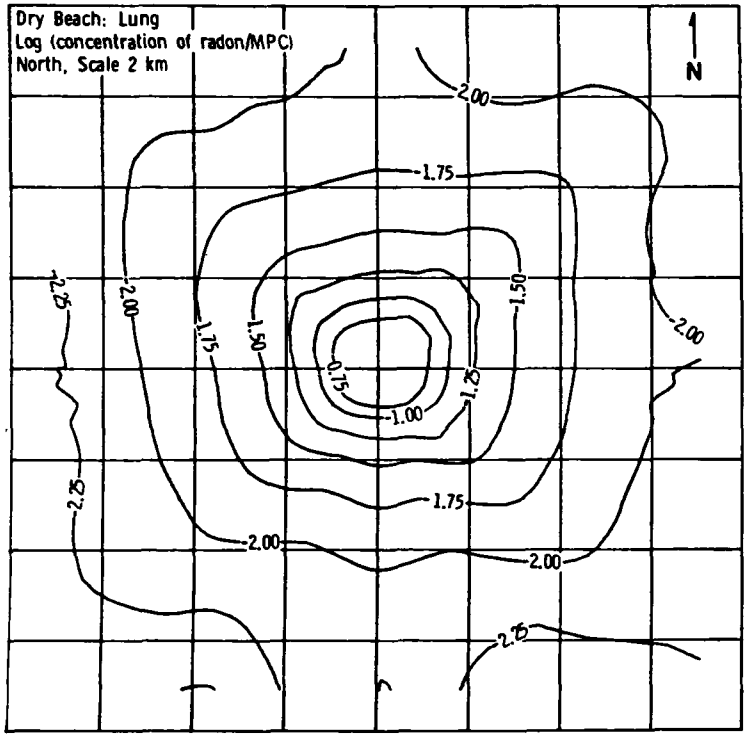
5. Concentration of Pb-210 in air as a function of distance from the geometric center of beach tailings predicted for the direction of maximum transport and for three surface moisture conditions: base case (dry), moist and wet beaches. The symbols superimposed on the curves do not represent the experimental data.



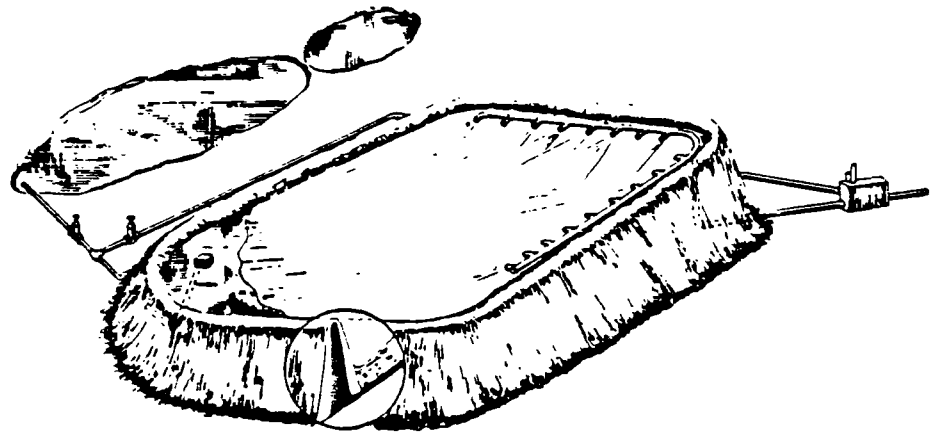
6. Ra-226 contamination of ground as a result of 20 years of mill operation as a function of distance from the geometric center of beach tailings and in the direction of maximum tailings transport, predicted for three surface moisture conditions: base case (dry), moist and wet beaches. The cross-hatched area represents the estimated surface activity from horizontal tailings flux. The symbols superimposed on the curves do not represent experimental data.



7. Concentration of Rn-222 in air as a function of distance from the geometric center of beach tailings predicted for the direction of maximum transport and for three surface moisture conditions (dry), moist, wet). The sumbols superimposed on the curves do not represent experimental data.



8. Safety factor isopleth, i.e., distances from the geometric center of the beach tailings having equal concentration of Rn-222 in air normalized to maximum permissible concentration (MPC) of radon (3 pCi/l). A safety factor of -3 indicates that the concentration of radon-222 in air is only 1/1000 of the MPC.



9. Sketch of a suggested design for minimizing fugitive dust and radon-222 emission from the surface of a tailings pond. Features include multiple ports for discharging mill effluents, recirculation of the solution over the tailings, and excess-solution evaporation ponds with plastic membranes and clay liners to minimize leakage. The retention area design is based on the Bear Creek model, with a clay-cored dike keyed to an impervious base.

DOSE MODELLING FOR RADON-PRODUCING ACTIVITIES IN HEAVILY POPULATED AREAS

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Philadelphia, Pennsylvania

Abstract

The Area Source Radiological Emission Analysis Code (AREAC) was used to estimate population and individual exposures resulting from radon effusion sources in two heavily populated areas. One situation results from disposal of radium and uranium processing wastes at a plant site over the past sixty years. The other situation occurs as a result of mining operations.

The mining operation results in radon contamination of the air near a metropolitan area. The source could affect 234, 386 persons living within a ten-mile radius. Results of the AREAC model calculations for radon diffusion and subsequent population exposure using meteorology and demography for the region are presented.

The waste disposal situation was modelled using measured concentrations of radium in soil and relating these concentrations to radon emanation rates and consequent area source strengths for radon. This provided input to the diffusion calculations employing regional meteorology and demography. The resulting radon concentrations at specific receptor sites and at sector centroids gave population exposure estimates. The disposal site is located in an area of many small population centers on the fringe of a metropolitan area. 47, 351 persons live within ten miles.

Introduction

This paper discusses use of a computer code to calculate the radiological impact of airborne radon in two situations involving relatively large populations. The results presented herein are theoretical ones from the computational model. The purpose of the work was to develop a tool to evaluate the effectiveness of proposed control measures in reducing population exposure. Another use of the computational model would be to guide a field measurement program and to provide a rational method for interpolation and extrapolation of a few well-chosen field measurements in the area of the radon source. No experimental radiation exposure data was obtained in the course of this work.

The computational tool used was AREAC (Area Source Radiological Emissions Analysis Code), developed by the U. S. Environmental Protection Agency's Office of Radiation Programs (Mi76). The code model was applied to a radium and uranium waste disposal site and to a radon-emitting mining operation using regional meteorological and demographic data. The paper briefly describes the computational methods used and presents results in terms of exposure at particular locations and to area populations as the consequence of radon gas emission in the two situations.

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The AREAC code applies the well-known Gaussian diffusion equation to an emission source of finite area. The source geometry can be either rectangular or circular, For meteorological input, the code uses the joint probability distribution for wind velocity and atmospheric stability (S168), along with an algorithm for atmospheric dispersion (Tu71) to generate dispersion coefficients from each of the specified source points to, first, up to 6 specific receptor sites and, second, to the 16 azimuthal sectors and up to 12 radial segments for population around the source. The dispersion coefficients multiplied by the area source strength and specified dose conversion factor to compute annual radiation exposure to individuals at the specific receptor sites and to the population distribution specified by angular and radial segment about the source.

A Mining Operation Emitting Radon

The modelling method and the results obtained can be better understood when applied to a relatively simple situation. In this situation, a mining operation emits approximately 6 microCuries per second of radon into the atmosphere as radon-contaminated ventilation air from an exhaust fan house, essentially a point source. The emitter is located between two metropolitan areas. Total 1970 census population within a ten-mile radius of the site was 234, 386. Two available sets of meteorological data were used in the calculations, since no specific data was obtainable nor directly applicable to the emission site. One data set was the ten-year average for the regional airport, about 8 miles distant; the other data set was from a nuclear power plant site 26 miles away. Neither set was truly appropriate for the emission site because of differing topography. The two sets of meteorological data also differ considerably. Nevertheless, computed individual and population doses are comparable, demonstrating that the modelling is not very sensitive to meteorology.

A dose conversion factor of 4.0×10^{12} (mrem/yr)/(Curie/m^3) (EP77) is used throughout this work. Resultant population exposures for the two meteorological regimes are presented graphically in Figures 1. and 2. Total population dose within the ten-mile radius and maximum individual dose within one mile are 293 person-rem/yr and 313 mrem/yr and 475 person-rem/yr and 336 mrem/yr for the cases of airport meteorological data and nuclear plant site meteorological data, respectively. This theoretical model employed a point source of emission directly into the atmosphere with straightforward diffusion to obtain the results presented. The next situation is more complex and demonstrates the area source feature of the AREAC code.

Waste Disposal Involving Radium and Uranium

The second situation modelled was the theoretical radiation exposure resulting from radon emitted by an old site contaminated by radioactive wastes from radium and uranium mineral processing. A detailed radiological survey (D078) includes measurements of soil contamination levels of ^{226}Ra on the site and its environs and as a function of soil depth for selected bore samples. Schiager's method (Sc74) for estimating radon

flux into the atmosphere from radium concentration in soil was used:

$$\phi_{Rn}\ (\text{pCi/m}^2\cdot\text{sec}) = 1.6\ C_{Ra}\ (\text{pCi/g}).$$

Schiager tested this relationship against measurements for the Salt Lake City Vitro uranium mill tailings pile with reasonable agreement - 6.8 pCi/l calculated radon concentration compared with a measured value of 10 pCi/l. For this work, the Schiager relationship was tested against 1975 field measurements for the Climax pile (Du77) with the following result: Calculated radon concentration at pile edge, 22 pCi/l; measured values, SE corner 30, NE corner 22, center 15, SW corner 38 and NW corner 26 pCi/l. The agreement was considered satisfactory.

Two source terms were computed. One was an areal average of radium concentration through the upper 6 feet of soil over the entire site. The other source used only the hot spot or relatively small area of high radium concentration near the surface in one place on the site. As in the first situation, two different sets of meteorological data were used. One set was from the regional airport 17 miles away. The second set was from a nuclear power plant site 30 miles away. Again, neither set was really appropriate because of the differing topography.

Results for the two differing sets of meteorological data generally agreed within a factor of 2 for individual and population doses which could theoretically result from radon emanation at the site to the 47, 351 persons residing in 1970 within a ten-mile radius. Figure 3 depicts population doses using a circular hot spot of radon emanation corresponding to a soil concentration of 760 pCi/g ^{226}Ra and airport meteorological data. Figure 4 shows the theoretical population dose estimates for the situation with 88 pCi/g over a rectangular site, again using airport meteorological data. A summary of population doses is given in Table 1. The model was also used to obtain estimated individual exposures at specific locations close to the site. A summary of the results of these calculations is given in Table 2.

Conclusion

A computational modelling method for estimating individual exposure and population dose has been described. The method and its application to two different sources of radon emission into the environment were presented. The model provides a tool to evaluate various radon emission control alternatives. The computational results obtained should be compared with field measurements and the model appropriately adjusted if necessary to utilize the method to obtain valid population dose estimates for these particular radiological situations and others like them.

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Du77 Duncan, D., G. Boysen, L. Grossman and G. Franz, Outdoor Radon Study (1974-1975), Technical Note ORP/LV-77-1, U. S. Environmental Protection Agency, Las Vegas, Nevada

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EP77 Radiological Quality of the Environment in the United States, 1977, Report EPA 520/1-77-009, U. S. Environmental Protection Agency, Washington, D. C. 20460
Mi76 Michlewicz, D., 1976, Area Source Radiological Emission Analysis Code, Report EPA 520/1-76-017, U. S. Environmental Protection Agency, Washington, D.C. 20460
Sc74 Schiager, K., 1974, "Analysis of Radiation Exposures on or Near Uranium Mill Tailings Piles", Radiation Data and Reports, 15, 411
Sl68 Slade, D., 1968, Meteorology and Atomic Energy, U. S. Atomic Energy Commission Report TID 24190, Oak Ridge, Tennessee
Tu71 Turner, D., 1971, Workbook of Atmospheric Dispersion Estimates, U. S. Environmental Protection Agency Publication AP-26, Research Triangle Park, North Carolina, 27711

TABLE 1

SUMMARY OF POPULATION DOSES NEAR DISPOSAL SITE

outer radius, miles	hot spot source Met 1	hot spot source Met 2	averaged source Met 1	averaged source Met 2
0.5	291	263	209	196
1	187	134	133	95
2	x	51	x	36
3	x	10	x	7
4	x	4	x	3
5	x	7	x	5

Population doses are person-rem/year

TABLE 2

COMPARISON OF RADIATION DOSES NEAR DISPOSAL SITE

distance, m. and direction	hot spot source Met 1	hot spot source Met 2	averaged source Met 1	averaged source Met 2
125, SSW	x	x	x	x
261, N	1.70	0.64	x	x
283, ENE	1.14	0.76	x	0.72
291, NE	1.58	0.70	x	0.50
335, E	0.46	0.54	x	0.48
753, WSW	0.16	0.05	x	0.05

Radiation doses are in rem/year
x indicates no calculation performed
Met 1 is meteorological data from nuclear plant site
Met 2 is meteorological data from regional airport

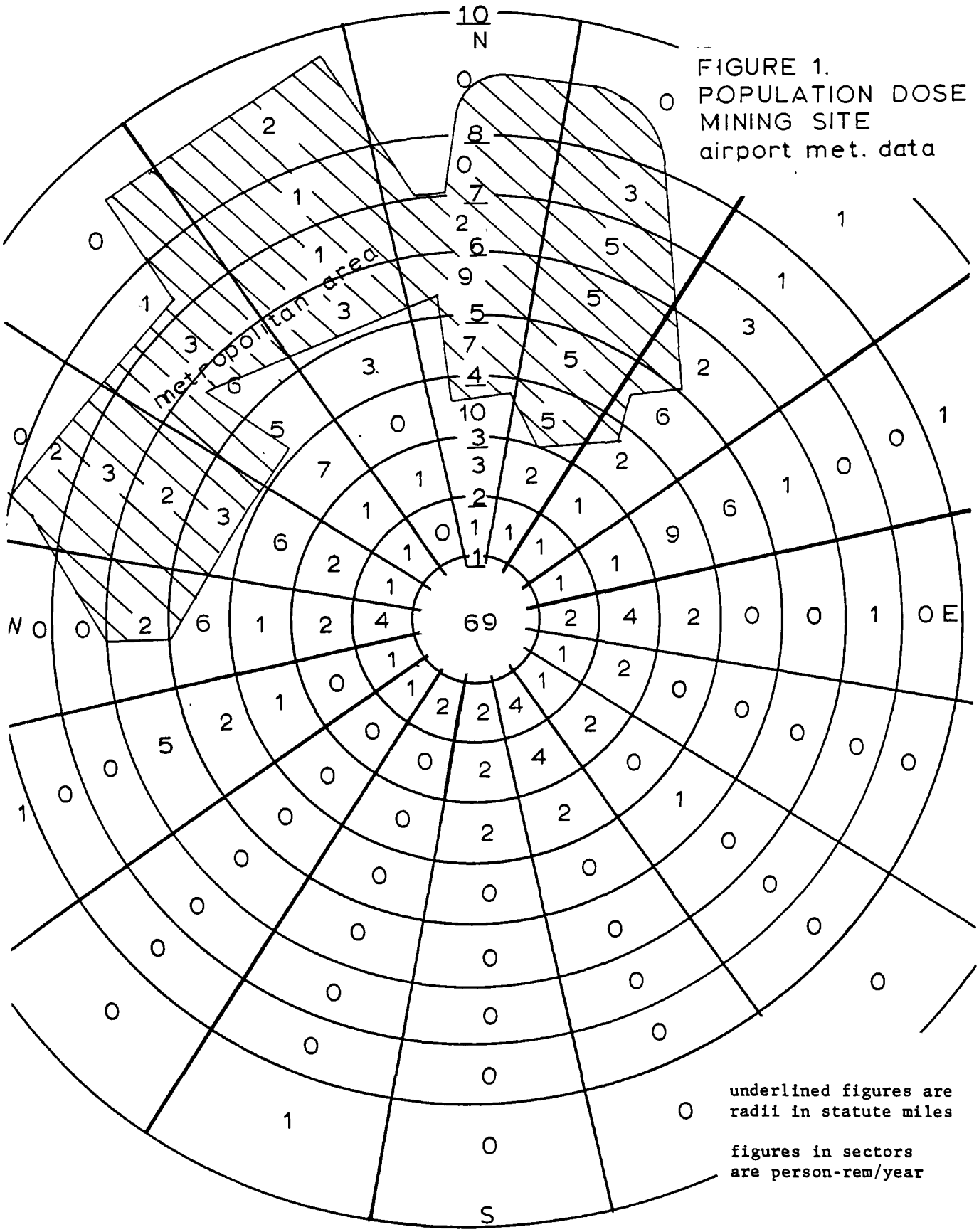
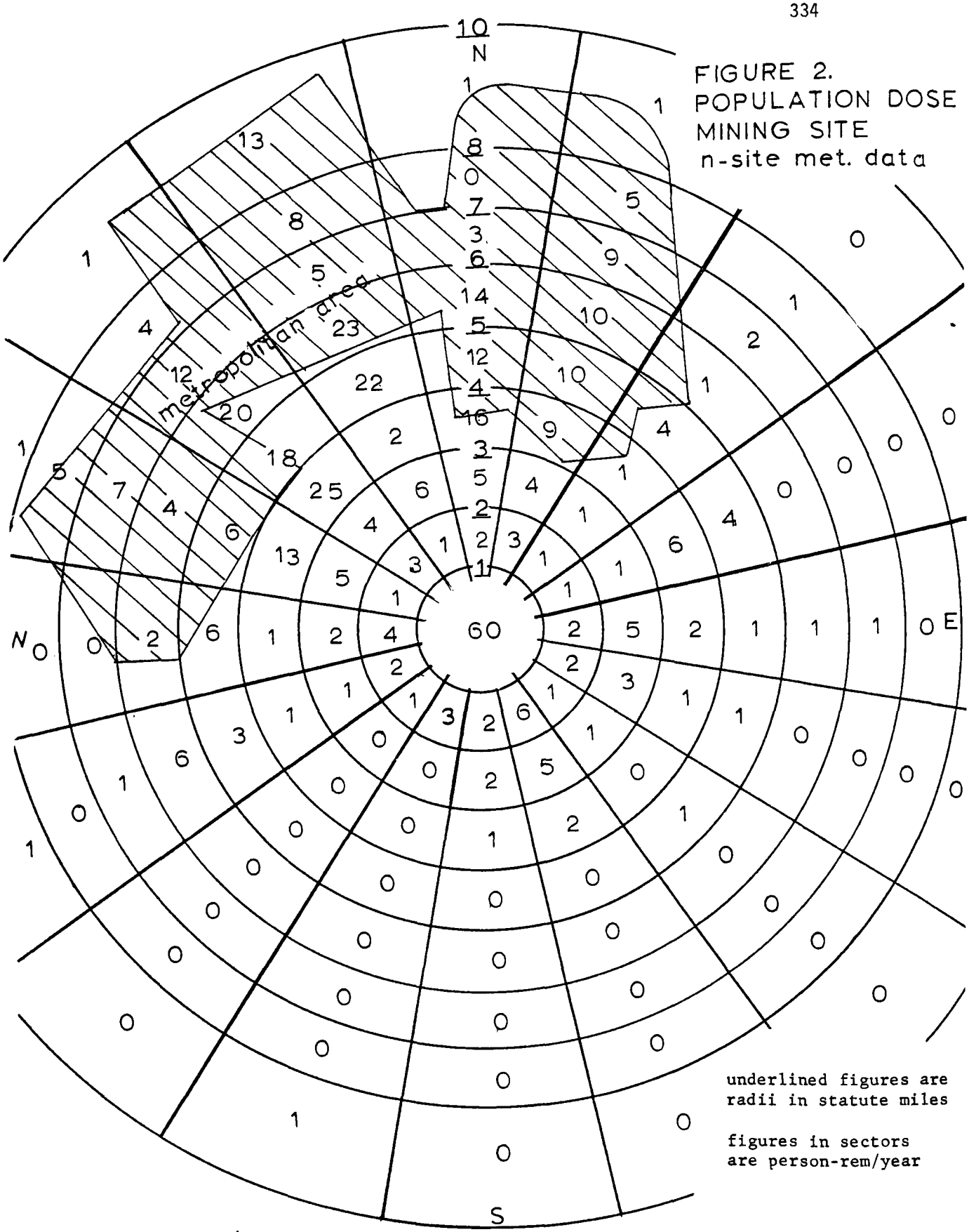


FIGURE 1.
POPULATION DOSE
MINING SITE
airport met. data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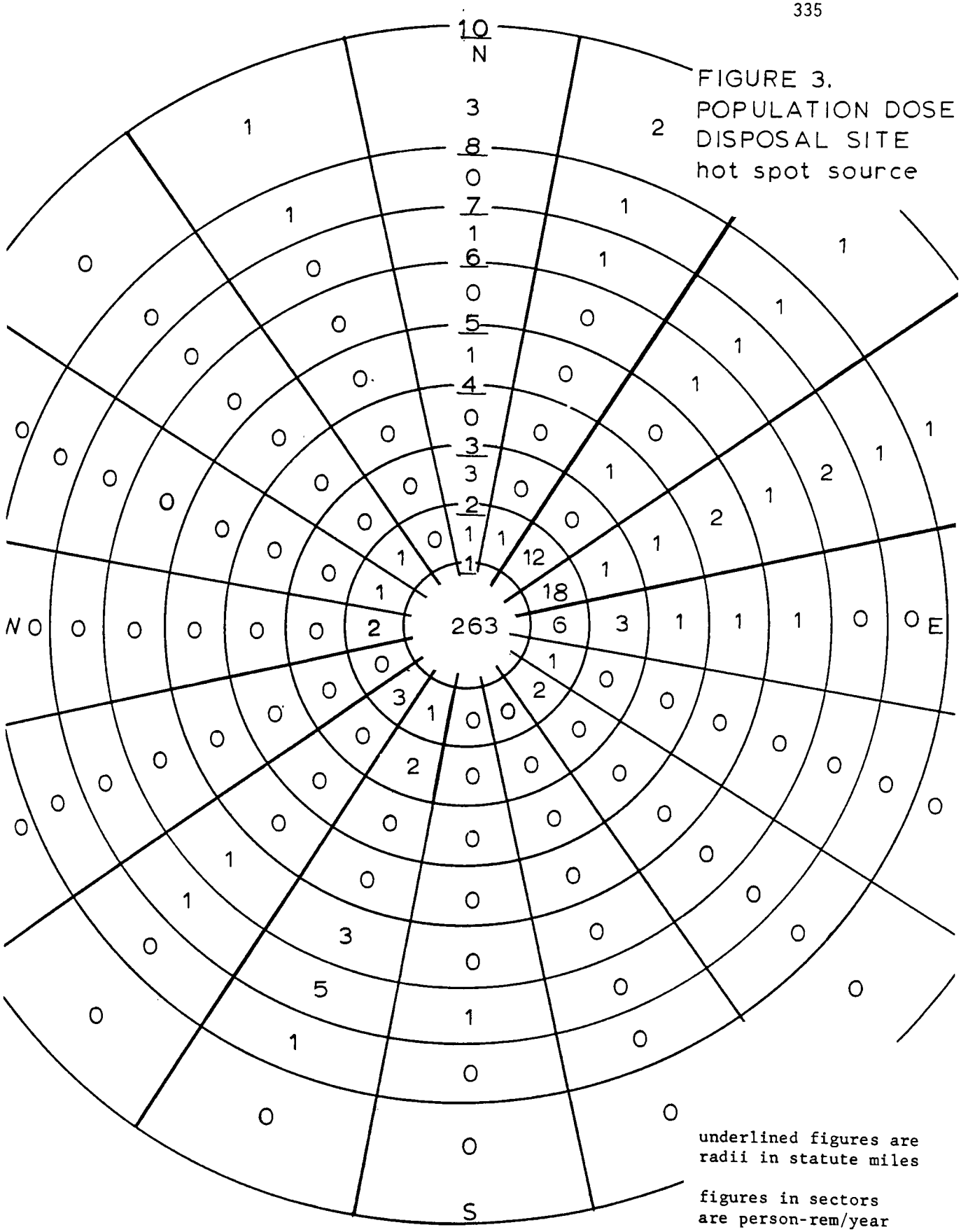

FIGURE 3.
POPULATION DOSE
DISPOSAL SITE
hot spot source

underlined figures are radii in statute miles

figures in sectors are person-rem/year

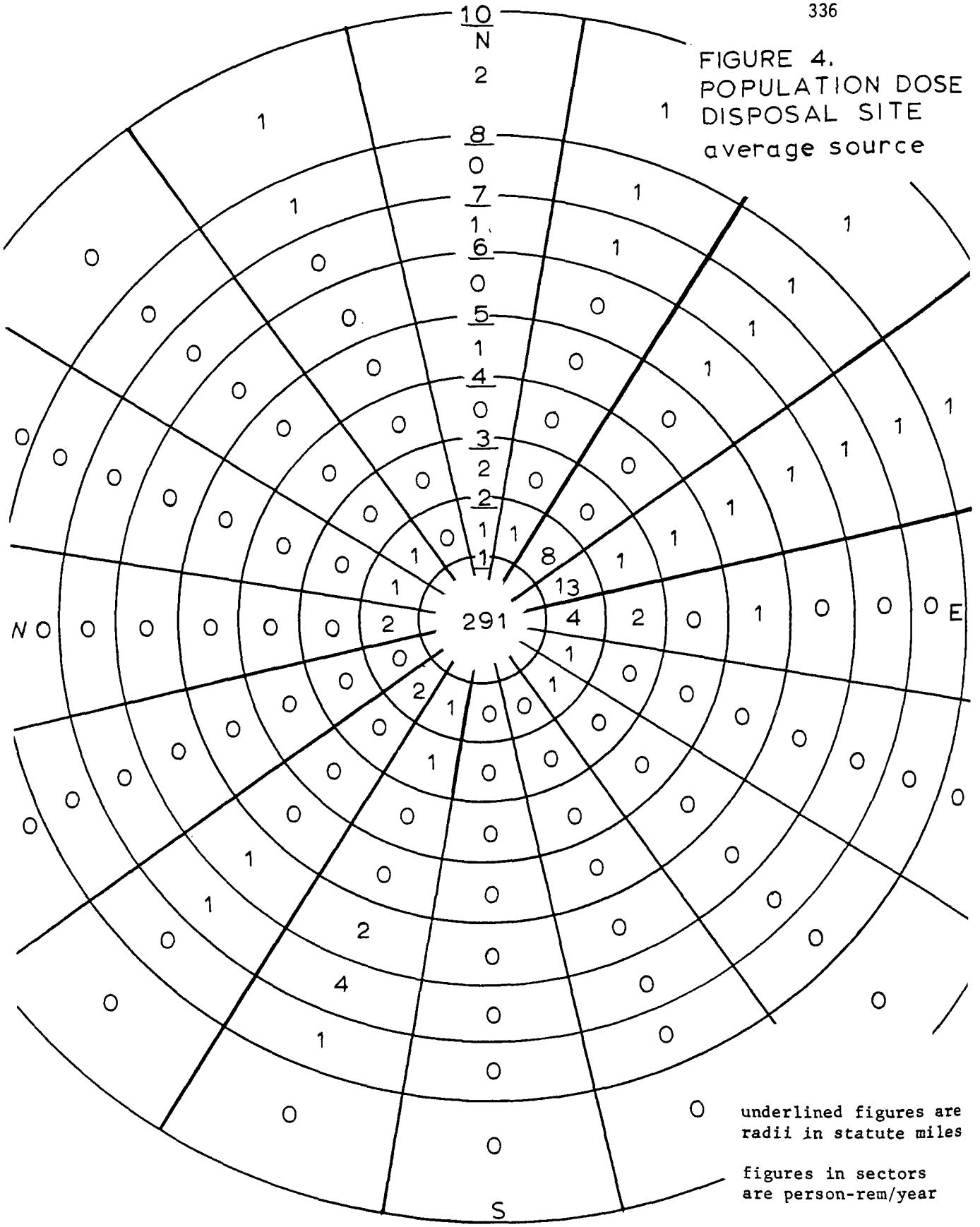

FIGURE 4.
POPULATION DOSE
DISPOSAL SITE
average source

RA-226 CONCENTRATIONS IN THE HYDROGRAPHIC BASINS NEAR URANIUM MINING AND MILLING IN BRAZIL

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"Abstract". A monitoring survey of the ^{226}Ra concentrations in river waters in the vicinity of the mining area and future milling facilities in the Poços de Caldas region began in January 1977. The objective of the monitoring survey is to establish a baseline to allow future comparisons between the ^{226}Ra concentrations in waters of the hydrographic basins of the Poços de Caldas plateau before and after the beginning of full scale commercial operations. Open pit mining started in July 1977 in the uranium deposits of Campo do Cercado, but the main uranium body has not been reached yet. Seasonal variations in riverflow are apparently accompanied by little variations in the ^{226}Ra concentrations in river waters. A crude calculational dosimetric model is in the process of being developed to estimate annual dose equivalent to an individual from ^{226}Ra via drinking water and irrigation patterns as a first step to calculate the collective dose equivalent commitment to the population of the Poços de Caldas plateau and surroundings.

"Introduction"

The brazilian region of Poços de Caldas is a remarkable example of non-explosive volcanic intrusive. The radiogeology of the Poços de Caldas region has been described by Roser et al. (Ro64), and later supplemented by other authors (Cu76)(Ad77). The Poços de Caldas plateau was formed when the central part of the alkaline plug, 35 kilometers diameter, thrust its way 400 meters above the adjacent substratum. Subsequently the central part of the plug, was lowered by chemical weathering and erosion , and further mineralization occurred (Cu76). Uranium deposits were discovered in this region by Frahya in 1948 (Fr50).

Most uranium ores from the Poços de Caldas deposits allow to recover about 0.2% U_3O_8(An74). There are two main uranium deposits in the Poços de Caldas plateau, namely, Campo do Cercado, and Campo do Agostinho; totalizing over 10000 metric tons U_3O_8 (An74). In the latter mining operations did not started yet. Although mining operations in the Campo do Cercado began in July 1977, the main uranium body has not been reached yet. Five hundred metric tons U_3O_8 are expected to be extracted annualy from Campo do Cercado through open pit mining when the operations will reach the commercial level. This projected annual production will enable PWR reactors to generate about 2.9×10^3 Mw(e)/year (Pa77). However, in order to extract annually 500 metric tons U_3O_8, a total amount of 1.40×10^6 metric tons of gross mineral ores will be mined at 18% efficiency (Fe78). This total amount of ore, after being processed, will be converted into 1.05×10^6 metric tons of sterile ores (i.e., < 0.01% U_3O_8) plus 3.5×10^5 metric tons of sludge. The sterile ores will contain about 30 g ^{226}Ra , while the 1.4×10^2g ^{226}Ra, initially in secular equilibrium with the ^{238}U

content of the 500 metric tons U_3O_8 extracted per year, will be contained in the sludge mixed in a volume of the order of $10^5 m^3$ liquid plus solid phases. Therefore, the concentration of ^{226}Ra in the sludge is expected to be at the μCi/ℓ level.

Figure 1 shows a map of the Poços de Caldas plateau with the location of Campo do Cercado indicated by a circle. Conventional leaching processes (Wo58) are being tested to extract uranium from the ores of Campo do Cercado. Milling facilities will be located in the same geographical region where the mining operations of Campo do Cercado are taking place.

The future tailings from the mining and milling operations in the Poços de Caldas region are expected to be kept in a way that ^{226}Ra leaching can be reduced as much as possible. However, the stabilization procedures to be adopted and the actual percentage of ^{226}Ra which will leach from the tailings are still unknown.

Although it is well recognized today that the radioactive contamination of water utilized in uranium mining and milling should be avoided, and coal mining may be more damaging for a local environment than uranium mining (NP77), detriment may result from the uranium mining and milling operations in Poços de Caldas in the case of significant ^{226}Ra contamination of waters of the hydrographic basin of the region. A recent study carried out under the auspices of the American Physical Society concluded that "for regional and local population exposure, radionuclides in uranium mill tailings are potentially at least as important as the actinide chain elements in high-level waste; the relative accessibility of mill tailings contrast with the isolation proposed for other actinide-containing wastes" (AP78). The rationale behind this statement becomes transparent through the fact that the long half-life of ^{226}Ra (1.602×10^3 years) imposes the necessity of considerable effort to find a solution to prevent any amount of ^{226}Ra, large enough to result in detriment, from leaching from the tailings of commercial uranium milling operations. To the best knowledge of the authors, there is not yet any long term solution for segregating, efficiently, uranium mill tailings from the surrounding environment, therefore, monitoring the environment is still the most effective way to detect faulty procedures or fails in the waste management of the tailings from uranium mining and milling operations. The local water contamination by ^{226}Ra resulting from uranium operations can only be assessed with any degree of confiability if the baseline of natural radioactivity in the surrounding region is previously established for future comparisons. A number of investigators have published reports of the impact on the local environment from operations in several parts of the world involving extraction and treatment of uranium for nuclear industry (Ru61)(Ts63)(Ha68)(Ha70)(Iy70)(Ki71)(Ka75)(Pr76)(Ea77)(Ko78), but the only report so far dealing with baseline studies prior to the beginning of uranium operations at any particular site is the one related to the work now being developed in Australia (Br78).

The monitoring survey of the ^{226}Ra concentrations in the waters of the main hydrographic basins of the Poços de Caldas plateau started in January 1977, six months before the beginning of the uranium mining opera-

tions in the region.Other investigators are undertaking studies on sedimentation of ^{226}Ra and on ^{222}Rn emanation from soils and atmospheric diffusion.

A simulation model to calculate the internal alpha dose to an individual from ^{226}Ra intake via the pathways of drinking water and irrigation paterns is in the process of being developed based upon scenarios consistent with today's state of art. This model will constitute the basis to calculate the collective dose commitment to the population of the Poços de Caldas plateau and surroundings.

"Sampling and Experimental Procedures"

The water drainage of the Poços de Caldas plateau occurs by three natural streams. The Rio das Antas crosses the plateau in the general direction south-to-north, as can be seen in the map shown in Figure 1, and drains about seventy percent of the waters which fall on the plateau . The Rio das Antas will be the natural recipient of liquid wastes from the mining operations in the Campo do Cercado and Campo do Agostinho, as well as from the milling operations. There is a plan to use water from Rio das Antas for the water supply system of the city of Poços de Caldas (PD70), located near the northern edge of the plateau. Two small dams , Saturnino de Brito and Bortolan, receive waters from the Rio das Antas, which is also tributary of the Rio Lambari. The latter together with Rio Pardo are the main contributors to the water volume of Graminha dam which appears in Figure 1. The Graminha dam covers thirty square kilometers and is near two cities, Palmeiral on the eastern shore and Caconde located five kilometers from the dam in the northwestern direction. The Rio Verde flows into the Rio Pardo forty kilometers downstream from its source, which is located ten kilometers from the center of Campo do Cercado in the southeastern direction, after draining about twenty percent of the waters that fall on the Poços de Caldas plateau by crossing the border of the volcanic intrusive through the eastern edge. The Rio Verde will also receive part of the liquid wastes from the mining operations in the Campo do Cercado. This river passes near two cities, Pocinhos inside the border of the plateau,and Caldas outside. A brook called Ribeirão da Prata, which runs across the city of Águas da Prata outside the western border of the volcanic intrusive, plus other smaller natural streams around the plateau,drain under ten percent of the water system.

Water samples are taken monthly from 28 sites of collection which are shown in Figure 1, and represent approximately the part of the hydrographic basins most likely to be affected by the uranium mining and milling operations. Water samples are collected in one liter plastic bottles and transported to Rio de Janeiro about five hundred kilometers apart from the collection sites without previous filtration and acidification. Once in the laboratory in Rio de Janeiro, the water samples are filtered through 0.45μm membrane filter and are subsequently acidified to a pH 2 or below with hydrochloric acid. Comparisons between samples filtered and acidified in the site of collection and then transported and those transported without these two preliminary procedures have shown that the adsorption rates in the first few weeks after collection differ very little. Less than five percent of the ^{226}Ra presented initially in solution is lost onto the walls of the recipients in which

the samples are transported without previous filtration and acidification. On one hand filtration through a 0.45μm membrane filter is a time consuming procedure and cannot be easily performed in the collecting sites of the Poços de Caldas region. On the other hand, acidification without previous filtration might prove misleading for determining the ^{226}Ra concentrations of the water samples, because if the suspended solids would contain significant amounts of ^{226}Ra, the acid solution could cause leaching, thereby resulting in wrong values for the ^{226}Ra concentrations in the water.

A simpler version of the well known de-emanation method (Ha67) is used to determine the ^{226}Ra concentration in the water samples. After filtration and acidification, a 120 ml aliquot is taken from each water sample for ^{222}Rn ingrowth into a 150 ml volume capacity bubbler. Co-precipitation with barium is not used because the levels of ^{226}Ra concentration of interest are high enough to be detected by direct de-emanation of ^{222}Rn, which is at a known state of equilibrium with ^{226}Ra in solution, from the bubbler into a scintillation flask. Light pulses from the scintillation flask are counted in a photomultiplier coupled with high voltage supply, scaler and timer, after equilibrium between ^{222}Rn and short-lived daughters is established. The experimental lower limit of detection with these simple procedures is about 0.20 pCi$^{226}Ra/\ell$. The detection system has been inter-calibrated with other somewhat similar systems used around the world through the International Atomic Energy Agency (IAEA) Coordinated Programme on Studies on the Source, Distribution, Movement and Deposition of Radium in Inland Waterways and Aquifers.

"Results and Comments"

Results of the analyses of the ^{226}Ra concentrations in the waters of Poços de Caldas region are shown in Table 1 for the wet and dry seasons, represented by the months of January and July respectively. In Table 1 the letters A, M, L, and R denote collection sites associated with the water system of the Rio das Antas, letter V denotes collection sites in the Rio Verde and some small tributaries, F-1 represents a collection site in the southern base of Morro do Ferro, and collection sites denoted P-1, P-2, and P-3 are located, respectively, in the Ribeirão da Prata in the city of Águas da Prata, in the source of mineral water called Fonte do Villela also in the city of Águas da Prata, and in the Ribeirão do Quartel which flows directly into the Ribeirão da Prata.

The ^{226}Ra concentrations in the wet season are, in general, slightly higher than those recorded in the dry season, though the variations are within the range of the experimental errors. However, for waters collected in the sites denoted by R-3 and P-3 the results for the dry season are higher than those for the wet season. Tentative explanations for the results, apparently anomalous, of R-3 and P-3 may be as follows: (i) R-3 is a collection site located in the Rio das Antas after the points of release of the sewage system of the city of Poços de Caldas, so ^{226}Ra may be dumped into the river with contaminated sewage: and (ii) P-3 is a collection site located in the Córrego do Quartel, in the town of Cascata, where in the dry season the water is usually clear, but in July 1977 there was resuspension of sediments because of the construction of

a small barrage upstream, so the ^{226}Ra might have been leached out of the suspensed sediments increasing the ^{226}Ra concentration in the water. Another anomalous result appears to be the high ^{226}Ra concentration (i.e., 3.9 $pCi^{226}Ra/\ell$) of the water sample collected in January 1977 in the site A-8, followed by non-detected ^{226}Ra concentrations (i.e., < 0.20 $pCi^{226}Ra/\ell$) in the months of July 1977 and January 1978. However, no plausible explanation has been found so far for this fact.

The water samples collected in the site P-2 present the highest ^{226}Ra concentrations in all months shown in Table 1. Such water samples are from a source of natural mineral water (Fonte do Villela). An earlier analysis of this mineral water has resulted also in a ^{226}Ra concentration about 30 $pCi^{226}Ra/\ell$ (Ha74). The water samples collected from the Ribeirão da Prata at the site P-1, 30 meters apart from the Fonte do Villela, have non-detected concentrations (i.e., < 0,20 $pCi^{226}Ra/\ell$), which indicates that the river water is not affected by the ^{226}Ra source for the Fonte do Villela nearby.

The water sample collected in the site M-1 in January 1977 presented the ^{226}Ra concentration of 26 $pCi^{226}Ra/\ell$. This site of collection has been destroyed by mining excavations in the center of Campo do Cercado.

The geographical configuration of the Poços de Caldas plateau is such that two hills, Morro do Cercado, and four kilometers north of this, Morro do Ferro, constitute the main elevations that divide the waters that fall in the region. The flowrate of Rio das Antas are shown in the graph of Figure 2a. Data on the rain precipitations in the Poços de Caldas region are summarized in the graph of Figure 2b; where the dots represent the monthly average of pluviometric records for the year 1977, and the vertical bars indicate the range of monthly average values recorded between 1961 and 1968. Observing the graph of Figure 2b one can conclude that in general the dry season is characterized by rain precipitations lower than 100mm/month between May and September, with a minimum in July. The maximum rain precipitation is usually reached in January, characterizing the peak of the wet season. The flowrate of Rio das Antas follows the pattern of rain precipitation in the Poços de Caldas plateau, and as can be seen from Figure 2a, the flowrate of this river in the wet season is about four times higher than in the dry season.

Figure 2c shows the monthly ^{226}Ra concentrations, for the year 1977, of the water samples collected in the sites M-4, A-1, and V-2, located respectively in the mining area, Rio das Antas, and Rio Verde. The ^{226}Ra concentrations in waters collected in the sites M-4, A-1, and V-2 had different distributions throughout the year 1977. According to Figure 2c, water samples collected in A-1 appear to have had slightly higher concentration in the wet season than in the dry season. Of course, further data are still necessary to confirm such observation. However, this can be explained by the following reasoning: on the one hand, the volumetric increase of water in rivers during the wet season dilutes the ^{226}Ra concentration in the river water; on the other hand, the amount of ^{226}Ra in solution in river water can be enhanced by the resuspension of ^{226}Ra adsorbed in the sediments. In the rivers of the Poços de Caldas region, the latter effect seems to predominate. Variations of ^{226}Ra concentration in waters as a function of time of the year have already been observed

elsewhere (Pa78), although under different conditions.

The water samples collected in M-4, which is located at a point only 2,5 km apart from the center of the mining operations of Campo do Cercado in a small tributary of Rio das Antas, presented, in 1977, ^{226}Ra concentrations higher than those water samples collected either in A-1 or V-2. Furthermore, between July 1977 and December 1977 the ^{226}Ra concentrations of the waters collected in the site M-4 increased from (2.8±0.5) pCi$^{226}Ra/\ell$ up to (6.0 ± 1.0) pCi$^{226}Ra/\ell$, while before July 1977 the ^{226}Ra concentrations never reached 2.8 pCi$^{226}Ra/\ell$ level. This remarkable increase in the ^{226}Ra concentration in the waters collected in the site M-4 indicates that the uranium mining operations are releasing ^{226}Ra to the small stream which flows into the Rio das Antas after crossing the Campo do Cercado in the southwestern direction.

The ^{226}Ra concentrations in the waters collected in the site V-2 were not detected (i.e., < 0.20 pCi$^{226}Ra/\ell$) until September 1977, when started increasing steadily up to (1.3 ± 0.4) pCi$^{226}Ra/\ell$ in January 1978. The collection site V-2 is located in an indirect affluent of the Rio Verde, and is 2.5 km distant, in the northeastern direction, from the center of Campo do Cercado. The site V-2 is near an area where preliminary work for future mining operations started around August 1977.

"Dosimetry"

A calculational dosimetric model is in the process of being developed to estimate the annual internal dose equivalent from ^{226}Ra intake via the pathways of drinking water and irrigation patterns, as the basis to calculate the collective dose equivalent commitment to the population of the Poços de Caldas plateau and surroundings. The dosimetry is essentially based upon exponential models recommended by the International Commission on Radiological Protection, ICRP (IC59), and used successfully in the computer program HERMES (F171) and in the report WASH-1258 (So73).

The ICRP hypotheses and recommended values (IC59)(IC74) were used for calculating the annual dose equivalent for the whole body and selected organs of an adult individual. Accordingly, the usage factor for water ingestion is 438ℓ/year (IC74). The human intake of the main food products of the region are the following: coffee - 23 kg/year; potato - 90 kg/year; corn, tomato, bean, and rice - 115 kg/year each (IC74). Further details of the dosimetric model and of the irrigation characteristics of the Poços de Caldas region will be given elsewhere.

Figure 3 shows a series of graphs of the annual dose equivalent, in mrem*/year, from ^{226}Ra via the pathways of drinking water and irrigation patterns, as a function of the ^{226}Ra concentration in the water. The upper line of each graphic bar in Figure 3 indicates an annual dose equivalent calculated under the assumption that ^{222}Rn is totaly retained in the organ, while the lower line of each bar indicates the contrary assumption, that is total ^{222}Rn escape from the organ. Experimental data

* 1 mrem = 10^{-3} rem = 10^{-5} sievert (= 10^{-5} l/kg = 10^{-1} ergs/g)

on ^{222}Rn escape from human organs are not generally available in the open literature, with the exception of the 0.6 fraction of ^{222}Rn that escapes from bone (Ro58).

Observing the graphs of Figure 3, one can say that when the ^{226}Ra concentration in the drinking water is 3 pCi$^{226}Ra/\ell$, the annual dose equivalent to the whole body is approximately 10 mrem/year, while to the bone the annual dose equivalent is about 15 mrem/year. However, if the ^{226}Ra concentration in the water to be used to irrigate plantations of coffee, potato, corn, tomato,bean, and rice is 30 pCi$^{226}Ra/\ell$, the annual dose equivalent to an adult individual, consuming these food products at the same rate as the reference man (IC74), can reach about 700 mrem/year to the whole body and over 1 rem/year to the bone. Although the maximum permissible concentration for ^{226}Ra in drinking water is generally lower than that for waters with potential to be used for irrigation purposes, the use of water with the same concentration may result in lower dose when used for drinking than when used for irrigation of food plantations. This somewhat striking observation should not take anyone by surprise, since in 1967, Eisenbud based on studies made in several parts of the world had already noticed that "the principal daily intake of radium is from food rather than from water" (Ei67).

"Concluding Remarks"

Evidently, further studies are needed to support any conclusion that one may decide to draw at this stage of an assessment like this, which constitutes only a preliminary step towards a long term project. However, several tentative conclusions and general observations deserve to be registered here for future references.

(i) The ^{226}Ra concentration levels in the river waters of the Poços de Caldas region are in general under 1.0 pCi$^{226}Ra/\ell$, with the exception of the waters of few small streams crossing the area where the uranium deposits are now being explored, even though the ^{226}Ra concentrations in these waters are still lower than 30 pCi$^{226}Ra/\ell$.

(ii) The ^{226}Ra concentrations in river waters of the Poços de Caldas plateau may be higher in the wet season than in the dry season, but the differences are slight, so more refined studies are to be untertaken to allow a definitive conclusion concerning this matter.

(iii) A model to calculate the collective dose equivalent to the population of the Poços de Caldas region from the ^{226}Ra existing naturally in solution in the river waters will allow to estimate the dose enhancement from any increase in the ^{226}Ra concentrations in the usable river waters. Appropriate data are now being gathered to calculate, according the ICRP recommendations, the collective dose equivalent from ^{226}Ra to the population of the Poços de Caldas region.

(iv) If ^{226}Ra, from the future tailings of the uranium mining and milling operations in the Poços de Caldas plateau, would be released to the environment in amounts such that the ^{226}Ra concentrations in the waters used to irrigate the typical food plantations of the region would reach 30 pCi$^{226}Ra/\ell$, one could expect an annual dose equivalent as high

as 700 mrem/year to the whole body (and over 1 rem/year to the bone) of an adult individual eating those foods at the same rate as the reference man.

(v) The highest annual dose equivalent via the pathway of drinking water under the present conditions of the Poços de Caldas region would be around 95 mrem/year to the whole body (150 mrem/year to the bone) of an adult individual who would drink daily water from the Fonte do Villela, with ^{226}Ra concentration just under 30 pCi $^{226}Ra/\ell$. Notwithstanding, the Fonte do Villela constitutes a touristic attraction and its water, according to one anonymous tourist, is good not only for those who are thirsty, but also is "good for health"!

"Acknowledgements"

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Table 1. Ra-226 concentrations in waters of the Poços de Caldas region.

Collection site*	Jan 77 wet season	Jul 77 dry season	Jul 78 wet season
		pCi ^{226}Ra/ℓ	
A-1	1.8 ± 0.7 †	1.2 ± 0.7	1.6 ± 0.7
A-2	0.6 ± 0.2	ND ††	0.3 ± 0.2
A-3	0.3 ± 0.2	ND	0.3 ± 0.2
A-4	0.4 ± 0.2	ND	0.2 ± 0.2
A-5	0.4 ± 0.2	ND	0.3 ± 0.2
A-6	0.3 ± 0.2	ND	0.3 ± 0.2
A'-6**	0.6 ± 0.3	ND	0.3 ± 0.2
A-7	0.5 ± 0.2	ND	0.4 ± 0.2
A-8	3.9 ± 0.7	ND	ND
A-9	1.0 ± 0.3	1.4 ± 0.4	1.7 ± 0.4
A-10	ND ††	ND	ND
L-1	ND	ND	ND
L-2	ND	ND	ND
R-1	0.6 ± 0.3	ND	ND
R-2	ND	ND	ND
R-3	ND	0.8 ± 0.3	ND
M-1***	26.0 ± 6.0	-	-
M-2	1.3 ± 0.4	1.1 ± 0.4	1.2 ± 0.4
M-3	5.0 ± 1.0	4.1 ± 0.7	6.0 ± 1.0
M-4	2.6 ± 0.5	2.8 ± 0.5	5.0 ± 1.0
V-1	ND	ND	ND
V-2	ND	ND	1.3 ± 0.5
V-3	ND	ND	ND
V-4	ND	ND	ND
P-1	ND	ND	ND
P-2	29.4 ± 6.0	28.0 ± 6.0	26.0 ± 6.0
P-3	ND	0.6 ± 0.3	ND
F-1	ND	ND	ND

* Geographical locations of collection sites are shown in Figure 1.

** A'-6 is a well twenty meters apart from collection site A-6 in Antas river .

*** M-1 was a water source annihilated by the mining excavations.

† 1σ due to statistical counting and calibration uncertainties.

†† ND = not detected, i.e. < 0.20 pCi ^{226}Ra/ℓ.

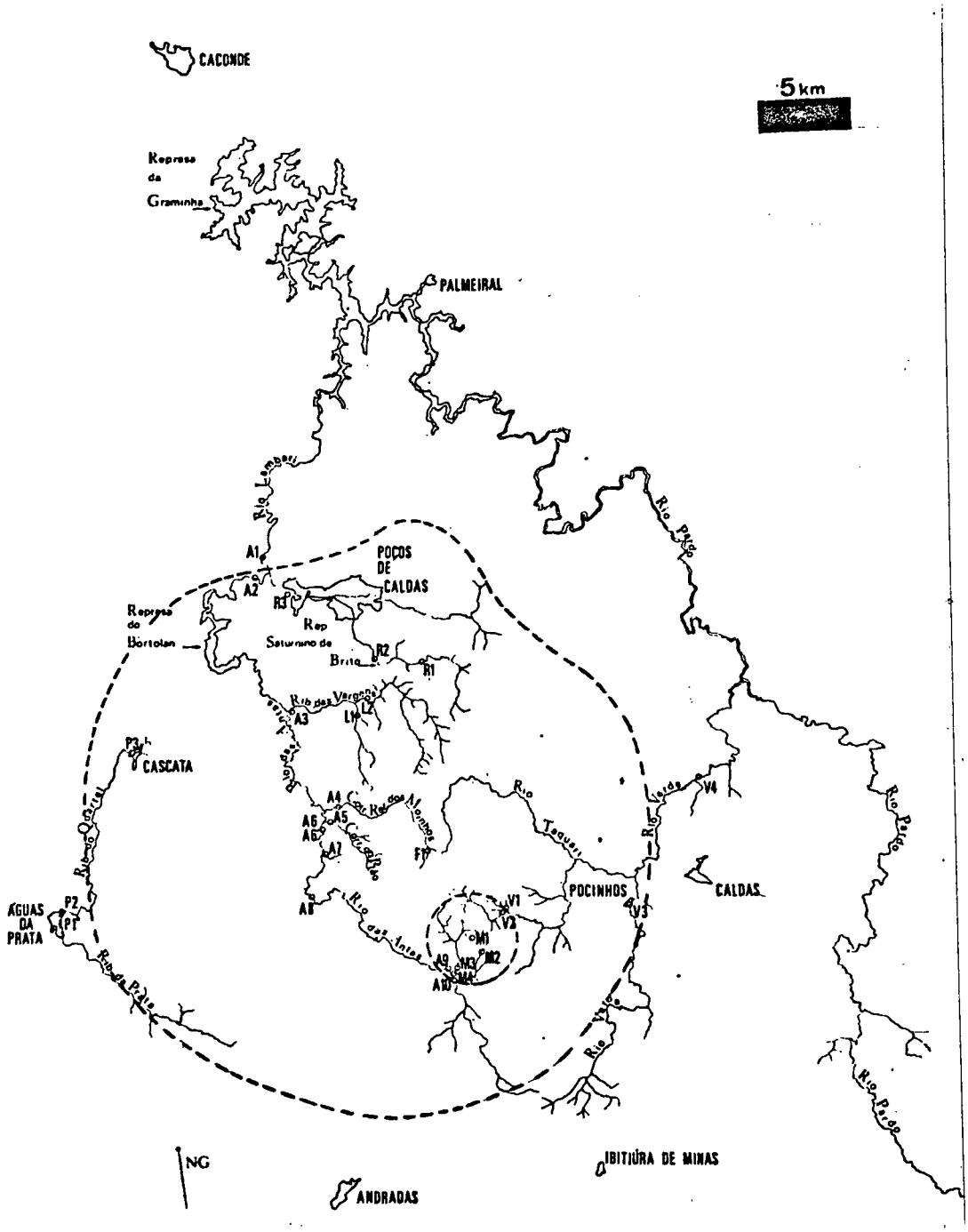

Figure 1. Sample sites for water collection in the Poços de Caldas region, and the main rivers of the hydrographic basins. Map is based upon Instituto Brasileiro de Geografia e Estatística (IBGE) charts.

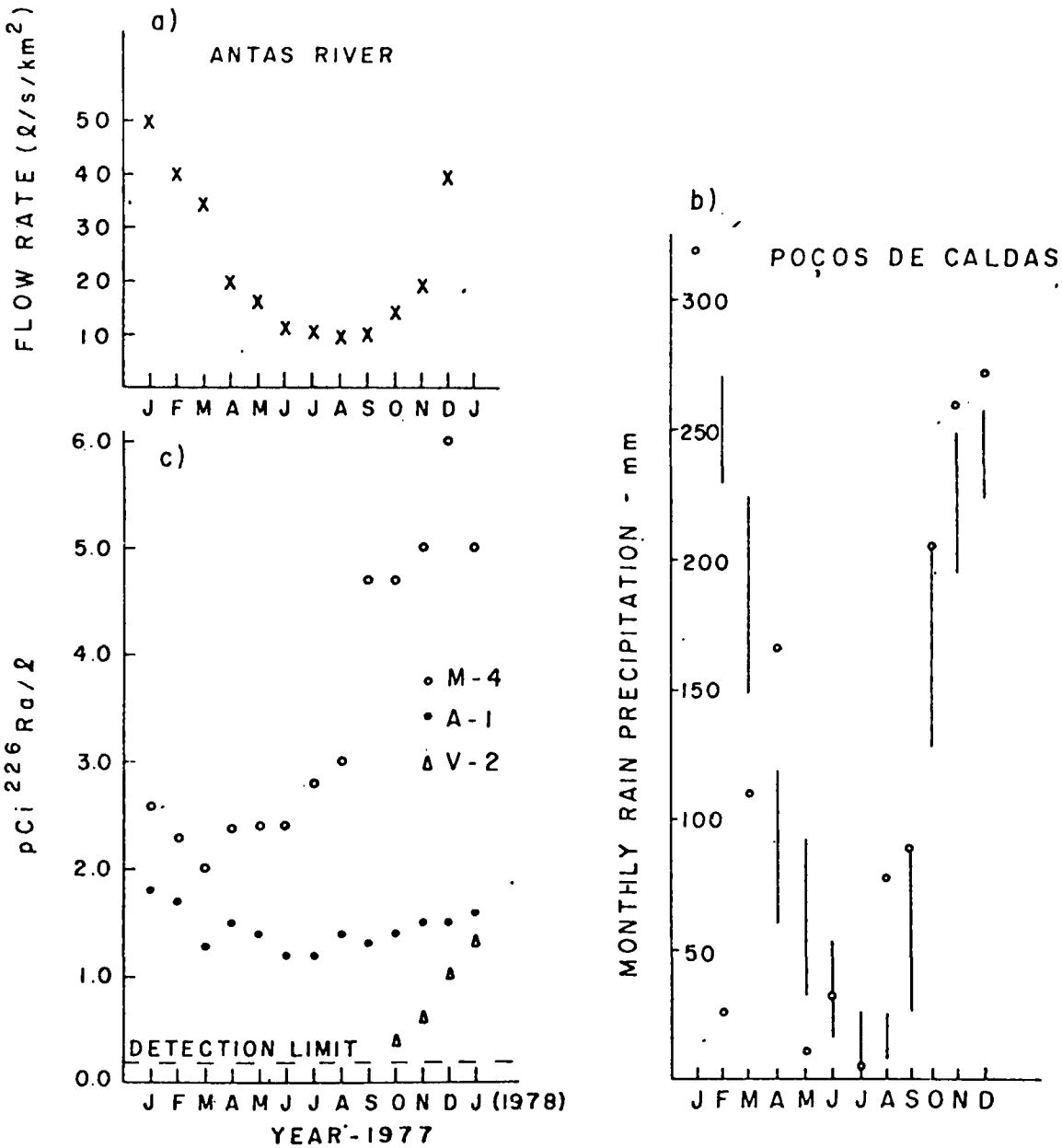

Figure 2. a) Flow rate of Rio das Antas; b) data on rain precipitation in Poços de Caldas; and c) Ra-226 concentrations in water samples collected in the mining area (M-4), Rio das Antas (A-1), and Rio Verde (V-2).

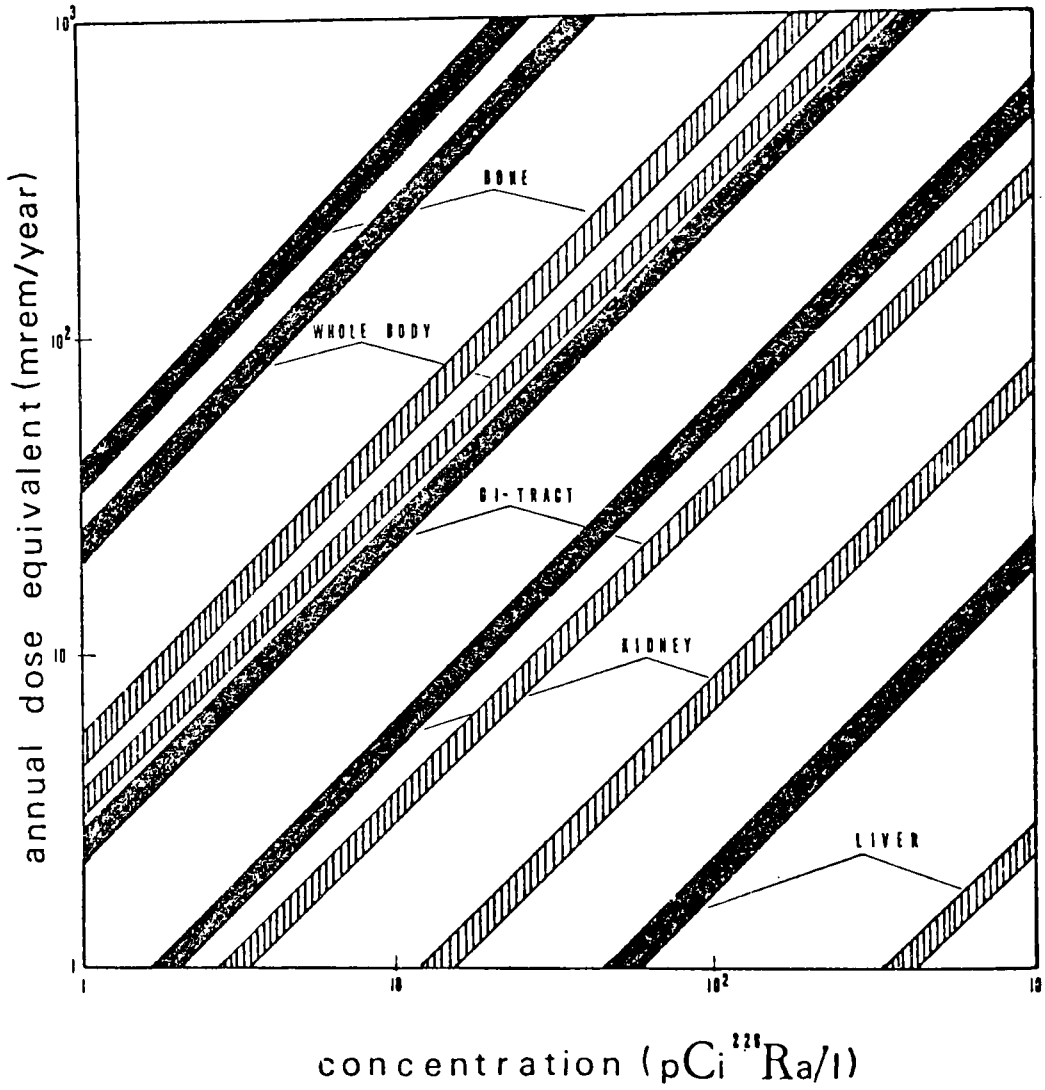

Figure 3. Annual dose equivalent from ^{226}Ra via the pathways of drinking water (bars with hachures) and irrigation patterns (dark bars). Upper lines of the bars indicate annual dose equivalent with no ^{222}Rn escape ,. while lower lines indicate total ^{222}Rn escape.

EVALUATION OF THE ENVIRONMENTAL DOSE COMMITMENT DUE TO RADIUM-CONTAMINATED SOIL

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Abstract

The Middlesex Sampling Plant located in Middlesex, NJ was a uranium ore sampling plant operating during the 1940s and 1950s. A radiological problem was identified during a routine program to resurvey selected former MED/AEC sites which are no longer under government control. The survey, when conducted by the U. S. Department of Energy (DOE), indicated that the Middlesex facility had a radium and radon problem on-site as well as off-site, where some of the contaminated soil was used as landfill. The old sampling plant is presently being used as a Marine Corps Reserve Training Center. Subsequent, more detailed studies have identified possible solutions to the contamination problem. The U. S. Environmental Protection Agency (EPA) is examining cleanup options based on a cost/benefit analysis utilizing the environmental dose commitment concept rather than an annual dose calculation. The practice of using dose to local populations as a basis for impact assessment can lead to a large underestimate of the total potential impact from the continuous environmental release of radon.

Introduction

Activities performed in connection with the Manhattan Engineer District (MED) efforts to develop the first atomic bombs and with postwar research sponsored by the U. S. Atomic Energy Commission (AEC) to develop nuclear energy led to the contamination of numerous sites in this country. The U. S. Department of Energy (DOE) has conducted a program to determine the present radiological status of those sites formerly used by MED/AEC and their contractors. Based on radiological monitoring most of the sites have been found to present no radiological hazard to present occupants. In such cases, the sites are approved for unrestricted use. At some of the sites, however, measurements of radioactive contamination levels have indicated a need for some form of remedial action before the site can be released for unrestricted use.

One of the locations surveyed, the Middlesex Sampling Plant in Middlesex, NJ, was used during its MED/AEC period for sampling, weighing, assaying and storing of uranium and thorium ores (FB78a). One of the major ores stored there was Belgian Congo uranium ore, which had a concentration of about 60% uranium. In equilibrium in the ore were radium and its decay products (FB78b). These elements formed the major constituents of the radioactive contamination found at the Sampling Plant site. The levels measured both on-site and off-site were found to exceed background measurements nearby by factors of up to 2000 (FB78b).

As a result of these findings the DOE has presented several possible remedial actions for the on-site and the off-site contaminated areas. The DOE presentation was based on levels of radioactive contaminants as measured on-

site. A measurement of gamma radiation levels or of alpha contamination would be a realistic indication of a potential radiological hazard attributable to fixed contamination.

Such a study of concentrations may tend to misrepresent the off-site hazard when one or more of the radioactive constituents is a gaseous product, capable of diffusing during its lifetime and moving off-site. With the diffusion of radon gas, the maximally exposed individual is not a valid reflection of maximum population dose. There is a greater distribution of the gas over a large area, leading to a severe underestimate of the population dose. Consideration should be given to evaluation of the total health impact of any suggested actions. Each option will involve exposure of some segment of the general population to radioactive materials. These will take the form of gases (radon emitted from the tailings) or particulates (dust laden with loosened radioactive soil from earth moving operations).

The short-term effects of operations have been addressed in the Engineering Evaluation and the Environmental Analysis, as have the financial costs. However, long-term effects of exposure to gases and particulates which have not been contained either prior to or during cleanup should be considered. Further, materials remaining after cleanup operations have been completed will lead to a very low level, but long term, dose to the general population. There should be a comparison of cost of the cleanup options and the anticipated health effects from materials remaining. This would give a basis for appraisal of each remedial action alternative.

Proposed Options

The Middlesex Sampling Plant and locations nearby affected by its operations require cleanup efforts of some type. In all cases, remedial actions would involve removal of buildings on site, excavation of soils and emplaced utilities such as the contaminated drainage system, asphalt coverings, lawns, and sidewalks. Follow up actions would include restoration of land surfaces to their original grade. The remedial actions being examined by DOE are based on levels of contamination which may be allowed to remain (FB78b). They include the following:

1. Interim storage on site : Areas of especially high contamination would be removed, packaged and stored on-site for the period ending with transfer to a permanent storage area. This would be used in addition to alternative actions and is designed to limit exposures to the public by packaging of contaminated portions of buildings and soils immediately. Additional clean up could then be implemented at a later time.
2. Long-term storage on-site : Packaged contaminated materials and soils from both on-site and off-site locations would be stored on the Sampling Plant site indefinitely. Such a program would require that the site not be open to public access for a prolonged period. Applicable criteria for such a long-term storage program would also need to be considered.
3. Removal of materials in interim storage and direct removal of site soils and buildings to a disposal site : This would follow implementation of alternative 1.
4. Direct removal of all contamination to a disposal site : Such a

program would require more time to implement, since arrangements for final disposition would need to be made before removal of contaminated materials could be initiated.

The DOE has presented these options in an engineering report. Evaluations of the options have been presented in a second report which presents an environmental analysis of the effects of all options. The effects discussed are examined in light of gamma radiation levels and radon concentrations now existing at each location. According to the DOE report potential health effects from radon daughter inhalation by occupants of the site buildings would result in one radiation associated lung cancer to building occupants every 70 years. External gamma exposures would yield the equivalent of one cancer incidence every 6,000 years. Projections of health effects have been made based on exposures to maximally exposed individuals off-site as well as to regular occupants of on-site facilities.

Environmental Dose Commitment

The assessments suggested here for deriving health effects projections depart in two significant respects from practice common in the past for assessing the significance of radiation exposures. The first of these is the use of the concept of environmental dose commitments (EDC). This concept considers the totality of doses to all populations over the lifetime of the radionuclide in the biosphere and not just to a maximally exposed individual located near a facility (US73).

The second departure from past practice is to evaluate potential health effects rather than to minimize radiation dose as the end point. In retrospect it is perhaps obvious that the focus for determination of the appropriate level for any guidance should be its public health impact, but in the past minimization of dose has served as a useful surrogate for the health impact because of uncertainties about the magnitude and shape of the relationship between dose and effect.

Under the concept of environmental dose commitment the health impact analysis thus considers the total impact of releases of radioactive materials to the environment by including radiation doses committed to local, regional, national, and worldwide populations, as well as doses committed due to the long-term persistence of some of these materials in the environment following their release. The analysis would serve to identify the effluents from the site (gaseous as well as wind-borne solids and soluble or insoluble contaminants carried off by water erosion) which represent the major components of risk to the population. This would lead to a better-defined view of the need to control long-lived contaminants as well as a recognition of the excess control measures for any short-lived radioactive materials.

To make a determination of the degree of control which can reasonably be required by any applicable standards, an analysis of cost-effectiveness of risk reduction must be performed. This will be discussed below.

For the Middlesex Sampling Plant Site calculations of the EDC may be performed with very little additional data. Population estimates for the area for the period of interest exist now. This would probably be 100 years, the period recommended by the EPA for institutional control of a waste repository.

The dose rates have been measured on-site. Off-site estimates of radon may be made knowing the source term and using existing wind rose data. From this information, a dose per year from each radionuclide of interest (basically radium and radon isotopes) may be found. Determining these values for the period of interest will yield the EDC. Based on the EDC in person-rem, health effects due to each deposited radionuclide may then be considered.

The DOE report has addressed radiological impacts in terms of potential health effects to occupants of the Sampling Plant buildings as well as to nearest off-site residents for radioactive contamination levels existing now. Consideration has also been given to contamination problems due to vegetation uptake factors for foods grown in contaminated soil off-site (FB78b). Environmental impacts due to remedial action implementation have been examined. As a short-term problem many of these impacts, such as releases of radioactive airborne particulates, would have their greatest potential effects on workers involved in the cleanup operations, rather than on the general population. In addition to air quality effects there would be radiological effects on local stream water quality. These effects have been discussed to some extent in the engineering report (FB78a) and should be considered in the decision-making process, although contribution their may be minimal.

Cost-Effectiveness Studies

The basic thrust of the health effects calculations described above would be to establish the need for and extent of remedial actions. The Engineering Evaluation presents three cleanup level options which should also be examined in this manner. Each of the alternative remedial action proposals mentioned above has been addressed with three cleanup levels for radium contamination of the soil: 10 pCi/g above background, 5 pCi/g above background, and background. Costs of these cleanup options have been listed in each case for each location (FB78a). It is at this point that a health effects analysis would prove most helpful in evaluating the cleanup options from a cost-benefit standpoint. By applying an EDC calculation to the cleanup levels presented, a risk assessment could be made which would allow decision making from a cost-effectiveness approach.

A total population health effects determination would be made for each of the suggested cleanup levels, applied to the 100-year period mentioned above. Such a determination would then permit a systematic evaluation of the cleanup level options in light of cost of implementation vs. reduction of health effects.

In addition to a study of health effects at each of the suggested cleanup levels, a second determination of health effects due to the storage options should also be made. The Sampling Plant site has been proposed for both interim storage (alternative 1) and long-term storage (alternative 2). While either of these options would be able to reduce dose to the public by removal of sources from public access to containment, there would still be some dose involved, first in the physical cleanup process and later at a much reduced level from the storage area. These options should similarly be examined to view the relative speed with which they could be implemented, thereby considering total doses involved.

Recommendation

A health effects analysis should be utilized to assess the suggested remedial actions for cleanup of the radiological contamination in and around the Middlesex Sampling Plant site. Such an assessment would include an appraisal of cost of each option compared to reduction of EDC. This would, in turn, give a risk reduction for all population exposed to the contamination from the site rather than a measure of dose reduction to the nearest off-site location. The use of the EDC will allow a decision-maker to proceed with greater knowledge of long-term risk and the cost associated with all options.

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ASSESSMENT OF RADON PROGENY INHALATION EXPOSURE FROM LOW-LEVEL WASTES OF PHOSPHATE MINING IN FLORIDA*

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Abstract

The redistribution of naturally-occurring uranium series radionuclides as a result of phosphate mining, processing, product use, and waste disposal presents several potential radiation pathways to man. Of particular importance is exposure to radon-222 progeny in structures built on reclaimed lands in Florida.

We analyzed indoor radon daughter sampling data from Polk County, and categorized the data by land and structure type. We determined the average population-weighted concentration in about 4,400 homes to be about 0.009 working level (WL) in addition to a background of 0.003 WL. We also determined that the average annual cumulative indoor exposure on reclaimed land was approximately 0.02 working level months (WLM). A relatively small number of houses on high-activity overburden accounted for 38% of the total population exposure.

We are proposing a generally applicable model to relate lung cancer risk to the average annual exposure, the risk coefficient, the expected lung cancer mortality from all other causes, the duration of the exposure and the number of years for observation of the effects. Health risk estimates were performed for present levels and population size, and also for several scenarios anticipating new growth and construction -- with and without imposed standards to limit indoor radon progeny levels. The model suggests that for an equilibrium condition, about one additional case of radiogenic lung cancer every two years in the Polk County population might be expected.

Introduction

Greater than average terrestrial concentrations of uranium and radium are associated with the phosphate rock matrix in Florida. The natural soil radium content of undisturbed (unmined) land increases with depth from the surface into the phosphate ore layer (Ro78). When the phosphate rock is mined, the overburden is removed by draglines and laid to one side. The matrix is then removed and slurried with water to a washer/beneficiation plant. Sand

* Research performed at the University of Florida/Gainesville, under contract with the Florida Phosphate Council.

tailings (waste by-products) are pumped back to previously mined areas or other disposal sites for use in land reclamation.

Prior to the development of the flotation process during the 1940's, the fine (<1 mm dia.) fraction of phosphate mixed with sand was not recovered with the pebble fraction, and was instead returned to the land as "debris." Later, during land restoration operations, the debris piles were redistributed as new land surfaces or fill.

Some unmined lands in the region with near-surface phosphate deposits (little, if any, overburden) and elevated radium concentrations have been identified. These have been termed by some as "mineralized" land (HRS78). In addition, unmined lands with enhanced natural radioactivity due to fill or other cover materials exist, and are referred to as "radioactive fill" lands. Often the distinction between "fill" and "mineralized" lands is less than adequate.

Thus, the redistribution of naturally-occurring uranium series radionuclides following phosphate mining, processing, product use, waste disposal, and land reclamation has increased the concentrations of near-surface radioactivity. Although several possible pathways to man may result from the technologically-enhanced radiation, the evaluation of radon progeny exposures in structures on these lands (primarily in Polk County) is particularly important.

The possibility of increased radiation-induced lung cancer from radon daughter inhalation prompted the Environmental Protection Agency (EPA) to consider the imposition of radiation standards specific to Florida homes and phosphate-related lands, and to designate overburden, slimes, and tailings from surface phosphate mining as "hazardous wastes" by reason of their radioactivity content (FR78).

The objectives of this study were 1) to assess the distribution of population exposures to indoor radon daughter concentrations, and 2) to estimate the lung cancer risks to residents of Polk County, whose homes are built on either reclaimed phosphate mine lands or unmined parcels with elevated soil radium levels.

Methods

Population exposures. We analyzed the indoor radon progeny data that were available for Polk County. Measurements were performed by the EPA (EPA75), the State of Florida Department of Health and Rehabilitative Services (HRS; HRS78), and the University of Florida College of Engineering (UF;UF78). We combined the UF quarterly sampling data for 25 homes with the additional data from a larger sampling of structures by the HRS to identify radiological characteristics according to land and structure type. The data for mobile homes resulted primarily from HRS measurements. Only a small amount of real data was available from the UF concerning the "fill" land category and from the HRS concerning "mineralized" lands. Since the radon progeny concentrations reported for houses on these two land categories were similar, they were combined into a single exposure analysis category.

We then employed population statistics to estimate the distribution of population exposures in Polk County. Less than 5% of the residences in Polk County are situated on reclaimed land (HRS78). From the sampling data we estimated that an additional 1% of all residential acreage (unmined) in the County could be classified as "mineralized" near-surface deposits or fill materials with elevated natural radioactivity.

Annual cumulative population exposures were estimated for each category from multiplication of the mean indoor radon progeny concentration (WL, above background) by an occupancy factor and a breathing rate correction factor (for continuous indoor exposure)*.

Risk analysis. The BEIR Report (NAS72) suggested that the mechanism of radiogenic lung cancer is dependent upon metaplastic perturbations (lesions, inflammations, etc.) in the bronchial epithelial tissues by nonradioactive irritants. Studies on underground miners tend to support the hypothesis that a synergistic relationship exists between exposure to inhaled radon daughter activity and other carcinogens or lung irritants (Ar76, He76), and that the combined effect is multiplicative rather than additive (Do77). It is therefore reasonable to presume that a given amount of radiation exposure to the lungs will be more harmful to a subject whose expectation of cancer is already high, than to one whose expectation is lower, i.e., that a given amount of lung exposure will not produce equivalent effects in all people.

We have proposed the following risk model to assist in the assessment of biological effects (lung cancer) from the inhalation of radon daughters. In simplest terms, the risk model states that the additional cases of lung cancer will be proportional to the product of the natural incidence and the cumulative exposure. The constant of proportionality is the risk coefficient. Lifetime risk per unit radiation (or cumulative exposure) was proposed as the best statistic for use in predicting lung cancers among populations exposed to radon daughters in air (Ar78). The present approach involves consideration of several time factors, including the latent period for lung cancer induction, the duration of the exposure, and the follow-up (observation) period. A linear, zero intercept dose response curve was assumed (Co78).

The expected number of lung cancer cases during a specified period of time among an unexposed population group may be represented by the symbol N_z. The absolute risk (N_{add}) is the additional lung cancer mortality due to the inhalation of radon decay products (above and beyond normal background levels) (Ja73a). The total number of lung cancer observed (N_{tot}) in a population exposed to the radiation hazard is therefore

$$N_{tot} = N_z + N_{add}. \qquad (a)$$

The percent increase in risk relative to the natural risk is

$$I_r = 100\ N_{add}/N_z. \qquad (b)$$

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* 36 WLM/WL-yr. To estimate cumulative WLM, the assumption was made that a third of one's total weekly inhalation is breathed occupationally. Thus, $3 \cdot 12 = 36$ "working-month equivalents" are breathed each year. We also assumed an average occupancy factor of 0.7.

Specifically, the expected number of "naturally occurring" cases of lung cancer (N_z) is

$$N_z = P_o \; Z \; t_2 \; (10^{-6}) \qquad (c)$$

where P_o is the population size, Z is the age-adjusted lung cancer mortality rate (cases/10^6 person-yr), and t_2 is the time frame or observation period (yr).

The additional risk is a function of the exposure level, the risk coefficient and the lung cancer mortality rate from all other causes. In a previous report, Jacobi (Ja73a) expressed this relationship as

$$N'add(E) = \alpha_{rel} \cdot E \cdot N'erw(E), \qquad (d)$$

where N'add(E) referred to the additional lung cancer mortality as a result of a cumulative working level exposure (E), α_{rel} was the relative risk coefficient or inverse doubling dose, and N'erw(E) represented the expected number of lung cancer deaths. Jacobi's equation can be restated more explicitly using new symbols as

$$N_{add} = K \; W_a \; t_1 \; N_z \qquad (e)$$

$$= K \; W_a \; t_1 \; P_o \; Z \; t_2 \; (10^{-6}), \qquad (f)$$

where K is the risk coefficient constant (WLM^{-1}), W_a represents the average annual exposure (WLM/yr) above background, and t_1 is the duration of the exposure (yr). W_a is the product of the average annual indoor radon progeny concentration, the breathing rate correction factor, and the occupancy factor. An appropriate expected lifetime should normally be used for the value of t_2 the observation period. The product of $W_a \; t_1$ is the equivalent of the cumulative working level month (CWLM) exposure unit frequently encountered in some of the literature.

The risk model must be modified to account for a changing population size if the analysis is performed over a lengthy period of time (Fi78). If P_o is the original population size and i is the annual rate of change, then the number of expected (natural) lung cancer mortalities in an expanding (or decreasing) population for t_2 years is thus

$$N_z \simeq P_o \; Z(10^{-6}) \sum_{n=0}^{t_2-1} (1 + i)^n, \qquad (g)$$

and the anticipated number of additional lung cancers due to the radiation component is

$$N_{add} \simeq K \; W_a \; Z \; P_o(10^{-6}) \left\{ t_1 t_2 + \sum_{n=1}^{t-1} [1 + i)^n - (1 + i)^{n-1}] \right.$$

$$\left. (t_1 - n) \; (t_2 - n) \right\}. \qquad (h)$$

The total number of cancer mortalities from all causes combined (N_{tot}) is the grand sum of equations (g) plus (h). We assumed that the annual rate of change, whether positive or negative, remains constant for t_2 number of years, and also that the values N_z and N_{add} are much less than the value P_o (Fi78).

The primary source for dose response data for human lung cancer risk following inhalation exposure to radon and radon daughters is the epidemiology of underground miners. A review (Fi78) of absolute risk factors for underground miners of various locations around the world resulted in the value 5 ± 4 (cases/10^6 person-yr-WLM) as the best estimate of the risk. Given this risk per unit exposure and the natural incidence of lung cancer in a population, the risk coefficient can be determined. The lung cancer incidence rate (Z) for all males in Florida is 449 cases/10^6 person-yr (HEW74). Assuming that Florida males have essentially the same "natural" risk as the base populations of the underground miners,* the risk coefficient is

$$K = N_{add}\ (WLM^{-1})/N_z \qquad (i)$$

$$= \frac{5\ (\text{cases}/10^6\ \text{person-yr-WLM})}{449\ (\text{cases}/10^6\ \text{person-yr})} = 0.011\ WLM^{-1}.$$

We therefore estimated the doubling dose (reciprocal of K) to be about 90 WLM, which compares with other reported values of 100 WLM (Au76), 110 WLM (Ja73b), and a value of 60 WLM used previously by the EPA (Mi77).

The natural age-adjusted incidence (Z) by county and state in the U.S. is available. The annual lung cancer incidence in Florida for members of the general population (males and females) is about 244 cases/10^6 person-yr (HEW74). The individual natural risk is strongly a function of age, sex, and smoking.

To tentatively delineate the impact and significance of radiation-induced lung cancer in Polk County, four population exposure scenarios were developed. For each of the scenarios a constant age distribution with time was assumed. We accounted for an assumed 10-yr lung cancer latent period (Au76) by setting an upper limit on the "effective" exposure period (t_1) of t_2 - 10 years. Furthermore, we assumed the risk coefficient K to apply equally well to female adults and children in the general population. Considering the many assumptions required and the limitations on the available data for radiation carcinogenesis, the reader's attention should be directed towards relative differences in calculated risk rather than on absolute numbers generated.

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* Actually the natural or expected incidence of lung cancer among the male population from which the fluorspar miners, and the American uranium and metal miners were taken is somewhat lower: 416 cases/10^6 person-yr (derived from NAS72). However, since a disproportionate number of miners were smokers, their expected incidence rate could have been higher than this. Therefore, the Florida male cancer incidence was chosen for the present analysis of a Florida population. The incidence for all males in the U.S. is 380 cases/10^6 person-yr (HEW74).

Results

We found the average indoor radon progeny "background" concentration in Florida homes to be about 0.003 WL (Fi78). Indoor levels were confirmed to be significantly higher in structures on reclaimed land. A summary of indoor radon progeny levels in Polk County homes by land and structure type is presented in Table 1. Since the radiological data were found to be log-normally distributed, we reported the geometric mean rather than the arithmetic average.

Table 1. Classifications of Polk County Structures and Their Indoor Radon Progeny Levels; A Composite of UF and HRS Data

Exposure Category		Radon Progeny Concentration, (WL)	
Structure Type	Land Type	Mean*	(Range)
Slab-on-grade	Undisturbed	0.003	(0.001-0.010)
	Higher-activity overburden and debris	0.043	(0.019-0.140)[a]
	Lower activity overburden	0.008	(0.004-0.018)[b]
	Tailings	0.008	(0.002-0.038)
	Unmined, near-surface radioactive deposits and fill	0.019	(0.003-0.045)[c]
Crawl Space Homes and Mobile Homes	Undisturbed	0.003	(0.001-0.010)
	Reclaimed	0.006	(0.001-0.014)[d]

*geometric mean

(a) combination of UF "debris" lands and the higher-activity population in the HRS "overburden" category.

(b) combination of the lower-activity population in the HRS "overburden" category, and the UF type "overburden".

(c) HRS "mineralized" and UF "radioactive fill" land types in this category are grouped together for convenience, and remain to be further defined.

(d) all reclaimed or otherwise altered land types.

Approximate population, structure type, and population exposure (to levels in excess of background) distributions are given in Table 2. We found the population-weighted radon progeny mean concentration in about 4,400 homes on disturbed lands to be about 0.009 WL above background. The corresponding "excess" population exposure was thus estimated to be about 117 WL-persons

above background. The average annual cumulative exposure (W_a) was found to be 0.009 WL • 36 WLM/WL-yr • 0.7 = 0.02 WLM for a typical individual resident

Table 2. Estimated Population Exposure from Elevated Radon Progeny Concentrations

Land Category	Estimated Residences	Estimated Persons	Excess Population Exposure WL-Persons	Excess Population Exposure %
A. RECLAIMED LAND				
1. Slab-on-grade: High-activity overburden and debris lands	322	1128	45.1	38
Low-activity overburden	843	2951	14.8	13
Tailings	995	3482	17.4	15
2. Crawl space and mobile homes: all reclaimed	1440	3603	10.8	9
Total:	3600	11164	88.1	75
B. RADIOACTIVE DEPOSITS, AND FILL				
1. Slab-on-grade	473	1656	26.5	23
2. Crawl space and mobile homes	316	790	2.4	2
Total:	789	2446	28.9	25
C. UNDISTURBED LANDS				
All structure types	74468	262390	---	---
Grand Total:	78857	276000	117.0	100

From Table 2 it can be seen that about 38% of the additional pupulation exposure appears to be attributable to a relatively small fraction of residences on the higher-activity overburden or debris lands where the indoor radon progeny concentrations were found to be distributed geometrically around a value of 0.04 WL above background. Since contributions to the total population exposure from the occupancy of public buildings and other non-residential structures were small, they were not included in the exposure analysis.

Having established probable population size and exposure distribution parameters, it was possible to estimate health risk implications. A summary of population risks to lung cancer according to equations (g) and (h) for four hypothetical 70-year scenarios is shown in Table 3. It can be seen that at current indoor radon progeny levels, population size, age distribution and number of dwellings, about 34 additional lung cancer deaths might be attributed to increased levels of indoor airborne radioactivity. If distributed evenly with time, the excess cancers might occur at the rate of one every two years. For Polk County as a whole, the 34 added cases over a 70-year period would represent an increase of 0.7% (too small to be detected statistically). However, the additional cases represent an increase of approximately 14.5% in expected lung cancer mortality among a base population of 13,610 persons at risk on reclaimed land. In particular, for residents of debris reclaimed land, 13 additional lung cancer deaths are estimated during the 70-year period according to the first scenario, which corresponds to a 67% increase over their natural risk.

The possible effect of restrictions on indoor radon progeny levels can be seen when scenarios three and for are compared. The result of decreased indoor activity might prevent about 12 cases, or about one very six years.

Table 3. Theoretical Population Lung Cancer Risk Scenarios for a 70-Year Exposure Period for Current and Restricted Indoor Working Level Concentrations[a]

Scenario	Level (avg.)	Population Growth rate/year	Lung Cancer Deaths N_z	N_{add}	N_{tot}
1	present (0.0086 WL)	0%	232	34	266
2	present (0.0086 WL)	1.5% first 30 years, and 0% next 40 years	332	47	379
3	present (0.0086 WL)	1%	334	42	376
4	restricted[b] (0.0064 WL average for existing structures and 0.0053 WL for new construction)	1%	334	30	364

(a) $K = 0.011$ WLM^{-1}, $W_a = 0.217$ WLM/WL-yr, $t_1 = 60$ yr, $t_2 = 70$ yr, $Z = 244$ cases/10^6 person-yr, and $P_o = 13{,}610$ persons.

(b) Limits of 0.02 WL for existing structures, and 0.01 WL for new structures.

Lifetime theoretical risks to individual residents ($P_o = 1$) were determined (Fi78); these varied from 0.00001 to 0.219 depending on the indoor level,

residency period, and smoking pattern. N_{tot} for the "average" individual (W_a = 0.02 WLM) was found to be 0.0175 (I_r = 2.4%) for 10-yr residency and 0.0195 (I_r = 14%) for lifetime residency on reclaimed land. For the average resident on debris land (W_a = 1.01 WLM), N_{tot} was found to be 0.019 (I_r = 11%) for 10-yr residency and 0.029 (I_r = 67%) for lifetime residency.

Discussion

In general, most indoor radon progeny levels are low, and the population health hazard is rather small. However, levels in some existing homes are unacceptably high, and these should be reduced. Debris category reclaimed lands may not be suitable as future home sites, unless proper construction methods are incorporated to limit indoor radon progeny levels.

The exercise of predicting health effects into the future is complicated by uncertainties in utilization of reclaimed lands and population trends. It is also difficult to predict future indoor radon progeny concentrations, since the technology is available to reduce current levels.

It is quite possible that the risk coefficient for underground miners, which we used in the risk analysis, is biased by other lung irritants characteristic of mining atmospheres. Therefore, better human risk data for non-miners is needed. For extension to the general public, the present risk coefficient likely produces a cautious overestimate rather than a nearest approximation of the biological effects from long-term low-level inhalation exposure to radon and radon daughters.

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RECOMMENDATIONS FOR REMEDIAL ACTION AND DECOMMISSIONING OF A RADIOACTIVE WASTE BURIAL SITE

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Abstract

For the past year, the Nuclear Fuel Services, Inc. (NFS) site located in West Valley, NY has been the subject of state and federal efforts to determine decontamination and decommissioning options. In 1978, the U. S. Environmental Protection Agency (EPA) issued Criteria for Radioactive Wastes for storage and disposal of all forms of radioactive wastes. Under an Atomic Energy Commission (AEC) license, NFS operated the only commercial fuel reprocessing facility in the United States. As a result of the reprocessing activities, the site contains liquid high-level radioactive waste, buried cladding hulls and defective fuel elements, a spent fuel storage pool, and a low-level burial ground. Low-level radioactive material contained therein also comes from sources other than NFS's operations. The site received a license from the State of New York to perform low-level burial operations and radioactive material was buried until 1975. Studies of the low-level burial area show radioactive gases have leaked through the trench caps and the caps are more permeable than the surrounding soil allowing water infiltration into the trenches. Active site maintenance is used to prevent trench water overflow through the trench caps. Other remedial actions have been described for the site and are undergoing implementation. The West Valley site will be examined to determine the extent of remedial action and decommissioning activities which may be necessary based on the proposed EPA environmental criteria for radioactive waste.

Introduction

The State of New York authorized the establishment of a commercially operated, low-level radioactive waste disposal area on part of a larger site containing a facility for the reprocessing of nuclear fuel. A site in Cattaraugus County approximately 30 miles southwest of Buffalo, New York known as West Valley was licensed as a disposal site and in November 1963 the first radioactive material was buried. Shallow trenches were dug to hold the waste and an earthen cap (cover) was placed over each trench after it was filled with the waste.

In the mid 1960s several burial trenches in the northern portion of the site (trenches 1-7) began to fill with water shortly after they had been covered. This posed a serious potential problem as the water could carry buried radionuclides out of the trenches and into the environment. To eliminate or reduce the water accumulation, burial procedures were changed for trenches in the southern portion (trenches 8-13) of the site. The new procedures were required by the State in 1968 and stipulated new trench capping methods were used to prevent surface water from entering the trenches.

By 1974 three of the trenches in the north area had developed high levels of water. Water levels in the south trenches, where modified capping procedures

had been used, remained low. To date, no significant water accumulation has occurred in the south trenches. In March 1975, water in one trench (trench 4) in the north area seeped through the trench cap. The contaminated surface runoff from this seepage was detected by the NYSDEC surveillance program and confirmed during an onsite inspection. A similar seepage was noted shortly thereafter along the west side of the cap on trench 5 (Ne75). Based on this occurrence, NFS closed the burial site and has not reopened it.

During the summer of 1978 remedial action was taken to prevent further water infiltration into the north burial trenches. The essence of this remedial action was to place additional earth over the cover of these trenches. Eight feet of dirt was placed over the trenches and compaction of the cover material was done using bulldozers. To date eight feet of dirt cover now exists over all burial trenches in the north and south portion of the site.

During the period from October 1963 through March 1975 more than 2,000,000 cubic feet of low-level radioactive waste were buried in the West Valley trenches. Kilocurie quantities of strontium-90, tritium, and cobalt-60 have been emplaced in the trenches along with such isotopes as radium-226, plutonium-238, plutonium-239, uranium-233, uranium-235, thorium-232, and americium-241 (Ke73)

Several reports and papers (Gi77a, Gi77b, En77, USDOE78) have been published detailing conditions at the low-level burial site at the West Valley site. These works identify environmental pathways by which radioactivity buried in the low-level trenches has been observed leaving the site. One such pathway involves the formation of gases, radioactive in nature, through chemical interactions in the burial trenches caused by reactions between water which has infiltrated into the trenches and the waste material, some of which is organic. Another pathway identified involves radioactively contaminated water percolating through burial trench caps and running off to adjacent streams. This pathway causes still another pathway involving the discharge of radioactive liquid from the trenches into a nearby stream. These releases occur in a controlled fashion after treatment has been performed on the radioactive material. Finally, lateral migration of small quantities of tritium through the strata adjacent to the trenches has also been observed, but to a very small extent.

In November 1978 EPA published proposed Criteria for Radioactive Wastes in the Federal Register (Co78). These criteria (a) define radioactive wastes, indicate which types of wastes should be controlled, and give examples of where these wastes originate; (b) state the goal of radioactive waste control and define limitations on institutional and other controls over certain time periods; (c) discuss the factors to be considered in assessing risk to the general public and the general environment; (d) discuss the factors which would result in unacceptable risk for different methods of disposal; (e) require that the selection, design, and operation of a disposal site must enhance isolation of radioactive wastes; and (f) discuss the appropriateness of retrievability of waste and communication of waste disposal locations to future generations.

Analysis

By reviewing the conditions at the site and comparing the status of the site to the proposed EPA criteria, determinations can be made as to potential

remedial actions and decommissioning actions needed to assure that the low-level site poses no unnecessary risk to public health and safety and to the environment and that all low-level radioactive materials are ultimately disposed of in an environmentally acceptable manner.

Proposed Criterion No. 1

Radioactive materials should be considered radioactive wastes requiring control for environmental and public health protection if they have no designated product or resource value and (a) are human-produced by nuclear fission or activation, fabricated from naturally radioactive materials into discrete sources, or as a result of regulatory activities are prohibited from uncontrolled discharge to the environment; or (b) contain diffuse naturally-occurring radioactive materials that, if disposed into the biosphere, would increase exposure to humans above that which would occur normally in pathways due to the preexisting natural state of the area. Examples of radioactive waste materials that should be subject to environmental protection requirements are:

All radioactive materials associated with the operation and decommissioning of nuclear reactors for commercial, military, research, or other purposes and the supporting fuel cycles, including spent fuel if discarded, fuel reprocessing wastes, and radionuclides removed from process streams or effluents.

Artifically produced radioisotopes, including discrete radium sources,for medical, industrial, and research use and waste materials contaminated with them.

The naturally-radioactive residues of mining, milling, and processing of uranium and phosphate ores.

This criterion addresses the issue of which materials should be considered as radioactive waste.

In general radioactive material buried at the low-level site would be considered radioactive waste based on this criterion.

Proposed Criterion No. 2

The fundamental goal for controlling any type of radioactive waste should be complete isolation over its hazardous lifetime. Controls which are based on institutional functions should not be relied upon for longer than 100 years to provide such isolation; radioactive wastes with a hazardous lifetime longer than 100 years should be controlled by as many engineered and natural barriers as are necessary.

This criterion addresses the issue of control of radioactive waste.

At the present time remedial action appears necessary to assure that this criterion is attained. Complete isolation of the radioactivity contained in the low-level burial area had not been accomplished before the summer of 1978. Whether the remedial action taken during 1978 will be sufficient to meet this criterion is as of yet unclear. Further actions such as using liners for the trenches may or may not be necessary to isolate the waste for long periods of time. However, it has not been positively demonstrated that liners would achieve successful isolation either.

Based on an inventory of material buried at the site (Ke73) one can calculate the approximate hazardous lifetime of the radioactivity buried at the site. If it is assumed that Ra-226, Am-241, and C-14 are the three isotopes which will remain hazardous for the longest period of time, in 1,000 years 3.64 Curies, 3.85 Curies and 392 Curies of Ra-226, Am-241 and C-14 would remain respectively. After 10,000 years 0.07 Curies and 133 Curies of Ra-226 and C-14 would remain. All the other isotopes buried on site should have activities below one Curie by 1,000 years. These estimates neglect any radioactivity that has left the site.

From this it can be seen that potentially hazardous levels of certain nuclides will remain in the low-level burial site well after the 100 years specified in criterion 2 as an upper limit for reliance on control by institutional functions. Based on this it would seem clear that the remedial actions taken to date be reviewed so as to assure that after 100 years the material buried will remain isolated from man and the environment for a sufficient period of time to allow certain long-lived radionuclides such as Ra-226, Am-241, and C-14 to decay away to innocuous levels. If the current remedial actions which have or will be implemented at the site cannot meet this isolation and control criterion, further actions should be prescribed.

Proposed Criterion No. 3

Radiation protection requirements for radioactive wastes should be based primarily on an assessment of risk to individuals and populations; such assessments should be based on predetermined models and should examine at least the following factors:

a. The amount and concentration of radioactive waste in a location and its physical, chemical, and radiological properties;
b. The projected effectiveness of alternative methods of control;
c. The potential adverse health effects on individuals and populations for a reasonable range of future population sizes and distributions, and of uses of land, air, water, and mineral resources for 1,000 years, or any shorter period of hazard persistence;
d. Estimates of environmental effects using general parameters or of health effects based on generalized assumptions for as long as the wastes pose a hazard to humans, when such estimates could influence the choice of a control option;
e. The probabilities of releases of radioactive materials to the general environment due to failures of natural or engineered barriers, loss of institutional controls, or intrusion; and
f. The uncertainties in the risk assessments and the models used for determining them.

This criterion addresses the issue of risk assessment.

To meet this criterion several actions should be undertaken. These are as follows:

a. Using existing inventories and data from ongoing studies, the physical and chemical properties of the waste should be reviewed to determine any unique characteristics which would lead to future problems in isolating the waste.

b. Alternative methods of control should be reviewed in the future as new methods become available. Existing methods which have not been considered, if any, should also be reviewed.
c. The potential adverse health effects on individuals and populations for reasonable future population sizes and distributions should be estimated for a period of up to 1,000 years. This should probably be in the form of a risk assessment and should address future land, air, water, and mineral resource uses.
d. Should the material buried at the site be shown to have a hazard potential greater than 1,000 years, an effort should be made to define this potential in terms of environmental and health effects for the hazardous lifetime of the waste.
e. The probabilities of release of the radioactive material buried at the site to the general environment due to failures of natural or engineered barriers, loss of institutional controls, or intrusion should be quantified.
f. The uncertainties in the analyses used to determine matters discussed in "a" through "e" above should be addressed.

Should the results of any of these undertakings listed in "a" through "f" show unnecessary risks, further remedial actions above what is currently being done will be necessary.

Proposed Criterion No. 4

Any risks due to radioactive waste management or disposal activities should be deemed unacceptable unless it has been justified that the further reduction in risk that could be achieved by more complete isolation is impracticable on the basis of technical and social considerations; in addition, risks associated with any given method of control should be considered unacceptable if:

a. Risks to a future generation are greater than those acceptable to the current generation;
b. Probable events could result in adverse consequences greater than those of a comparable nature generally accepted by society; or
c. The probabilities of highly adverse consequences are more than a small fraction of the probabilities of high consequence events associated with productive technologies which are accepted by society.

This criterion addresses the issue of unacceptable risk.

To fulfill this criterion three determinations should be made:

a. Risks to future generations will be no greater than those acceptable by this generation. This does not mean risks must be equal to or less than those imposed on current generations. They could be greater as long as the current generation would accept the risk.
b. Probable events will not create adverse consequences greater than those of a comparable nature generally accepted by society.
c. Probabilities of highly adverse consequences are no more than a small fraction of the probabilities of high consequence events which are associated with productive technologies and which are accepted by society.

Should it be impossible to make reasonably assured affirmative determinations on these points it will not be possible to make a positive determination on the acceptability of the risk at the West Valley site and further remedial attention will be needed to asssure this criterion is attained and the risk is acceptable.

Proposed Criterion No. 5

Locations for radioactive waste disposal should be chosen so as to avoid adverse environmental and human health impacts and, wherever practicable, to enhance isolation over time.

This criterion addresses location and waste isolation.

Since the site currently exists, it is questionable whether a criterion involving siting is applicable. If it is found that the site for reasons of geology, hydrology, and meteorology cannot avoid adverse environmental and human health impacts and that removal of the waste and disposal in another place is less risky, then this criterion might apply and might cause evaluation of this alternative (the authors note that there does not seem to be any substantial evidence supporting this premise at this time). However, this criterion should be closely scrutinized if the site is considered for future disposal.

Proposed Criterion No. 6

Certain additional procedures and techniques should also be applied to waste disposal systems which otherwise satisfy these criteria if use of these additional procedures and techniques provide a net improvement in environmental and public health protection. Among these are:

a. Procedures or techniques designed to enhance the retrievability of the waste; and
b. Passive methods of communicating to future people the potential hazards which could result from an accidental or intentional disturbance of disposed radioactive wastes.

This criterion addresses supplementary protection goals.

Two areas of remedial action are evident from this criterion. One involves developing the best possible monitoring system around the site with the goal of detecting potential problems before they become hazardous. The other involves designing better passive means of communicating the potential hazards to future generations. The idea of retrievability of waste does not seem feasible since the material is already buried. However, should the site be considered for use in the future it would be wise to consider upgrading waste containerization to allow retrievability for as long a period of time as is reasonably possible.

Summary

The past history of the West Valley low-level radioactive waste burial site reveals that radioactivity has been and can be released to the environment. Remedial actions have been undertaken to alleviate this problem. Work must now

be done to assess the hazard potential of the radioactive material over time, to assure that the risk attributable to the site is adequately assessed, to assure the acceptability of the risk, and to assure that the controls at the site are adequate. This work may show that more remedial attention is needed at the site in the form of engineered barriers and safeguards to meet the proposed EPA criteria for radioactive waste. Upgrading monitoring and passive communications safeguards also seems desireable.

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ACCEPTABLE RESIDUAL RADIOACTIVE CONTAMINATION LEVELS FOR SITES OF DECOMMISSIONED NUCLEAR FACILITIES

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Abstract

Examination of existing guidelines and regulations has led to the conclusion that there is need for a general method to derive residual contamination levels that should be acceptable to the public in return for use of any site of decommissioned nuclear facilities. The method used for this study is to determine these acceptable environmental levels based on a maximum annual dose to an individual from the residual contamination via all environmental pathways. A maximum annual dose criterion of one mrem is selected for purposes of illustrating the method. Acceptable residual radioactive contamination levels for the sites of three current-design nuclear facilities are presented.† The reference facilities considered are: a Fuel Reprocessing Plant (FRP), a Pressurized Water Reactor (PWR), and a small Mixed Oxide Fuel Fabrication Plant (MOX). Using maximum annual dose as a basis permits an accurate accounting of the impact of radionuclide mixtures at each unique nuclear site. The results presented consider all probable radiation exposure pathways contributing significantly to the maximum annual dose to an individual from chronic exposure to the residual radioactive contamination on the site. For chronic radiation exposure, the year in which the maximum annual dose occurs depends upon the chemical and physical characteristics of the residual radionuclides, the body organ of reference, and the radiation exposure pathway. The acceptable residual radioactive contamination levels are based on the calculated maximum annual dose resulting from the radionuclide mixture accumulated from a calculated annual release during the facility operating lifetime. The results of this study show acceptable radioactive contamination levels at plant shutdown in units of microcuries per square meter of 5.0×10^{-3} for the FRP, 1.2×10^{-2} for the PWR, and 1.3×10^{-2} for the MOX.

Introduction

Our examination of existing guidelines and regulations has led us to the conclusion that there is a need for a general method to derive acceptable radioactive contamination levels that can be applied for the release of any decommissioned nuclear site for public use (Sc77, Sm78, Je79). Some guidance currently exists defining the levels of radioactive surface contamination which are acceptable to the U. S. Nuclear Regulatory Commission for the termination of operating licenses (NRC74, AEC70). Other suggested guidance is directed toward specific types of nuclear facilities, or accident situations involving radioactivity (CFR49-76, ERDA75, He74, Gu64, Ha75, ANSI78).

None of these guidelines is sufficiently flexible to accommodate the various radionuclide mixtures or site specific features unique to each nuclear

† Based on work prepared for the Division of Engineering Standards, Office of Standards Development of the U. S. Nuclear Regulatory Commission.

facility. We believe that the methodology used to calculate acceptable residual radioactive contamination levels should be based on a more general concept; a concept that is capable of accommodating these unique radionuclide mixtures and site specific features. One general concept for determining acceptable radioactive contamination levels is to compare established annual dose limits with calculated annual doses to members of the public.

There are currently no unique regulations or specific guides on acceptable maximum annual dose to individuals living on or near a decommissioned site. Guidance that could be interpreted as annual dose limit recommendations specifically for the cases of interest here include:

- Recommendations of the International Committee on Radiation Protection (ICRP), Publication 9 (ICRP66)
- Surgeon General's Guidelines (DHEW) (PHS71)
- Appendix I of 10 CFR 50, Guides for Design Objectives for Light-Water-Cooled Nuclear Power Reactors (NRC) (CFR10-76)
- Proposed Federal Guidance for the Environmental Limits of Transuranium Elements (EPA) (EPA77)
- 40 CFR 90 Environmental Radiation Protection Requirements for Normal Operations of Activities in the Uranium Fuel Cycle (NRC) (CFR40-77)

The purpose of this paper is to describe our methodology for determining acceptable residual radioactive contamination levels based on the concept of limiting the annual dose to members of the public. It is not our purpose to recommend or even propose dose limits for the exposure of members of the public to residual radioactive contamination left at decommissioned nuclear sites. Thus, example acceptable levels of residual radioactive contamination are calculated only to demonstrate the annual dose-based methodology for an assumed annual dose of one millirem. The reference sites we considered are a Fuel Reprocessing Plant (FRP), Pressurized Water Reactor (PWR), and a small Mixed-Oxide Fuel Fabrication Plant (MOX).

The following terminology is used in developing the annual dose-based methodology:

- Disposition Criteria The acceptable radioactive contamination levels for public use of decommissioned nuclear sites, based on a maximum annual dose limit.
- Organs of Reference Radiation doses are calculated for specific organs of the human body. In this study these organs of reference are the thyroid glands, lungs, total body, and bone.
- Exposure Pathways The radiation exposure pathways represent ways by which people are exposed to radiation. Exposure pathways of concern in calculating the dose to members of the public located on a decommissioned nuclear site are inhalation of radionuclides, external exposure from radioactive surface contamination, and the ingestion of food products containing radionuclides.

- Decay Periods The continually changing mixture of the radionuclide inventories results in annual doses that are time-dependent due to radioactive decay. This dependence is demonstrated by calculating the doses at the time of the facility shutdown, and at 10, 30, and 100 years after shutdown.

- Annual Dose The annual dose is the radiation dose equivalent calculated during any year following continuous exposure. It is the sum of the doses received during the year of interest from all exposure pathways including the dose resulting in that same year from the intake of radionuclides during previous years. The highest value calculated for any year following the unrestricted public release of the site is referred to as the maximum annual dose.

- Maximum Exposed Individual The maximum exposed individual is assumed to reside at the location of the highest airborne radionuclide concentration, and maximized exposure pathway parameters are used.

- Unrestricted Use The unrestricted use of the decommissioned site means that the potential exposure to members of the public from residual radioactive contamination levels will not exceed the maximum annual dose limit as may be established by Federal and state regulatory agencies. Decommissioning a site will, in general, result in the unrestricted public use of land areas that the public had been denied use of during the nuclear facility operational life.

Maximum Annual Radiation Dose

The annual dose from radiation exposures originating from sources external to the human body tends to decrease with time after shutdown due to radiative decay of the residual radioactivity. Annual doses to organs of reference resulting from chronically ingested or inhaled radionuclides, however, tend to increase with time after shutdown until a maximum value is reached. The annual dose from internal sources then tends to decrease with time due to both radioactive decay of the residual radioactivity and biological elimination of radionuclides deposited in the organ of reference. For continuous exposure to a radioactively decaying source, the year in which this maximum annual dose occurs depends on the chemical and physical characteristics of the radionuclides, the organ of reference and the environmental pathways considered.

A fundamental relationship for the calculation of radiation dose to man from environmental pathways for any radionuclide is given as follows:

$$R_{ipr} = C_{ip}\ U_p\ D_{ipr} \qquad (1)$$

where:

R_{ipr} • the radiation dose equivalent or committed dose equivalent from nuclide i via pathway p to organ r;

C_{ip} • the concentration of nuclide i in the media of pathway p;

U_p • the exposure rate or intake rate associated with pathway p; and

D_{ipr} • a radiation dose equivalent or committed dose equivalent factor: a factor for a given nuclide i, pathway p and organ r that converts a specified concentration of the radionuclide and the intake rate of that radionuclide to the radiation dose equivalent or committed dose equivalent.

Specific equations tailored to each exposure pathway are derived from Equation 1. A more complete discussion of the radiation dose model, the variation of annual dose with time after shutdown, and the parameters used can be found in the literature (Sc77, Sm78, Je79). The principal difference among pathways is the manner in which the radionuclide concentrations in air, soil, or food products are calculated as an integral part of the computerized models used in this study (So74, Ba76). They are functions of such parameters as the radionuclide release rates, resuspension rates, deposition rates, root uptake parameters, and atmospheric dispersion.

Disposition Criteria Methodology

The methodology we used to determine disposition criteria based on annual dose is shown in Figure 1. The three steps in this methodology are:

- Compute the Maximum Annual Doses The maximum annual doses to the organs of reference resulting from the radioactive contamination present at the radioactive decay times of interest are calculated using the radiation dose methodology discussed previously.

- Compute the Contamination Levels The residual contamination levels, or disposition criteria, expressed in units of microcuries per square meter ($\mu Ci/m^2$) are calculated for the organs of reference using a normalized annual dose value of 1 mrem.

- Determine the Maximum Acceptable Contamination Level The maximum acceptable contamination level at the assumed maximum annual dose is determined by selecting the most restrictive calculated organ dose derived from all exposure pathways. This value is dependent on the composition of the radionuclide inventory.

Example Calculations

Radioactive contamination is expected to be present on the site as a result of effluents released during normal operations over the anticipated plant life. The concentration of deposited radionuclides was estimated using an NRC computer model (Sa76). Continuous annual releases at a constant level were assumed over the plant life. We assumed the effective release height to be 100 meters for the FRP and 10 meters for the PWR and MOX. Deposition values reported are for the area within the site boundary (the area within a radius of 1,000 meters from the point of release). Offsite contamination levels are expected to be lower.

The resulting surface contamination compositions for the reference FRP, PWR and MOX are shown in Tables 1, 2 and 3 for various times after plant shutdown. It should be noted that the contamination levels defined for the site by Tables 1 through 3 are probably higher than might be encountered at real facilities. This is primarily because no credit was taken for weathering effects on the radioactive contamination either during the facility operating life or during the time after shutdown. For specific sites, comprehensive measurements will be necessary at shutdown to characterize the quantity and mixture of the deposited radioactive contamination.

Maximum annual doses calculated for the site radionuclide inventories (Step 1 in Figure 1) are listed in the literature (Sc77, Sm78, Je79). Acceptable contamination levels for a dose of 1 mrem/yr are calculated (Step 2 of Figure 1), and disposition criteria based on the most restrictive annual dose to any organ of reference (Step 3 of Figure 1) are determined. These values are listed in Table 4 in units of $\mu Ci/m^2$. The dominant radionuclide contributor to the organ doses are listed in Table 4 to help illustrate the dependence of the calculated doses on the assumed radionuclide mixture.

For the FRP, the disposition criteria are controlled at all times after shutdown by the dose to the thyroid gland because of the dominance of ^{129}I in the radionuclide mixture. For the PWR, because of ^{90}Sr in the radionuclide mixture, the deposition criteria are controlled by the dose to bone at all times after shutdown. The primary pathway for both ^{129}I and ^{90}Sr deposition in humans is the consumption of leafy vegetables grown on the site.

The change in limiting organ dose from lungs to bone between 0 and 10 years after shutdown for the MOX reflects the impact of the time-dependent resuspension factor used in our analysis. We assumed that the quantity of material available for resuspension decreased with time after ground deposition using the Anspaugh model (An75) until a constant value is reached around 20 years after deposition. With more material resuspended at short times after shutdown, the dose to the lungs is limiting. As the amount of resuspended material decreases, the translocation of material from the lung and GI-tract to the bone controls the annual dose.

Summary and Conclusions

The methodology that we have presented in this paper can be used to calculate defensible acceptable residual contamination levels that are directly relatable to risk assessment with the proviso that an annual dose limit will be established. Our methodology is shown to be flexible enough to permit variations in the radionuclide mixtures and site specific data required in the calculation of the annual doses. The maximum annual dose should be used for comparison to an annual dose limit, and not a 50-year committed dose equivalent or the first year dose. The first year dose is not conservative when internal exposure is the dominant pathway, and it is not appropriate to compare a 50-year committed dose to an annual dose limit.

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Table 1. Estimated Maximum Radioactivity Deposited on the FRP Site Over a 30-Year Operating Lifetime

Radionuclide	Deposited Radioactivity ($\mu Ci/m^2$) at Selected Times After Shutdown			
	Shutdown	10 Years	30 Years	100 Years
^{89}Sr	1.5E-5	--(a)	--	--
^{90}Sr	1.4E-3	1.1E-3	6.9E-4	1.3E-4
^{90}Y	1.4E-3	1.1E-3	6.9E-4	1.3E-4
^{91}Y	2.8E-5	--	--	--
^{95}Zr	5.5E-5	--	--	--
^{95}Nb	5.5E-5	--	--	--
^{103}Ru	4.9E-5	--	--	--
^{106}Ru	2.4E-3	2.6E-6	2.9E-12	--
^{110}Ag	2.0E-6	8.8E-11	--	--
^{125}Sb	2.7E-5	2.1E-6	1.3E-8	--
^{127}Te	2.1E-6	1.7E-16	--	--
^{129}Te	2.5E-7	--	--	--
^{129}I	6.4E-3	6.4E-3	6.4E-3	6.4E-3
^{131}I	1.6E-5	--	--	--
^{134}Cs	5.5E-4	1.9E-5	2.3E-8	--
^{137}Cs	2.0E-3	1.6E-3	9.9E-4	2.0E-4
^{141}Ce	5.1E-6	--	--	--
^{144}Ce	7.4E-4	9.9E-8	1.8E-15	--
^{147}Pm	3.1E-4	2.2E-5	1.1E-7	1.0E-15
^{154}Eu	6.8E-5	3.0E-5	6.0E-6	3.1E-8
^{155}Eu	3.7E-5	8.7E-6	4.8E-7	1.9E-11
^{234}U	2.0E-7	2.0E-7	2.0E-7	2.0E-7
^{235}U	4.4E-9	4.4E-9	4.4E-9	4.4E-9
^{236}U	7.4E-8	7.4E-8	7.4E-8	7.4E-8
^{238}U	8.0E-8	8.0E-8	8.0E-8	8.0E-8
^{238}Pu	1.6E-4	1.5E-4	1.3E-4	7.3E-5
^{239}Pu	2.0E-5	2.0E-5	2.0E-5	2.0E-5
^{240}Pu	3.1E-5	3.1E-5	3.1E-5	3.0E-5
^{241}Pu	2.7E-3	1.3E-3	3.0E-4	1.9E-6
^{242}Pu	8.6E-8	8.6E-8	8.6E-8	8.6E-8
^{241}Am	1.0E-4	1.5E-4	1.9E-4	1.9E-4
^{243}Am	4.6E-7	4.6E-7	4.6E-7	4.6E-7
^{242}Cm	9.4E-6	1.7E-12	--	--
^{244}Cm	3.6E-5	2.4E-5	1.1E-5	7.4E-7

(a) Dash indicates deposition is less than 10^{-15} $\mu Ci/m^2$.

Table 2. Estimated Maximum Radioactivity Deposited on the PWR Site Over the 40-Year Plant Lifetime from GESMO (NRC76) Study Annual Releases[a]

Radionuclide	Deposited Radioactivity ($\mu Ci/m^2$) at Selected Times After Shutdown			
	Shutdown	10 Years	30 Years	100 Years
^{54}Mn	4.2E-4	9.1E-8	4.3E-15	--[b]
^{58}Co	1.5E-4	--	--	--
^{60}Co	5.3E-3	1.4E-3	9.8E-5	8.7E-9
^{59}Fe	6.3E-6	--	--	--
^{89}Sr	7.5E-6	--	--	--
^{90}Sr	9.0E-4	7.0E-4	4.2E-4	7.1E-5
^{90}Y	9.0E-4	7.0E-4	4.2E-4	7.1E-5
^{131}I	2.8E-5	--	--	--
^{133}I	2.8E-6	--	--	--
^{134}Cs	5.2E-4	1.8E-5	2.1E-8	--
^{137}Cs	9.3E-3	7.4E-3	4.7E-3	9.3E-4

(a) Normalized to the reference PWR power rating of 1175 MWe and plant capacity factor of 0.75.

(b) A dash indicated values less than 10^{-15} $\mu Ci/m^2$.

Table 3. Estimated Maximum Radioactivity Deposited on the MOX Site Over a 10-Year Operating Lifetime

Radionuclide	Deposited Radioactivity (pCi/m^2) at Selected Times After Shutdown			
	Shutdown	10 Years	30 Years	100 Years
^{234}Th	1.5E-4	--(a)	--	--
^{233}Pa	2.4E-6	2.4E-6	2.4E-6	2.4E-6
^{234}U	3.0E-4	3.0E-4	3.0E-4	3.0E-4
^{235}U	7.0E-6	7.0E-6	7.0E-6	7.0E-6
^{236}U	6.2E-7	6.2E-7	6.2E-7	6.2E-7
^{237}U	4.5E-3	--	--	--
^{238}U	1.5E-4	1.5E-4	1.5E-4	1.5E-4
^{237}Np	2.4E-6	2.4E-6	2.4E-6	2.4E-6
^{238}Pu	8.8E-1	8.1E-1	7.0E-1	4.0E-1
^{239}Pu	7.2E+0	7.2E+0	7.2E+0	7.2E+0
^{240}Pu	3.5E+0	3.5E+0	3.5E+0	3.5E+0
^{241}Pu	1.8E+2	1.1E+2	4.2E+1	1.5E+0
^{242}Pu	8.0E-4	8.0E-4	8.0E-4	8.0E-4
^{241}Am	1.9E+0	4.1E+0	6.3E+0	6.8E+0

(a) A dash indicates values less than 10^{-7} pCi/m^2.

Table 4. Disposition Criteria on the Reference FRP, PWR, and MOX Sites Corresponding to a Maximum Annual Dose of 1 mrem per Year[a]

	Time After Shutdown Years	Limiting Organ	Dominant Radionuclide	Disposition Criteria $\mu Ci/m^2$
FRP	0	Thyroid	^{129}I	0.0050
	10	Thyroid	^{129}I	0.0038
	30	Thyroid	^{129}I	0.0029
	100	Thyroid	^{129}I	0.0020
PWR	0	Bone	^{90}Sr	0.012
	10	Bone	^{90}Sr	0.0096
	30	Bone	^{90}Sr	0.0091
	100	Bone	^{90}Sr	0.0098
MOX	0	Lungs	^{239}Pu	0.013
Class Y	10	Bone	^{239}Pu - ^{241}Am	0.49
	30	Bone	^{239}Pu - ^{241}Am	0.23
	100	Bone	^{239}Pu - ^{241}Am	0.077

(a) Includes doses from inhalation, external exposure, and ingestion of food products.

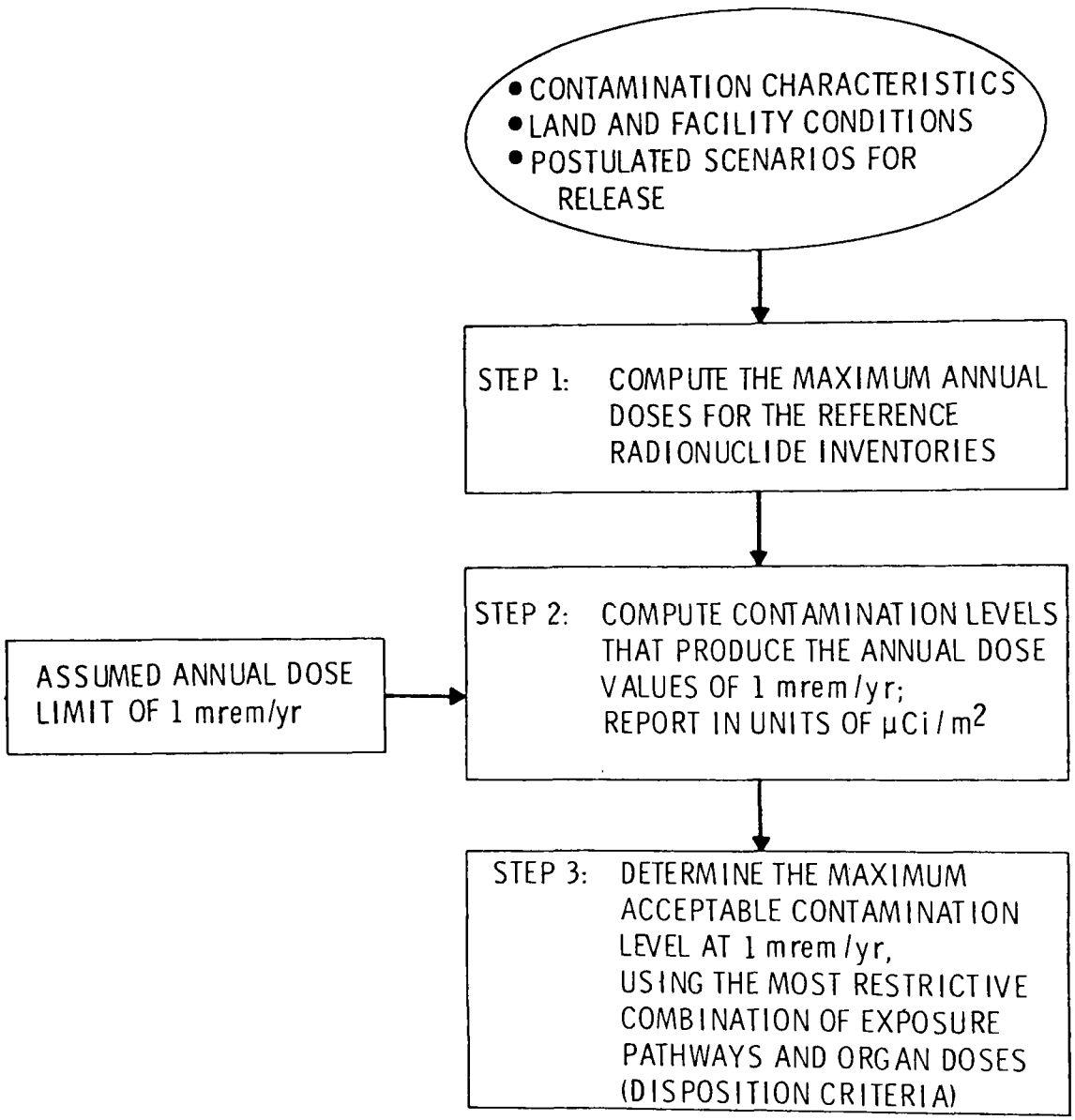

Fig. 1. Disposition Criteria Methodology

DOSIMETRIC AND RISK/BENEFIT IMPLICATIONS OF Am-241 IN SMOKE DETECTORS DISPOSED OF IN NORMAL WASTES

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Abstract

The risks associated with ^{241}Am in residential-type smoke detectors have been assessed (using a linear dose-response model) by evaluating collective dose commitments (70 years) to the public delivered during normal detector use and disposal. The risk of radiation induced harm results mainly from external exposure, with only a small contribution from waste management practices. A reference case of 100 million detectors distributed in residences at a rate of 10 million per year leads to an external dose of 340 man-rems per year to the U.S. population and a cancer risk of 0.07 cases per year. Collective background is 20 million man-rems per year. Lives saved by detectors from fires are estimated at 2400 cases per year, for a benefit (lives saved) per risk (potential cancers induced) ratio of 34,000.

Objective

The analysis reported here is based partly on the results of a study (Wr79) conducted for the Ionization Smoke Detector Bureau of the National Electrical Manufacturers' Association to assess the potential radiation risks associated with home-type ionization smoke detectors containing ^{241}Am, and put these risks in perspective relative to the benefits to be gained; similar evaluations have been reported elsewhere (Nu78; Or77; Na77a).

This evaluation addresses only single station (home-type) ionization smoke detectors; a reference case has been chosen for analysis in which 10 million detectors, having a mean effective life in the home of ten years, are produced and distributed per year. At equilibrium there would be 100 million detectors deployed, each with two microcuries of activity, less than that distributed in the past but representative of current output.

Approach

The approach taken was to assess the potential radiation exposure to individuals and populations associated with the deployment of these detectors. The collective and individual risks were then estimated by assuming a linear dose-response relationship and multiplying the dose by an effects coefficient established from human and animal information. For this purpose, total cancer induction was taken as the measure of risk.

The benefit to risk ratio is expressed as the potential number of lives saved from fires versus the estimated number of lives at risk as a result of radiation exposure. In addition, the individual and collective doses associated with the deployment of the detectors can be compared to natural background as a means of putting the exposures in perspective.

Finally, a comprehensive literature review was conducted and pertinent calculations were adopted from other analyses, with suitable modifications to the reference U.S. case chosen. In addition, detailed consideration was given to specific situations not analyzed previously.

Detectors, ^{241}Am Source Characteristics, and Integrity Tests

Detectors consist of a "tamper-proof" chamber housing the ionization source, electronics to sense a change in voltage when smoke enters the sensitive volume, and warning or signaling alarms.

The active portion (^{241}Am source) is manufactured by three different manufacturers by similar but not identical processes. The following is a brief description of the general process:

The majority of Ionization Chamber Smoke Detectors (ICSD) utilize the oxide form of the alpha-emitting radionuclide ^{241}Am ($^{241}AmO_2$). Americium oxide is uniformly mixed with gold, formed into a briquette and sintered at above 800°C. The briquette is then mounted between a backing of silver and a front cover of gold or gold/palladium alloy and sealed by hot forging. The composite material thus formed is cold rolled to give the desired activity loading which ranges from 0.01 to 2.5 $\mu Ci/mm^2$. An additional corrosion resistant material, either rhodium or gold, is commonly electrodeposited on the top surface. Total foil activity ranged from 0.5 to 130 μCi in the past with modern designs utilizing 1 to 2 μCi ^{241}Am. The sources are commonly fixed onto metallic holders usually stainless steel or plated brass by soldering or crimping them to the holder wall.

Tests of detector and source integrity have been made by various organizations including the source and detector manufacturers, independent testing laboratories, and government-related laboratories. Tests include acid leaching, abrasion, corrosive atmospheres, and high temperatures simulating fires (Ba76b; Ea77; Gr76; Ha75,78; Hi76; Ni69; Sw69).

Removable fraction (i.e., by wiping plus immersion in water) ranged between 10^{-3} to 10^{-6}. In most fire tests the airborne fraction is normally one to two orders of magnitude less than the mechanically removable fraction; the latter averaged 10^{-3} in the reports surveyed. We have used 10^{-2} for our analysis for conservatism and to take into account the possible effect of high temperature incineration.

The number of detectors in use has increased greatly in the last few years while at the same time the average amount of ^{241}Am per detector has decreased. In 1970, for 53,000 smoke detectors (not just home-type) containing ^{241}Am, the average activity per detector was 79 μCi with a total of about 4.7 Ci cumulative in all detectors. By 1975, the average activity of ^{241}Am per detector had dropped to 16 μCi, but the total Ci used, 10.8, increased because the number of detectors increased to 388,000. By 1977, detector sales had increased to 7.3 million (this is based on a calendar year from July 1 of the previous year through June 30 of the reference year) with an average activity of 5.7 μCi, and a total amount of ^{241}Am of 42 Ci that year (Nu78).

Thus to mid-1977, nationally, there has been about 105 Ci distributed in detectors. Industry figures show that in calendar year 1978, slightly more than 11 million ionization detectors were sold, with an average activity of about 4 μCi.

For 1979 the expected activity per detector is less than 2 μCi (Ha78). Thus although there has been an enormous expansion in production, the total activity distributed in detectors has increased less rapidly than the number of units because advances in technology has allowed the use of less activity per unit. Sales of 10 to 11 million units are expected in 1979.

Radiological Characteristics of ^{241}Am

Americium is an actinide element of atomic number 95 first identified late in 1944. Americium-241, the isotope used in smoke detectors, has a radiological half-life of 433 years and emits alpha particles and gamma and X-ray photons with abundances as follows: $\bar{E}_{\alpha}$ = 5.48 MeV; γ_1 = 59.5 MeV, 36%; γ_2 = 0.026 MeV, 2.6%; and Neptunium L-X rays with an average energy of 18 keV emitted with an abundance of 37.6%.

Dosimetry and Risk Evaluation

Based on the results of animal experiments and some metabolic information obtained *in vivo* in man, americium is retained primarily in three organs, lung, after inhalation, from which it is transported primarily to liver and skeleton (Du73; In59; Wr72).

The model and metabolic parameters of ICRP-19 (In72) were used in the calculations of inhaled dose. In calculating the ingestion dose, the ICRP-2 (1959) model was used for which a GI absorption of 10^{-4} and other metabolic parameters taken from ICRP-19. Calculations of the inhalation dose were made with the DACRIN Code (Ba75; Ba76a) similar to those first reported by Strom and Watson (St75) but modified to use ICRP-19 rather than ICRP-2 parameters (Wa78).

Estimates of cancer risk were compiled (Bai76; Be72; Ene77; Ma72; Ma76a; Me7 Ne75) from several publications to derive a risk of 1.2×10^{-1}/μCi ingested. Per unit activity, inhalation carries four orders of magnitude greater risk than ingestion. Risk and dose estimates are shown in Table 1. Americium-241 oxide probably behaves intermediately between Class W and Y. The risk estimate for inhalation of Class W is 2.7×10^{-2}. For external exposure, we used the BEIR absolute linear risk coefficient, 2×10^{-4}/rem (Be72).

The 59 keV gamma ray gives a dose rate of 9 nrads per hour at 1 meter from a detector containing 1 microcurie.

Assessment of Exposure and Risk to House Occupants for Normal ICSD Use - External Dose

A. Individual Dose

The most important component of dose turns out to be external dose from the detectors.

The potential for external exposures by smoke detectors has been evaluated by the NEA-OECD (Or77), which developed a methodology to make an assessment of individual and collective doses from the detectors. This methodology was applied by Johnson (Jo78) to the United States population and will also be adopted in the present evaluation. In addition, Graham (Gr78) has calculated and measured dose rates from different detectors containing ^{241}Am. The excess dose noted by Graham above 9 nrads per hour per microcurie of ^{241}Am at 1 meter results from inclusion of the X rays in the external dose estimates.

Although the detector housing normally provides some shielding from the X rays, they are not completely eliminated. The X rays have not been included in the dose estimates here however because they are not sufficiently penetrating to be considered as "whole body" radiation, and their application in "total body" risk estimates would be inappropriate.

The average annual dose, according to the formula developed by OECD and for persons sleeping approximately 6 feet from a detector, 8 hours per day, would be 14 microrads, or roughly 80 minutes of natural background. The dose varies as the inverse square from the detector and at one meter would be four times larger. Little information exists on distances people actually sleep from detectors on the average, so this might overestimate or underestimate the dose.

B. Collective Dose Estimate

The collective dose has been evaluated by assuming that 100 million detectors containing 2 μCi each are distributed in 50 million homes, with 90% of the units being positioned in hallways and the other 10% in bedrooms. Again assuming an average distance of 2 meters, the collective dose is estimated as 340 man-rems. The assumption is made that 3 people are exposed for 1 hour per day in the halls, and 2 people for 8 hours per day in the bedroom.

If all the detectors were installed in halls this estimate would be 232 man-rems and if all were installed in bedrooms, 1872 man-rems, assuming an occupancy of 2.95 people/structure unit.

Summary of Calculation of Average Annual Collective (Dc) and Individual (Di) Dose

$$Di = \frac{\Gamma\, At}{d^2}\,(O_h + O_b);\ \text{and}$$

$$Dc = (D_{ih}P_hF_h + D_{ib}P_bF_b)\,\frac{n}{2}$$

where

Γ is the specific gamma ray constant for ^{241}Am = 8.7 nrad per hour per μCi at 1 meter;
A = ^{241}Am activity of detector, 2 μCi;
n = number of installed detectors (taken as 2 times the number of dwelling units);
t = hours in year, 8760

d = average distance of detectors from people, taken as 2 m;
F_h = fraction of detectors in hallways = 0.9;
F_b = fraction of detectors in bedrooms = 0.1;
P_h = three people in the hall;
P_b = two people in the bedroom;
O_b = occupancy factor in bedroom taken as 8 hours per day (2 m from detector);
O_b = occupancy factor in halls (2 m from detector) taken as 1 hour/day;
O_h = occupancy factor in halls (2 m from detector) taken as 1 hour/day;
D_{ih} = individual dose rate in hall; and
D_{ib} = individual dose rate in bedroom.

For these values, D_i becomes 14.2 μrads per year, with 12.6 from the bedroom (D_b) and 1.6 (D_h) from the hall. The collective dose D_c is 340 man-rads.

Exposure and Risk for Waste Disposal

The dose associated with disposal is evaluated by considering both incineration and burial of detectors. Since home-type detectors have no requirement for special disposal, it is prudent to assume that all of them will be disposed of in normal refuse.

A. Individual and Population Dose from Incineration

The analysis adopted here is based on the NEA-OECD (Or77) analysis of this problem. Assumptions used are:

Population feeding one disposal route	= 1.5×10^6
Number of private homes (2 ICSD's/home)	= 5×10^5
Average activity in single station	= 2 μCi ^{241}Am
Total amount of refuse	= 5×10^8 kg/yr
Removal efficiency from stack	= 0.9 (90%)

For the reference case of 10 million units per year, this would involve 100 incinerators.

With well-designed sources less than 1% of the activity is likely to escape or become airborne. If it is assumed that the stack is 50 m high, and that removal of 90% of the particulates from the effluent takes place (a likely circumstance since all modern incinerators will have particulate removal devices, such as electrostatic precipitators or scrubbers, generally with efficiencies exceeding 90%), then a maximum downwind concentration averaged over a year is calculated (Br64), as $\sim 10^{-11}$ μCi/m^3. Assuming a breathing rate of 20 m^3 per day, leads to an annual inhalation of 1.5×10^{-7} μCi. This gives a dose commitment to lung and bone of 100 and 80 μrems respectively, roughly 1/5000 normal annual background alpha doses to these organs.

The estimation of collective dose to the U.S. population from incineration is given in Table 2.

It is assumed that 10 million units are sold per year each having an activity of 2 μCi. In order to assess the amount that might be inhaled from eventual incineration of these devices, one needs to know the fraction of the total units that might be incinerated, the fraction of the activity that would be released and the fraction of the activity that would return from the environment back to man. All of these can be estimated to give an order of magnitude of the collective risk. From studies of fallout it is known that cumulative human retention of plutonium inhaled from weapons testing debris has been about 3×10^{-8} (Ri75). This refers to debris that has descended rather uniformly over the U.S. In the case of a large number of point sources, such as incinerators, the distribution would not be uniform throughout the U.S. but rather would be concentrated in areas which have a greater population density where the incinerators are located. For purposes of this analysis it is assumed that this will introduce a 30 fold factor greater collective inhalation by people. It is also assumed that 10% of the units produced eventually are incinerated, and that the fraction of material released from an incinerated unit is one part in a hundred. This leads to an estimate of 2×10^{-3} μCi collective inhalation with a concurrent risk estimate of 2×10^{-4} cancers per year from the operation of the whole industry from incineration.

Clearly, modern incinerators will have particle filtration systems designed to minimize the total mass loading to the environment which will reduce the emission below the amount chosen for this analysis. From this calculation it is clear that one is dealing with a very small risk, referenced to a U.S. population size of 200 million. It is possible that the estimate of the cumulative activity retained may be too low or too high, but in any event is probably not in error by two orders of magnitude which would still leave this risk estimate a very small one.

One may ask whether or not there would be a significant addition to the dose from resuspension of material which deposits on the ground. Several studies suggest that the dose from resuspension of insoluble actinides would at most be comparable to that received on direct inhalation (Wa74). This might, therefore, increase the risk estimate by a factor of two.

B. Activity in Incinerator Slag or Ash

The concentration of ^{241}Am in incinerator ash or slag may be evaluated as follows: in the U.S. (Table 3) about 20% of solid waste is left as ash after incineration. It is assumed that all of the ^{241}Am will remain with the ash. Estimates of solid waste generation per capita of 2.7 kg/day have been made for 1980 (En77).

Assuming two detectors in each of 50 million homes, with a mean life of ten years each, leads to the disposal of 10 million units per year. At present, some 8 to 13% of solid waste is incinerated, the rest going to landfill.

Future incineration could increase or decrease. Accordingly, it is assumed that 10% are incinerated.

For total incineration of all solid waste, the average concentration in ash or slag is obtained by dividing the total ^{241}Am discarded in detectors by the residual ash weight from one year's worth of incinerated solid waste, or:

$$\frac{(10\times10^{6}\ \text{detectors/y})(2\times10^{6}\ \text{pCi/detector})}{(50\times10^{6}\ \text{houses})(2.7\ \text{kg/d, person})(2.95\ \text{persons/house})\times (365\ \text{d/y})(0.21\ \text{g ash/g solid waste})(10^{3}\ \text{g/kg})} = 0.66\ \text{pCi/g}$$

For 10% of waste being incinerated, an average concentration would then be 0.07 pCi/g of waste slag which is small compared to the normal alpha actinide content in soil, about 5 pCi/g. However, there is no available data on alpha emitters in solid waste incinerator residue with which to compare this expectation.

The NEA-OECD study concluded that the majority of the activity would remain with the slag, and an analysis of the various uses to which slag might be put has shown these routes of exposure to be unimportant.

C. Disposal: Burial or Sanitary Landfill

The risk from inhalation to an individual from resuspension in the vicinity of an uncovered landfill can be estimated using the approach developed by Johnson (Jo78). It is assumed that the ^{241}Am is uniformly distributed and resuspends from soil as does uranium, a natural actinide. Johnson concludes that an average air concentration of 5.4×10^{-4} pCi/m^3 results from average uranium soil content of 0.93 pCi/g.

The concentrations of residue in landfill can be evaluated from the calculations of ^{241}Am in slag from incineration. In landfill it is assumed that the waste is mixed with an equivalent of soil, and that the fivefold reduction in mass associated with incineration does not occur. Thus, this average concentration would be:

$$\frac{0.7\ \text{pCi/g}}{10}$$

which would lead to a local air content due to resuspension of:

$$\left(\frac{0.07}{0.93}\right)\times 5.4\times10^{-4} = 4.0\times10^{-6}\ \text{pCi/m}^3.$$

This substantially overestimates the expected concentration in air since the resuspension value is valid only for a much larger area source than a single or multiple landfill. Thus, for 70 years, inhalation of 20 m^3/day would lend to an inhalation of 2 pCi, associated with a lifetime risk on the order of magnitude of 10^{-7}.

The collective risk has not been evaluated for this route but is clearly limited and likely to be less than by other pathways.

D. Dose from Ingestion

A relatively simple calculation shows that the potential risk from ingestion is less than the risk evaluated for inhalation. Bennett (Be78) has shown that ^{241}Am from weapons testing, which is widely distributed in the

environment, is very poorly returned to man via diet. The cumulative deposition on the U.S. is now about 5000 Ci, and the current annual average dietary intake about 0.4 pCi. If no change in the fraction in diet occurs over the next 70 years, then lifetime intake would be 0.4x70 = 27 pCi. In 200 million people, this represents an integrated lifetime intake of 5.4×10^3 μCi, or 10^{-6} of that distributed in the environment.

Assuming that 10% of the ^{241}Am in smoke detectors leaches or otherwise becomes available to move about the environment, the collective ingestion risk for disposal of the reference 10 million units (integrated 70 years into the future) would be:

$$\left(\frac{2\ \mu Ci}{\text{detector}}\right)(10^7\ \text{detectors})(0.1)(10^{-6})\ 1.5 \times 10^{-5}\ (\text{risk}/\mu Ci) = 3 \times 10^{-5}.$$

An alternative approach may be taken by calculating the ^{241}Am content on food grown directly on old landfill converted to agricultural uses. This approach has been used by the Nuclear Regulatory Commission (Nu78) using a concentration in waste of 0.05 pCi/g of waste. The concentration of 0.07 pCi/g derived here is comparable.

Since ^{241}Am is discriminated against by most animals, uptake by vegetation represents the most significant likely route.

If it is assumed that the waste is comparable to soil, that the ^{241}Am is evenly dispersed, that all the ^{241}Am in detectors becomes available, that plant uptake is 10^{-4}, and that 10 g daily of a person's diet comes from plants grown in waste containing such detector origin contamination, then:

$$7 \times 10^{-8}\ \frac{\mu Ci}{g} \times 10^{-4} \times 10\ \frac{g}{d} \times 365\ \frac{d}{y} \times 70\ y \simeq 2 \times 10^{-6}\ \mu Ci \text{ on a per capita basis}$$

If one million people had such diets then the collective intake would be equal to that derived from the assumption of uniform distribution.

Summary of Disposal Intakes and Risks for 10 Million Units/Year

	Collective Activity (μCi)	Risk
Incineration (inhalation)	2×10^{-3}	2.4×10^{-4}
Landfill (ingestion/food)	2	3×10^{-5}

The collective estimates of potential harm (risk) are summarized in Table 4.

Risk/Benefit

In 1977, 9,950 civilian fire deaths were estimated for the U.S., of which 8,600 occurred in structure fires, and 7,800 in residential structures.. Accordingly, "the United States fire death problem is heavily concentrated in residential fires." It is likely, therefore, that the ICSD's will be useful in reducing this toll of 7,800, which will be used for the analysis here.

There undoubtedly will be a reduction of nonfatal injuries to civilians (33,400 in 1977) and possibly also to firefighters (106,100 in 1977) if fires are detected earlier and response accordingly can be more rapid.

Property loss from fires in structures was 5.2 billion dollars, and if the proportion of property loss from residential fires to all structure fires is the same as the relative loss of life, then the value of residential fire property loss would have been about 4.7 billion dollars (De78).

Approximately 9,950 persons lose their lives in fire each year in the U.S. (46.4 fire deaths/million persons). This is almost two times higher than any other technically advanced country reporting, with Canada ranking second with 29.2 fire deaths/million persons. Two-thirds or approximately 8,600 persons/year, lose their lives from fires occurring in buildings, 9 out of 10 of which occur in private homes. Since about 75% of these fires occur during the night, people are usually asleep and are not aware of the developing fire until it is too late to save themselves. From these data, one could reasonable conclude that if the dwelling had been equipped with some type of early warning fire detection device, many of these fatalities could have been avoided (Br77, De78).

A number of studies have been conducted which allow a rough estimate of the effectiveness of smoke detectors in reducing the fire death rate.

Halpin, et al. (Ha77) have assessed the potential impact of fire protection systems by studying the outcome and histories of 73 fatal fires in the Maryland/Washington, D.C. area between June, 1976 and 1977. The method chosen was to make a thorough investigation of all fires in which fatalities occurred and then to examine the data to make a professional judgment as to the likely efficacy of various fire protection systems. The fires were primarily residential in classification.

There were 114 fatalities in these 73 fires, and about 2.6 million dollars in property losses. The conclusions were that 100 of the 114 fatalities could have been saved and 114 of the 123 injuries prevented, if fire protection systems had been installed. Property losses would have been about 73% lower if detector systems had been operating.

A 90% potential effectiveness of smoke detectors could be inferred as a result of this analysis. However, this study showed, as did others, that the human factor reduced the effectiveness below that theoretically attainable. People do not always realize that their lives are in danger when warned of a fire, and do not always behave accordingly, i.e., in a manner to insure their own survival.

The mortality rate was greatest among the young (less than age 9) and elderly (greater than 60), being 20 and 24% respectively of the total. This is consistent with the distribution of deaths in national fire statistics (Na77b). Thus, life shortening is particularly high among the young involved in fires. In Canada, McGuire and Ruscoe (1962) concluded that ICSD's may save up to 45% of average adults, and 35% of children and the infirm who die in fires.

Although neither this nor other studies provide a highly accurate basis for judging the effectiveness of smoke detectors, the potential effectiveness is nevertheless up to 90% and the practical effectiveness, although lower, is still substantial. Thus, the use of smoke detectors should be coupled with an educational program for proper response in case of an alarm.

For the purpose of this report an effectiveness of 45% will be assumed, consistent with the data reported by Halpin, et al. (Ha77) and the figure used in the NEA-OECD study (Or77).

Ten million detectors installed in 5 million homes, with about 2.95 persons/household, would serve a population of 14.75 million. For a home fire rate of 7,800/year, and an effectiveness of 0.45, and a U.S. population of 215 million, we get lives saved as

$$7{,}800 \times \frac{14.75}{215} \times 0.45 = 240 \text{ lives/year}$$

If these are introduced over a ten-year period, then the number of lives saved per year will be cumulative, or about 2,400 per year.

We may now compare this with our estimates of 7×10^{-2} potential indirect effects per year, or a benefit to risk ratio of 34,000 lives saved for every potential life shortened. The former are identifiable early deaths averted and the latter are non-observable effects which result from a calculation assuming a linear relationship between radiation dose and biological effects at levels well below natural background.

The major contribution to risk comes not from internal exposure but from external exposure. The risk could also be expressed as a fraction of natural background. For individuals the dose to a maximally-exposed individual was shown to be less than 1% of the natural background exposure. On a collective basis the natural background delivers about 20 million man-rems to the U.S. population per year, whereas doses from 100 million installed ICSD's would collectively be about 400 man-rems, or one part in 250,000.

Thus, the dose from ICSD's is trivial relative to natural background, the variation in natural background, and from most other human activities involving alteration of natural radiation exposure.

The collective disposal and external risks are shown in Table 4. The risks from external exposure (7×10^{-2}) appear to be two orders of magnitude greater than the risks from internal exposure (2×10^{-4}); waste disposal practices do not contribute significantly to the risk. The benefits, by any reasonable measure, appear to be four to five orders of magnitude greater than the risks.

Translated into human terms the potential for saving lives far exceeds the risk associated with the widespread deployment of smoke detectors containing ^{241}Am.

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TABLE 1

50-Year Committed Dose Equivalent and Risk Values Adopted for ^{241}Am, 0.5 Micron Particles

Inhalation	rem/μCi Class Y	rem/μCi Class W	Linear Estimate of Cancer Risk/rem	Risk/μCi Inhaled or Ingested Class Y	Risk/μCi Inhaled or Ingested Class W
lung*	680	69	1.6×10^{-4}	1.1×10^{-1}	1.1×10^{-2}
liver	500	1000	10^{-5}	5×10^{-3}	10^{-2}
skeleton	740	1600	4×10^{-6}	3×10^{-3}	0.6×10^{-2}
			TOTAL	1.2×10^{-1}	2.7×10^{-2}
Ingestion ($f_1 = 10^{-4}$)					
liver	0.9		10^{-5}	9×10^{-6}	
skeleton	1.4		4×10^{-6}	5.6×10^{-6}	
			TOTAL	1.5×10^{-5}	

* The dose to lung is to the pulmonary region where lung cancers are most often found in inhalation studies with actinides in experimental animals (Bai76).

TABLE 2

Calculation of Collective Dose and Risk from Incineration of Smoke Detectors

Basis:

One year's production (P) of 10 million units
Activity per unit (A), 2 μCi
Fraction of units incinerated (0.1) F_I
Fraction of activity released from the detector during incineration (F_R) = 10^{-2}
Fraction of activity released from detector which is released from the incinerator stack, F_S - 0.1
Cumulative fraction of that released and is retained by man, by inhalation, Cm = 10^{-6}
Ratio of activity inhaled to that retained, Fm = 10
Risk per unit activity inhaled, R = 0.1 μCi (cancer induction)

Then, the total estimate of risk from one year's operation until complete disposal would be:

$$PAF_IF_RF_SCmFmR$$

which is 2×10^{-4} cancers, associated with the collective inhalation of 2×10^{-3} μCi.

TABLE 3

Composition of Solid Waste
Average for 21 U.S. Cities, 1966-1969

Category	Weight (%)
Food	18.2
Garden	7.9
Paper	43.8
Metals	9.1
Glass and Ceramics	9.0
Plastics, rubber, leather	3.0
Textiles	2.7
Wood	2.5
Dirt, ash, etc.	3.7
	100.0

Composition	Typical Composition
Moisture	28.2
Carbon	25.6
Oxygen	21.2
Hydrogen	3.5
Sulfur	0.1
Nitrogen	0.6
Ash	20.8

TABLE 4

Collective Disposal Risks

		Lifetime Committed Cases/year
Incineration		2×10^{-4}
Burial		2×10^{-5}
Firefighters		10^{-5}
External	person-rem	
home	340	
transport	2	
warehousing	6	
retailing	17	
Total	365	7×10^{-2}

BANQUET ADDRESS

MANAGING LOW-LEVEL RADIOACTIVE WASTES:

BIOETHICAL CONCERNS

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INTRODUCTION

Now that the high-level waste problem has attracted considerable public exposure and exploitation, the mounting controversy over low-level waste management has begun to compete for center stage. The strategy planned at Critical Mass in 1974 continues to be successful. At that forum, Dr. Margaret Mead exhorted her audience "to make people feel that everything they value in the world is at stake" if the development of nuclear energy is allowed to continue. She stated then that "Americans aren't afraid of dying suddenly but of dying slowly." She therefore urged her listeners to concentrate on those aspects of nuclear power that would evoke the most fear, namely, the long-range deteriorating effects of low-level radiation. The association in the public mind between low-level wastes and low-level radiation continues to bear fruit in the strategy of inducing fear. However, there is reason to be convinced that in the long term, reasonable arguments will prevail. To that end, the public must be confronted with the social necessity of an equitable mangement of hazards having the potential for harmful health effects and social consequences.

By "equitable management" I mean that policy makers should first be well informed about the broad spectrum of both natural and ordinary hazards that may have health effects for large segments of a population, then make comparisons of actual costs per capita to reduce them, and only then set criteria and standards that will get the most public health protection for the many out of a finite amount of money. Hazard management is equitable only if it is proportional in relation to the actual harm that can be identified and reduced by expenditures of human effort, time, and money.

In view of ethical requirements for social justice and equity, we need to address three major problems: the adequacy of conceptual tools for assessing biohazards; the disagreement among scientific experts; the origins of value-conflicts underlying expert disagreement.

I. CONCEPTUAL TOOLS: How we structure hazard-management.

Contrary to a popular misconception, "hazards" have neither a bare facticity nor an intrinsic morality predetermining how human beings should behave in relation to them. Hazards are not baldly "there" in nature or in human transactions with it. What people regard as hazardous in any given era reflects what they have come to know about their environment, and what they value as essential or desirable on a scale of real possibilities. In short, human beings structure hazards. In that sense, hazards are human artifacts. A hazard is not by definition "toxicity of substance" or "violence of event" or "magnitude of consequences" that can be known, classified, and predicted. A hazard exists only when, and to the degree that, harmful exposure of and assimilation by the human body or other valued living systems becomes a genuine, not merely imaginable, possibility. And that possibility exists only when there is an inability or failure to devise and maintain controlling actions or safeguards.

Because there are vast uncertainties about "how the world works," it serves no human purpose to bewail our "legacy of risks to future generations," and then make the fraudulent claim that the goal of hazard management must be to assure centuries of control over toxic elements or prediction of future adverse events. I concur with Prof. William Clark in his statement that hazard management is "the adaptive design of hazard structure," and that the primary goal of hazard management is "to increase our ability to tolerate error and to take productive risks."[2] This statement stands in sharp contrast to a popular yet unexamined notion -- expressed as well as anyone by Wolf Häfele -- that "we are locked in a world of untested hypotheses (of unimplemented trials) because we dare not let experience prove us wrong. The costs of failure have grown too great."[3] Not only does this notion reflect the New Pessimism, the defeatism and pseudoscientific dire predictions, pervading our cultural climate; but it also constitutes in itself the ultimate hazard · the failure to design and maintain structures of social resiliency. It is the social

ideal of resiliency that has been a major driving force behind the emergence of highly complex and technologically advanced societies. The social ideal of resiliency impels us to cope with the risks that are inherent and unavoidable in the human condition by doing risk-analysis.

The hazy connection between hazards and risks has given rise to another popular misconception. If popular literature on the subject is any indication, "risk" is steadily acquiring the moral opprobrium reserved for other four-letter words. I do not intend to add to that moralizing. Suffice it to say that "risk" has begun to carry all the baggage associated with uncertain consequences of so-called "hard" technology in a world of big, bad, centralized, corporate industrialism. What accounts for this state of affairs? At the very least, many have adopted the uncritical assumption that risk is a normative concept for certifying consequences to human beings that are harmful, dangerous, or "bad." These contrast sharply with consequences that are beneficial, pleasurable, or good, and by implication, risk-free. We have already grown accustomed to graphs which imply this dichotomy: one axis measures risks; the other axis measures benefits. This assumption is altogether understandable because it reflects a basic value-conflict about the nature of risk-taking.[4] For some persons, risk-taking is by definition hazardous, harmful, and perhaps the result of some demonic compulsion suppressing nobler human pursuits. For others, the word risk stands for the opportunity to undertake what is challenging and venturesome, innovative and fulfilling to the human spirit in its endeavor to live "the good life." This value-conflict has developed because risk-taking is not inherently good or bad -- neither in a psychological sense nor in a moral sense. The fact that the concept of risk is negatively overloaded in practical usage has no theoretical justification. In any case risk-taking is inherent in the capacity of a social -- hence moral -- being to make conscientious, consequential decisions.

Because of a facile identification of risks with hazards, a false antithesis has been set up between risks and benefits -- as if there were a way to have one without

the other. Granted that it is a common phrase already entrenched in our regulatory lexicon, "risk-benefit analysis" is misleading. "Benefit" is a term that stands for a known or virtually certain result or reward. Risk implies an unknown or uncertain outcome, a mere possibility weighted on the side of probability that intended benefit will not materialize and, instead, harm may occur. As William May suggests, it would be far more accurate to talk about either harm-benefit analysis (so that both words would refer to expected outcomes) or risk-hope analysis (so that both words would clearly signal possibilities only.[5] The trouble with the phrase risk-benefit is twofold: it fails to express a proper symmetry, and it tends to obscure the primacy of benefit within the normative structure of human action.[6] That is to say, the primary motivating force of human activity is the foreseen and intended benefit which can be gained or lost. Even in the case of activity undertaken to avoid harm or injury, it is the intended benefit of that avoidance that is primary. In other words, in concrete decisions, what is actually "at risk" is the possibility that intended benefit may be gained or lost. If risk-analysis is going to reflect the normative structure of human action, then the concept of risk must focus primarily on the benefit acquired or foregone. When harm results, it is clearly unwanted and unintended. Risks and benefits are inseparable, not antithetical.

I will be the first to admit that these remarks will appear to be hopelessly ivory-towered and out-of-touch, in view of entrenched interests in doing risk-benefit analysis as usual. But I refuse to be deterred, because they have important implications for the common good of society.

In my view, a major problem about the growing dispute over low-level radiation hazards and low-level waste management is the inadequacy, not of risk-analysis, but of harm-benefit analysis. The first order of business here should be to gain some refinement in the concept of benefit. In one of the most comprehensive and insightful studies of risk to date,[7] Okrent and Whipple suggest that we should make qualitative distinctions which reflect significantly different types of benefit, namely those goods

(1) essential to society (e.g. food, water, energy at sufficient levels, in short basic goods);

(2) beneficial or advantageous to society (e.g. most manufacturing);and

(3) of peripheral, if any value to society (e.g. aerosal deodorants which have readily available substitutes at similar cost and lower likelihood of harm.)

Using these refinements, we can distinguish between corresponding levels of harm. Just as there are basic goods, there are also basic harms that may result from being deprived of goods essential to subsistence and material well-being. The obligation of a society to avoid basic harms and provide access to basic goods has been formulated in the ethical principlesof justice and equity. As for second-level benefits which are advantageous because they improve the quality of life of a society, the total outcomes of any social policy toward such improvements will have an unclear mix of benefits and harms. In our era, one of the most difficult questions we face as a society is how to make comparative judgments about the moral desirabilities of various harms and second-level benefits, especially when they are different in kind.

Automobile and airplane manufacturing afford major economic benefits to employees, to capital investors, and to the general health of international economies. Yet each time someone drives a car or enables an airplane to take off, the benefits one pursues may entail the possibility (risk) of unintentionally causing the death or serious impairment of a fellow human being. Any society must, at some point in policy formation, deliberately decide how we ought to balance economic benefits and costs against possible harm or loss of life.

According to critics of such balancing, a human life is of infinite value,and its loss or impairment cannot be put in a class with other "negative consequences," much less be given a finite monetary value. To do so indicates the moral bankruptcy of our materialistic, consumerized, decadent society. Cost/risk/benefit quantifications, say its critics, manifest a loss of respect for the sacredness of human life.

Those who defend this conceptual tool have sometimes used simple observations, such as "There are necessary tradeoffs in any public policy decision," or "Everyone

puts a finite, monetary value on one's life when buying life insurance, installing safety mechanisms in a home or auto, taking hazardous jobs because they pay higher wages." Such analogies are true enough, but not sufficient. The public must be educated and confronted with the fact that any society or viable economy has but a finite amount of money to spend on health protection and safety, and that the ethical problem is to get the most protection for the most people from this finite amount. To put a government, or an industry, or a company in a position of financial insolvency on the grounds that their financial liability for loss of life is infinite violates equity and justice. These principles express the obligation of society to provi access to basic goods and a reasonable quality of life for its citizens as a whole.

As a conceptual tool which attempts to enhance informed consent, cost/risk/benefit quantifications are merely one tool among many others whereby policy makers endeavor to allocate finite amounts of money in a just and equitable manner. They are not tools for putting some callous "dollar value" on human life or injury as a moral judgment of individual worth, much less of using economic losses to society as a measurement of personal expendability. We are in fact maximizing the value we as a society place on human life when we endeavor to allocate limited amounts of money in such a way as to reduce widespread hazards, thereby preventing as much loss of life and protection from injury as possible.

The fact that our tools for balancing economic costs against risks to human life are not morally or ethically objectionable does not amount to saying that they are psychologically easy and acceptable to the general public. Far from it. The task of public education in this matter is monumental. Furthermore, I concur with my colleague in social ethics, George Pickering, in his observation that "we are going to have to do more than find some level of 'acceptable risk'; we are going to have to come to terms with the question of 'justifiable harm.' There are, after all, some kinds of harm which cannot be avoided; but there are other kinds of harm which any society should not allow and against which it should adopt protective or remedial measures to the best of its ability."[8] Which is which becomes the problem.

We must face up to the discomforting task of formulating a more enlightened concept of, and method of informed consent to, unavoidable hence justifiable harm, and not divert attention away from it by focusing exclusively on "acceptable risk" criteria. In my judgment, our failure to undertake this task lies at the root of the second problem I noted at the outset: namely, the frustrating dilemma of a policy-maker who wishes to set safety standards on the basis of informed consent -- yet when he turns to scientists upon whom he relies for "expert testimony," he finds that they have basic disagreements about what data should count, how it should be interpreted, and what level of health protection is "acceptable" or "safe enough to be safe."

II. EXPERTS AND STANDARDS FOR "SAFETY": How we have institutionalized dissensus and value conflicts.

Aaron Wildavsky has recently observed, "Experts are used to disagreeing, but they are not so used to failing to understand why they disagree."[9]

From my reading, research, and reflection on the problem of "expert dissensus" (vs. consensus) especially in the matter of radiation protection, I have come to the conclusion that at the heart of the matter lies a misconception about safety, especially as it relates to risk estimates and risk acceptability.

A case in point is the unending controversy over whether or not there is a threshold for radiation below which no harmful effect occurs. A threshold concept has been generally accepted for most toxic elements. It carries the implication that below a threshold dose any exposure is "absolutely safe." But over the twenty years of evolution in radiation protection philosophy, the ICRP and NCRP came to adopt a conservative assumption, namely that it would be more prudent to assume some harmful effect from any radiation dose, however small, than to assume a threshold dose and then discover data proving it to be false. This conservative assumption carries the implication that there is no absolutely safe radiation dose except zero, and every dose greater than zero entails a corresponding risk of genetic or somatic harm. In the ensuing process of applying a linear no-threshold hypothesis to the development of standards, regulatory institutions and some of theirexpert advisers seem to have forgotten that their quest for radiation limits rests only on a hypothesis, a conservative assumption, and not on a scientifically established fact.
As Dr. G. Hoyt Whipple has observed, "The data on the biological effects of radiation can be interpreted in terms of a threshold dose, but even the vast amount of radiobiological data cannot conclusively prove the existence, or absence, of a threshold."[10]

Given this state of affairs, the dilemma of the policy maker could be mitigated if two factors in the controversy were clarified: (1) the meaning of safe;

and (2) the meaning of threshold.

As noted above, a profound misconception of "safety" dominates the controversy over radiation protection. The working assumption of policy makers and regulators has been that safety is an intrinsic, measurable, absolute property that any given system, or product or activity can and should possess. Our society has institutionalized and appointed the regulator to measure approximations to that elusive property. The mandate of the regulator is to make evermore stringent regulations, presumably to come ever closer to that property by reducing risks. However, the only risks he is expected to monitor and minimize are a small percentage of the total spectrum of risks tolerated by members of society as a whole. Intent on making a set of risks publicly "acceptable," as an index of "safety," the professional regulator must continue to propose risk-reduction without regard to economic costs or social impacts of ever-changing regulations. Seemingly, he is "only giving the public what it wants," namely safety. This spiral is likely to continue unless or until the public comprehends the fact that safety is not an intrinsic property measured by approaching zero-risk. Safety is an evolving, relational value-judgment derived from current personal or social priorities. Risks can be scientifically measured, quantified, and predicted in probabilistic terms. Safety, however, cannot be measured, much less pre-determined by the presence or absence of risks. Judgments of safety are judgments about the justifiability or unjustifiability of harm. The process of reasoning whereby ethical safety-policy decisions are made ought to be dictated--not by risk avoidance, an impossible ideal -- but by comprehensive risk/risk assessments and cost/risk/benefit ratios. When these comparisons make it clear that a point of diminishing returns on allocations of money, time, and effort has been reached by comparison with other potential hazards in a society, then the particular product or process under scrutiny is "safe enough." If indeed unintended and unwanted harm should occur despite carefully wrought safety-policy decisions, then such harm can be judged "justifiable" because unavoidable or negligible by comparison with other harms and essential benefits.

If policy makers were more circumspect about the process of reasoning from which they ought to derive ethical safety-policy decisions, there might also be less ambiguity about a disputed "threshold concept."

With their increasingly sophisticated measurements in radiobiology, specialists are capable of identifying, and extrapolating from, even minute effects of exposure to radiation. But it is a qualitatively distinct cognitive leap to make the value judgment that a zero-threshold for so-called "safe" radiation exposure ought to be written into regulatory standards. Certain radiobiologists make this value judgment. However, scientific judgments about putative effects from radiation exposure cannot and ought not to be substituted for an ethically responsible value judgment about "safety." For the policy maker, the practical threshold concept cannot be evaded. There can and must be a practical threshold below which the possibility of unintended, unwanted, and comparatively insignificant harm becomes ethically justifiable. This justification derives from a reasoning process which concludes that such effects are unavoidable and negligible by comparison with other greater radiation exposures -- both naturally occurring and applied by humans -- and with other potential hazards against which citizens ought to be protected first.

The near clinical paranoia about cancer which some citizens experience cannot be avoided. Indeed it may even be exacerbated by policy makers and politicians who are trafficking in the fearsome mystique now surrounding radiation sources. The pathologic fear of radioactivity and radiation will in time be overcome, just as mankind has transformed its fear of fire, steam locomotives, electricity, the automobile and the airplane. Meanwhile, however, the fear of cancer is only a symptom of much more pervasive fears about the fate of our human species.

The dissensus among scientists may mean that we need to devise innovative institutional methods for dealing fairly with their complaints without undermining still further public confidence in expert professionals, in safety-policy decisions, and in regulatory actions. But to do so with circumspection, we must recognize the origin of basic disagreements over value judgments.

III. UNDERLYING PHILOSOPHIES: Why we derive conflicting values.

The mounting controversy over low-level radiation and low-level wastes has revealed basic value-conflicts. We are compelled, therefore, to probe more deeply into the origins of these value-conflicts -- namely, the philosophical and ethical principles from which values derive their justification.

In this regard, it is fortunate that the Natural Resources Defense Council (a self-styled public interest group in the USA) has recently disseminated a "Report" supplementing its highly critical "Comments on Criteria for Radioactive Waste" proposed by the Environmental Protection Agency.[11] I consider it a fortunate development -- not because these political tracts are likely to advance either the public interest or public understanding of the complex issues they purport to address -- but because it throws an illuminating spotlight on the NRDC's ethical and philosophical assumptions.

In their critical Comments, the NRDC authors repeatedly chastize the EPA and proposed criteria for evading what they choose to call "the fundamental mandate of EPA" and "an uncompromisable standard" -- namely, "non-degradation of the environment." This is their rendering for "protection of the environment." The NRDC authors commend the EPA at one point for comparing hazards from human activity to hazards from the "pre-existing natural state of the area." (7) As their reason for feeling that this is an appropriate standard, the NRDC authors state that "it emphasizes the role of a trustee as one who maintains the non-renewable environment as it was originally, to pass on to the next trustee." This fundamental goal is a key consideration, says the NRDC, "because if any degradation is allowed (in the name of 'allowable radiation exposure'), there is no clear bound at which degradation becomes, by anyone's standard, too much." (3-4)

In their supplementary Report, the NRDC authors make the claim that the "public's attitude toward the environment is one of non-degradation."(26) Citing the Atmospheric Test Ban Treaty, the demise of the Plowshare Program, and

public concern over long-lived nuclides and tailing piles, the NRDC claims that it speaks for the public on two contentions:

> (1) "No amount of radioactive contamination is 'acceptable'. . . .
>
> (2) "The effects of radiation on future generations are of prime import and can not be discounted."(26)

On behalf of the public, the NRDC authors are of the opinion that -- given their assumption that longevity of hazardous lifetimes of radioactive wastes constitutes an unfair imposition of hazards and risks upon unconsulted future generations -- the ethical principles of equity and participation require waste disposal criteria to be neutral to future generations. Although admitting that the ideal of a totally neutral allocation of benefits and risks is unattainable, it nonetheless serves a purpose useful to the NRDC political strategy: it

> "finds practical application in refuting the arguments that a present commitment to nuclear power is fair because investments in a technological society now via nuclear power will benefit the future as a result of an enhanced society, more than they hurt as a result of waste hazards."(11)

The NRDC authors preface their own proposed criteria by stating that "the least unfair way of managing intertemporal relationships is for each generation to try to leave the earth as it was when they arrived. As a goal, the only acceptable distribution of hazards and benefits is the neutral allocation, where no pattern of benefits and hazards is imposed."(28)

From their version of a theory of Justice and Equity, the NRDC authors derive a criterion which purports to consider only the risks to future generations, and to ignore the net benefits of using nuclear energy. The original unmined ore bodies and their cumulative risks to future generations are to be established as the measure against which cumulative risks from nuclear operations of all types (mining, milling, fuel processing, decommissioning, waste disposal) are to be compared for acceptability. Considerations of cost are secondary. If it should happen that our society does not

> "wish to bear the monetary costs of justice, then we should explicitly acknowledge that we prefer being wealthy and evil to being poor and righteous and not try to justify our moral vacillation with a cloud of cost/benefit models."(29; emphasis added)

These NRDC tracts give us much to ponder about beams and motes in the eyes of special interest politics, and in that regard, they are a helpful exercise in edification. But as for providing the public with thoughtful and persuasive analysis of the intellectual questions posed by human transactions with natural and man-made radiation sources, they are an exercise in obscurantism.

By espousing as a fundamental philosophical principle, "non-degradation of the environment," the NRDC joins with the Sierra Club in defining a "degraded environment" as any place that human actions have affected or changed. Formulas such as these obscure two questionable assumptions:

(1) that an untouched "natural environment" by definition manifests a superior, if not sacred order which human interventions violate to some degree; and secondly,

(2) that a trustee of a so-called "natural environment" can do nothing more nor less than pass it along in its original pristine state; to do otherwise is to be guilty of a moral wrong.

The philosophy of non-degradation has a long history, as is clear to anyone who has read Book I of Georgius Agricola's DE RE METALLICA, published in 1556.[12] This sixteenth century inventory of objections to disturbing the earth cite the following:

> "The earth does not conceal and remove from our eyes those things which are useful and necessary to mankind, but on the contrary, like a beneficent and kindly mother she yields in large abundance from her bounty and brings into the light of day the herbs, vegetables, grains, and fruits, and the trees. The minerals on the other hand she buries far beneath in the depth of the ground; therefore they should not be sought. But they are dug out by wicked men who, as the poets say, are the products of the Iron Age. (6-7)
>
> . . . The strongest argument of the detractors is that the fields are devastated by mining operations. . . the woods and groves are cut down, for there is a need of an endless amount of wood for timbers, machines, and the smelting of metals. And when the woods and groves are felled, then are exterminated the beasts and birds, very many of which furnish a pleasant and agreeable food for man. Further, when the ores are washed, the water which has been used poisons the brooks and streams, and either destroys the fish or drives them away. . . Thus it is said, it is clear to all that there is greater detriment from mining than the value of the metals which the mining produces. (8)
>
> And next they raise a great outcry against other metals, as iron, than which they say nothing more pernicious could have been brought into the life of man. For it is employed in making swords, javelins, spears,

> pikes, arrows -- weapons by which men are wounded, and which cause slaughter, robbery, and wars. . . . It is claimed too, that lead is a pestilential and noxious metal, for men are punished by means of molten lead. (11)
>
> They contend that, inasmuch as Nature has concealed metals far within the depths of the earth, and because they are not necessary to human life, they are therefore despised and repudiated by the noblest, and should not be mined, and seeing that when brought to light they have always proved the cause of very great evils, it follows that mining is not useful to mankind, but on the contrary harmful and destructive." (11-12)

The NRDC appeal to a non-degradation philosophy only demonstrates that those who are ignorant of history are condemned to repeat it.

Furthermore, the philosophy of non-degradation uncritically assumes the idea that a benign environment is rapidly being ruined by human beings. However, the historical record attests that an untamed environment has repeatedly wrought massive human degradation through catastrophic effects of famines, plagues, floods, tornadoes, earthquakes, etc. The fundamental problem, therefore, is not to maintain some simplistic "non-degradation" of the environment. Rather the problem is a complex one of devising appropriate means to protect both life-sustaining and aesthetic qualities of the biosphere, and at the same time develop technologies which provide basic human goods as a necessary condition for maintaining a preferred environmental quality. As a fundamental, meaningful principle for securing that environmental protection, "non-degradation" is vacuous.

As for preoccupation with risks to future generations and their proposed principle of neutrality, the NRDC authors seem committed to perpetuating a politically powerful, yet no less fraudulent myth -- namely, that the hazards of radioactive wastes foist unprecedented risks onto unconsulted future generations because the index of their hazard to the future is measured by and equivalent to the longevity of their radioactive half-life. This is absurd. Intellectual honesty should compel those who know better to state as often as necessary that any risks of adverse health effects from radiation sources -- both to present and future generations -- must be measured only in relation to environmental pathways which determine the degree of likelihood of harmful exposure of and assimilation by the human body.

Perhaps time alone can domesticate the exaggerated fears which surround radiation in general, and radioactive wastes in particular. In the meantime, our concern for the risks bequeathed to future generations will be better expressed if we reject outright two simple-minded notions: (a) first, that such risks have an existence in splendid isolation from the benefits which justify them, and (b) secondly, that such ethical principles as equity and participation require a neutral allocation of risks and benefits to the future.

The first notion merits rejection because the legacy of any generation to an immediate as well as remote future is not mere "risks" and "hazards," but to the contrary, an entire social order striving to provide material well-being, institutional stability, and creative freedom for its citizenry. Risks and promises, harms and benefits are inseparably interdependent within any sustainable social order.

As for the second notion, the ethical principle of equity requires a society to provide its citizens with reliable access to those basic goods which sustain material well-being. The principle of participation requires a society to provide institutionalized methods of consent for its citizens, who in turn are obligated to contribute to and abide by outcomes of those methods. It is nonsense for anyone to arrogate to themselves the wisdom either to decide for future generations what is in their best interest in securing basic goods and protection from basic harms, or to suppress -- under the guise of "neutrality to the future" -- any method of devising conceptual tools which might enable the present generation to deal constructively with its uncertainties and responsibilities toward the living. Ethical responsibility is primarily for the living who happen to be the only foundation we have to provide for the well-being of future generations.

From a bioethical perspective, there is ample justification for policy makers in the present generation to establish criteria and standards for health protection by reference to naturally occurring radiation sources from which man-made applications are derived. But it is not justifiable on the basis of a pseudo-philosophy of nondegradation or trusteeship over some pre-existing "natural state."

Those responsible in society for providing basic goods, methods of informed consent, and an equitable management of biohazards have an ethical obligation to derive value judgments of safety, acceptable risk, and justifiable harm from a philosophy of congruence with a pattern of benefits and harms already established by naturally occurring radiation sources with which human beings have lived and evolved throughout recorded history. That is to say, the philosophy of congruence and of logical consistency require a policy maker to form value judgments on the relative benefits of providing protection against radiation by first taking account of wide variations in personal exposures and population exposure from naturally occurring background sources.

External sources of exposure include cosmic rays, together with the radionuclides they produce, and primordial radionuclides in the earth. Internal exposure from natural radionuclides inhaled or ingested via food and drinking water augment that external exposure appreciably (e.g. potassium-40 adds 17 mrem.) Large segments of the population in the United States receive natural external radiation doses varying from 41 to 105 mrem per year simply because of geographic location. Variations in natural exposure to thorium in monazite sands along the southeastern coast of India range from 130 mrem to 2,800 mrem; while on the coast of Brazil, exposure ranges from 90 to 2,800 mrem with an average of 550 mrem per year. There is no scientifically established evidence, despite contrived attempts to prove it, that there are basic harms to those so exposed.

Human tolerance for, indeed dependence upon, such wide variations in natural radiation sources for several millenia demonstrate that increments from man-made applications of those natural sources can be kept well within the range of those variations without inflicting unjustifiable harm or deprivation of basic goods.

I am fully aware that this conclusion is contrary to what has been assumed by regulatory agencies when they have set excessively conservative standards in the past. With our increased knowledge of the benefits and harms of natural radiation, however, there is ethical justification for their gradual revision.

It is a matter of fact that the largest increment from man-made radiation exposure comes from medical and dental health practices, and these exposures are 10 to 100 times greater than other man-made sources which by contrast are stringently regulated.

From the perspective of bioethics, the inequitable management of biohazards in general, and of radiation protection in particular, has become a matter for national embarrassment. There is clearly a category of negligible risk and negligible harm which in practice ought to be ignored. This category conincides with the ethical principle of justifiable harm.

An application of the philosophy of congruence and of negligible harm is already a part of the public record in testimony submitted to the Nuclear Regulatory Commission in the matter of Perkins Nuclear Station, North Carolina.[13] In public testimony, comparisons have been made between radon releases which result from mining and milling of uranium, with radon naturally released from the earth. According to the record, Dr. R. L. Gotchy "provided calculations out to 10,000 years of the comparative population exposure resulting from radon emanation from the nuclear fuel cycle compared to the naturally occurring exposures. These calculations show that exposures due to radon releases from mining and milling are insignificant compared to natural background radiation exposures." Although agreeing with Dr. Gotchy's estimates based upon the data he had used, Dr. Hamilton "decried extrapolations of health effects into the distant future as being misleading" and not truly meaningful, because they do not take any account of repair mechanisms. When questioned by the NRC Board, Dr. Hamilton testified that:

> . . . variations in normal living style, traveling about the country and going indoors or outdoors result in doses that are many orders of magnitude greater than the increase in dose resulting from radon-222 emanating from tailings and mining.

He concluded that low levels of radiation such as these are "completely insignificant and without any reality."

The NRC Board concluded its findings by stating that the best mechanism available to characterize the significance of increments released by mining and milling is:

> to compare such releases with those associated with natural background. The increase in background associated with Perkins is so small compared with background, and so small in comparison with the fluctuations in background, as to be completely undetectable. Under such a circumstance the impact cannot be significant.

IV. CONCLUSION: Suggested Bioethical Principles for Setting Criteria and Standards for Radiation Health Protection

In view of the above reflections, I suggest that the following principles might better serve as guidance in the formulation of social policies for radiation health protection:

(1) Any involuntary risks imposed by social policies for radiation protection must be congruent with, must not be in excess of, and may be reasonably less than, those involuntary risks imposed by the wide variations in naturally occurring toxic elements and harmful effects from our natural environment.

(2) There must be evidence that basic goods and essential benefits cannot be more satisfactorily obtained through other alternative means which entail fewer risks.

(3) The basic goods and essential benefits must be demonstrated or virtually assured, and they should outweigh the possibility of basic harm to human well-being.

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SESSION F

ENVIRONMENTAL - PUBLIC HEALTH ASPECTS II

Session Chairperson

R. M. Fry

State of Kentucky

A REVIEW OF ENVIRONMENTAL SURVEILLANCE DATA AROUND LOW-LEVEL WASTE DISPOSAL AREAS AT OAK RIDGE NATIONAL LABORATORY

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ABSTRACT

White Oak Creek and Melton Branch tributary surface streams flow through the Oak Ridge National Laboratory (ORNL) reservation and receive treated low-level radioactive liquid waste which originates from various Laboratory operations. The streams receive additional low-level liquid waste generated by seepage of radioactive materials from solid-waste burial grounds, hydrofracture sites, and intermediate-level liquid-waste sites. Over the years, various liquid-waste treatment and disposal processes have been employed at ORNL; some of these processes have included: settling basins, impoundment, storage tanks, evaporation, ground disposal in trenches and pits, and hydrofracture. Burial of solid radioactive waste was initiated in the early 1940's, and there are six burial grounds at ORNL with two currently in use. Monitoring at White Oak Dam, the last liquid control point for the Laboratory, was started in the late 1940's and is continuing. Presently, a network of five environmental monitoring stations is in operation to monitor the radionuclide content of surface waters in the White Oak watershed. In this paper, the solid waste burial grounds will be described in detail, and the environmental data tabulated over the past 29 years will be presented. The various monitoring systems used during the years will also be reviewed. The liquid effluent discharge trends at ORNL from the radioactive waste operations will be discussed.

INTRODUCTION

Six solid radioactive waste disposal areas (SRWDA) have been used since the operation of Oak Ridge National Laboratory (ORNL) began in 1943. The location of these SRWDA's are shown in Figure 1. Location of the first three SRWDA's were selected primarily for convenience (We 76). Very little geologic or hydrologic considerations were given to the site selections.

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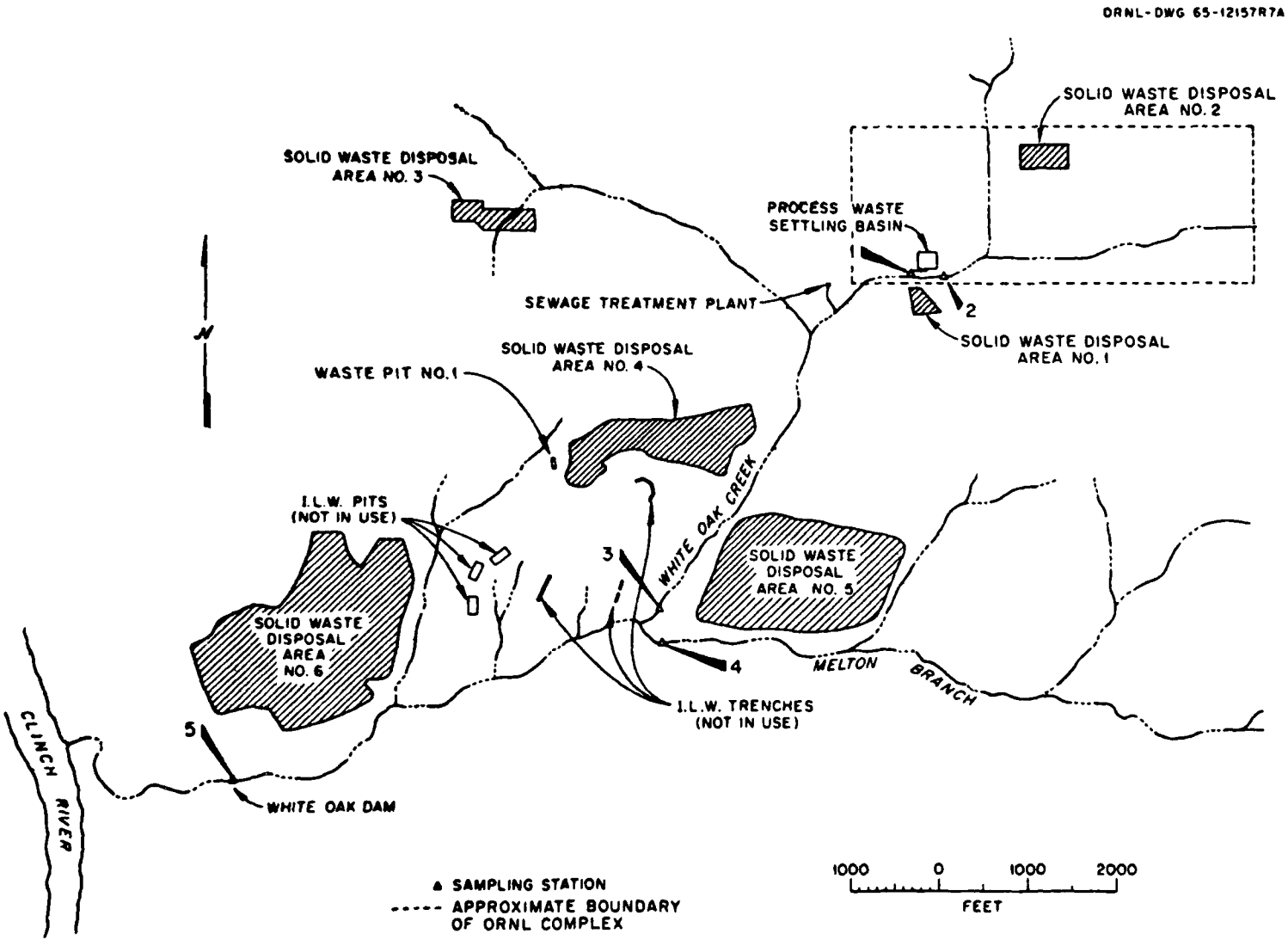
* Operated by Union Carbide Corporation under contract W-7405-eng-26 with the U.S. Department of Energy.


ORNL-DWG 65-12157R7A
SOLID WASTE DISPOSAL AREA NO. 2
SOLID WASTE DISPOSAL AREA NO. 3
PROCESS WASTE SETTLING BASIN
SEWAGE TREATMENT PLANT
2
SOLID WASTE DISPOSAL AREA NO. 1
N
SOLID WASTE DISPOSAL AREA NO. 4
WASTE PIT NO. 1
1
I.L.W. PITS (NOT IN USE)
WHITE OAK CREEK
3
SOLID WASTE DISPOSAL AREA NO. 5
SOLID WASTE DISPOSAL AREA NO. 6
4
MELTON BRANCH
I.L.W. TRENCHES (NOT IN USE)
5
WHITE OAK DAM
CLINCH RIVER
▲ SAMPLING STATION
APPROXIMATE BOUNDARY OF ORNL COMPLEX
1000 0 1000 2000
FEET

As the volume of waste increased, more attention was given to site selection. Areas underlain by Conasauga shale formation make excellent sites for waste disposal, as the shale is easily excavated and has ion exchange properties that inhibit the migration of water-soluble nuclides through the soil. Melton Valley is underlain by this formation and is the location of three of the SRWDA's that became operational since 1951. The current operational status and land area of the solid-waste areas are given in Table 1. Other sources of contamination on the site include settling basins, impoundments, trenches, and pits.

Table 1. Operational status of ORNL Radioactive Solid Waste Storage Area

SRWDA	Operating Dates	Status	Land Used (acres)*
1	1943 - 1944	Closed	1
2	1944 - 1946	Closed	4
3	1946 - 1951	Closed	7
4	1951 - 1959	Closed	23
5	1959	Operating	33
6	1969	Operating	68

*One acre = 4047 m^2.

SOURCES OF CONTAMINATION

SRWDA No. 1

SRWDA No. 1, with a total area of one acre, is located at the foot of Haw Ridge. It is at the edge of the Laboratory complex and is about 25 ft south of White Oak Creek (We 76). This site was selected on the basis of its proximity to the Laboratory, and no consideration of waste leaching into the water system was given. Waste was dumped into open trenches and backfilled. There are no available records showing the quantity or kind of solid waste disposed of in these areas. Very little monitoring data is available around SRWDA No. 1 (We 76). The date of closure of this area was 1944; it was closed because water was found in one of the trenches.

In 1946, the site was surveyed for surface contamination, and soil samples were analyzed. The results from only two areas indicated activity above background. Water samples from two wells and a surface seep in this area were analyzed for ^{90}Sr, ^{137}Cs, and transuranic elements in 1975. Low concentrations of ^{90}Sr (0.4 dpm/mℓ) were present in one of the wells. No detectable quantities of ^{137}Cs and transuranic elements were found (Du 75).

SRWDA No. 2

SRWDA No. 2 was operated between 1943-1946, and covered a total area of about four acres. The site is located on the lower half of a hill near

the east entrance of the Laboratory. Selection of the site was most likely based on consideration for the reduction of personnel exposure during the transportation of the waste (We 76). No attention was given t environmental protection.

There are no available records documenting the quantity or kind of solid waste disposed of in this area. It has been ascertained that solid waste contaminated by beta or gamma activity was placed in black iron dru and buried in the trenches. Liquid waste contaminated by plutonium was placed in stainless steel drums and either buried in trenches or stored without burial in a "natural ravine" eroded in the denuded slope (We 76).

The use of the SRWDA No. 2 site was later found to be incompatible with the long-range land-use planning at the Laboratory, and the operatio was terminated in 1946. After closure, most of the waste is said to have been exhumed and reburied in SRWDA No. 3. The stainless steel drums containing liquid plutonium waste were removed intact, but the black iron drums containing beta-gamma solid waste had deteriorated. Thus, the surrounding earth was also removed and reburied at SRWDA No. 3. The hillsid of the SRWDA No. 2 site was then bulldozed to smooth out the irregulariti and was seeded (We 76).

During August, 1976, 13 core samples were collected at various point in SRWDA No. 2. Water samples were also collected from the core holes. Activity levels in water samples were found not to be significantly diffe from baseline samples when analyzed for ^{3}H, gross-alpha, and gross-beta activity. A representative portion of the homogenized whole core was use for this analysis. The average uranium and plutonium concentrations were found to be 0.47 pCi/g and 0.06 pCi/g, respectively (Oa 77). The average radioisotope concentration for soil samples near the perimeter of the DOE area in Oak Ridge have been found to contain 0.66 pCi/g of uranium and 0. pCi/g of plutonium (Oa 76).

The average ^{137}Cs concentration for the upper third and the entire core was measured to be 0.7 and 0.3 pCi/g, respectively. Both of these values are substantially lower than the value of 1.0 pCi/g, the average value of samples collected in 1976 from 16 sites throughout eastern and central Tennessee (Oa 76). It should be noted that these soil samples a from cores several feet long and are being compared with topsoil samples. For ^{90}Sr, the average values for the cores were $\leq$ 0.57 and $\leq$ 0.53 pCi/g for the upper third and the entire core, respectively (Oa 76).

SRWDA No. 3

SRWDA No. 3 is about 0.6 miles west of the west entrance to the Labc complex. The site is a flat, forested area at the foot of Haw Ridge. Waste Area No. 3 presumably was chosen because of its proximity to the Laboratory, out-of-sight location, and because the soil could be readily excavated (We 76). The area became operational in 1946. Alpha-contamina wastes were dumped in unlined trenches and covered with concrete, whereas the beta-gamma waste was covered with native soil.

Samples of well water from the area were analyzed in 1964 and indicated small amounts of the trivalent rare earths (TRE), ^{90}Sr, ^{89}Sr, and ^{3}H (We 76) Well samples were also collected in 1973 and analyses indicated ^{90}Sr levels as high as 3.0 dpm/mℓ. Soil samples were collected and analyzed during 1978 and the results are given in Eldridge (79).

SRWDA No. 4

During 1948-50, a study (St 51) of the geology and hydrology of the Laboratory site was conducted. Disposal of waste in the Conasauga shale belt was recommended. SRWDA No. 4 was opened in 1951 in the closest area to the Laboratory underlain by Conasauga shale. Trench orientation was variable and lacked any consistent relationship to original site topography (We 76). Auger holes one to two feet in diameter in this area were used for the disposal of higher radioactive level waste. The site was closed in 1959 and resulted in a total disposal area of 23 acres.

A number of small seeps have developed near the rim of the terrace in the center third of the area, and others are reported to have developed in the central part of the site. During 1959 and 1960, sampling of wells and streams in and near this area indicated that both ground water and surface water were contaminated (We 76). Eight of the sixteen wells showed beta-gamma contamination. Water samples from two seeps indicated contamination of ^{90}Sr, ^{137}Cs, ^{95}Zr-^{95}Nb, ^{60}Co, and TRE. The section of White Oak Creek flowing by SRWDA No. 4 indicated radioactive contaminants of ^{106}Ru, ^{90}Sr, ^{210}Po, ^{239}Pu, and TRE (We 76). In 1964, water samples were collected from six wells and one seep and each were found to contain $^{89,90}Sr$, ^{3}H, TRE, and minor amounts of ^{106}Ru (We 76). Discharges of ^{90}Sr from SRWDA No. 4 versus precipitation are given in Table 2 from Stueber (78).

Table 2. ^{90}Sr discharges vs. precipitation

Water Year*	Precipitation cm	Total ^{90}Sr Discharge Ci
1967	154	2.7
1968	114	2.0
1969	102	2.1
1970	122	1.6
1971	123	1.2
1972	120	2.4
1973	181	1.6
1974	175	5.2
1975	147	3.2
1976	124	5.1
1977	129	2.3

*September 1 - August 31

Soil samples were collected in 1973 along the south side of SRWDA Nc These samples contained small amounts of ^{60}Co, ^{137}Cs, and ^{90}Sr (Du 76). soil along White Oak Creek east of the area has been contaminated by seep from SRWDA No. 4 and discharges of the creek. Near this site is a contam nated flood plain area which was once flooded by an intermediate pond. A was constructed in early 1944 to help create an intermediate retention pc between the Laboratory and White Oak Lake. The dam was breached in late 1944, and a small pond remained until 1950 (Du 76). Results of analyses soil and sediment are given in the section on flood plains.

SRWDA No. 5

SRWDA No. 5 opened in 1958 and consisted of two sections on the hill east of White Oak Creek and south of Haw Ridge. This area was opened in 1958, because space in SRWDA No. 4 was approaching exhaustion. Initially, same burial procedures were used at this site as had been used at the pre ceding sites; that is alpha-contaminated waste was interred in the lower of the area and capped with concrete, and the beta-gamma contaminated was was simply covered with weathered shale. This segregation procedure was discontinued sometime during the operational life of the site. The trenc lengths vary from < 40 feet to > 500 feet. These trenches were oriented parallel to the topographic slopes (We 76). Water samples from several w were collected in 1964. The principal contaminants found were ^{90}Sr, ^{89}Sr ^{106}Ru, ^{3}H, and TRE. Several new wells were cored and sampled. The data suggested that at this time, only minor movement of radioactivity had occurred. In 1960, samples from these wells indicated that SRWDA No. 5 w the major source of ^{3}H (We 76) in White Oak Creek.

Most of the transport of radionuclides in the surface water is monitored at Station 4 on Melton Branch. Additional data are given in th section on monitoring stations. In 1974, thirteen small seeps were sampl along the south edge of the area. These samples contained measurable amo of total alpha, ^{90}Sr, ^{3}H, and ^{125}Sb. Eleven of the samples contained con trations of ^{90}Sr ranging from 9×10^{-8} to 6.1×10^{-5} µCi/mℓ (Du 76).

SRWDA No. 6

SRWDA No. 6 is located immediately northwest of White Oak Lake. This site is about 70 acres in size and was opened in 1969. Trenches initiall were excavated as long as possible, but are now limited to a length about feet. This procedure was initiated in order to reduce the collection of water in the trenches to an acceptable level (We 76). Some monitoring around this area has been completed. The results indicate some movement radioactivity, but it is too early to judge the true meaning of the resul

Floodplain Areas

There are four floodplain areas on the site that are contaminated. For the purpose of this paper, only the floodplain that was established b the construction of the dam in 1944 will be discussed.

During 1944, a study of the ^{137}Cs distribution in soil, roots, ground vegetation, overstory, litter, mammals, flies, and insects from the 2 ha (20,000 m^2) flood plain area was made (Vo 76). The highest concentration found in soil was 84,400 pCi/g. All ^{137}Cs concentrations below the 17-cm depth, outside the floodplain area were equal to background (Vo 76). Concentrations in the roots ranged as high as 12,500 pCi/g. The concentration of ^{137}Cs in ground vegetation ranged from 4.6 to 182 pCi/g. Other results can be found in Voris (76).

Waste Ponds

Another source of radioactive discharges from past waste treatment procedures are the waste ponds. Three waste ponds have been in use at ORNL over the years of operation. An example of the activity in these ponds is from a study in 1977 (Ta 77) of Waste Pond 2. This study indicated approximately 5 Ci of $^{237,240}Pu$, 200 Ci of ^{137}Cs and 33 Ci of ^{90}Sr in the bottom sediment. Another source of discharged waste is the Intermediate-Level Waste Pits.

Seepage Pits and Trenches

In 1951, the construction of pits for disposal of intermediate-level liquid was begun. The first pit was opened in 1951 and immediately closed because of its poor location. The second pit was opened in 1952, and large quantities of intermediate-level waste were disposed of for the first time. Pits 3 and 4 became operational in 1955 and 1956, respectively. Trenches 5, 6, and 7 were opened between 1960-1962. These trenches were taken out of routine service in 1965 as part of a plan to implement disposal of intermediate-level waste by hydrofracture (Du 75).

Small amounts of ^{90}Sr and ^{137}Cs have been observed in seepage from trenches 6 and 7. The major seepage problem has been with ^{106}Ru, as seen in Table 3 and Figure 2.

Table 3. Annual discharges of ^{106}Ru to the Clinch River

Year	Ci	Year	Ci
1959	520	1965	69
1960	1900	1966	29
1961	2000	1967	17
1962	1400	1968	5
1963	430	1969	1.7
1964	191	1970	1.2

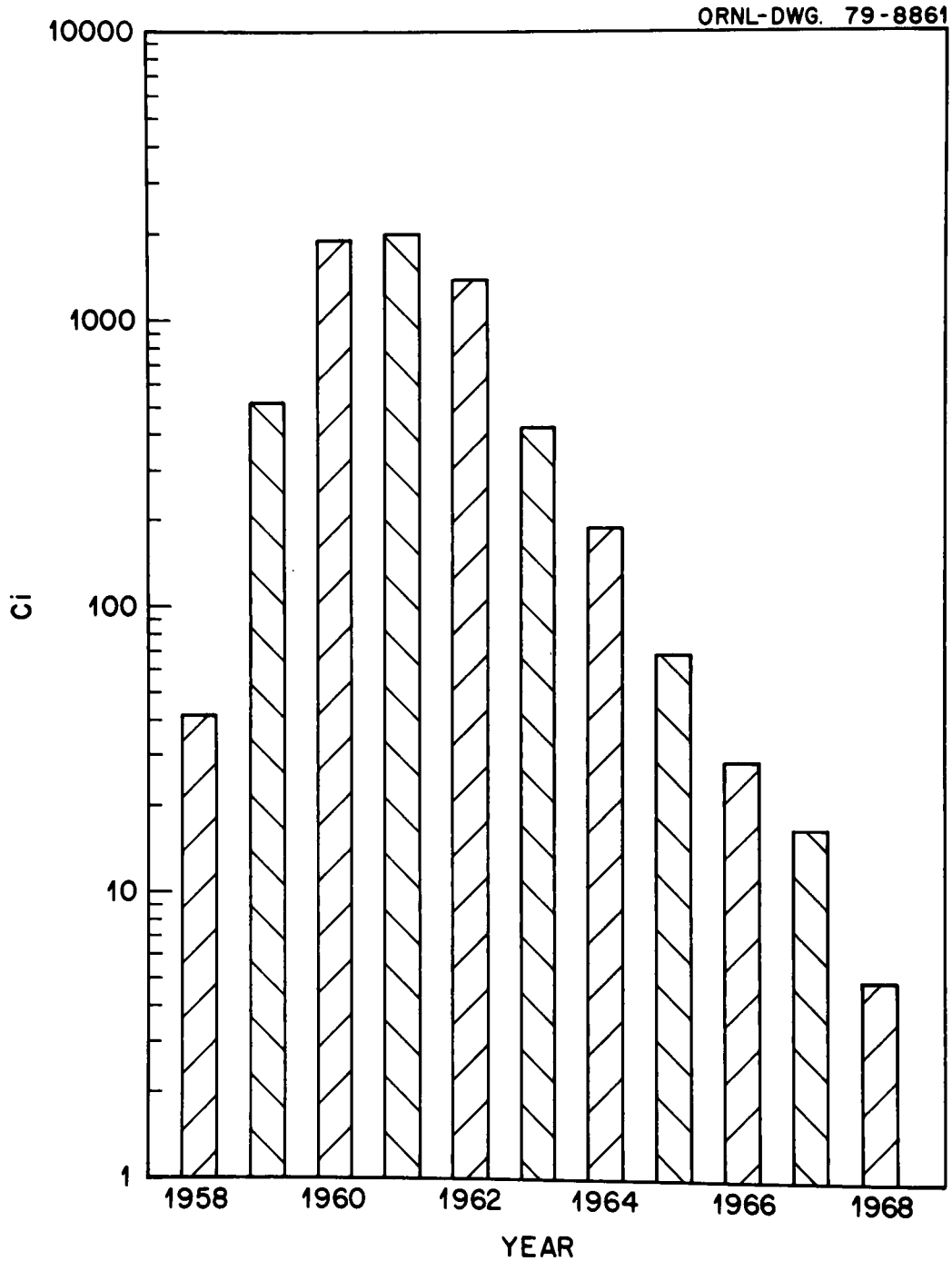


Fig. 2. Annual discharges of ^{106}Ru to the Clinch River.

WATER SAMPLING

Low-level radioactive liquid wastes originating from ORNL operations are discharged, after preliminary treatment, to White Oak Creek and Melton Branch, which are small tributaries of the Clinch River. The radioactive content of the White Oak Creek discharge is determined at White Oak Creek Stations 1, 2, and 3 and at White Oak Dam, which is the last control point along the stream prior to the entry of White Oak Creek into the Clinch River. Samples are collected at several locations in the Clinch River, beginning at a point above the entry of the wastes into the River and ending at Center's Ferry near Kingston, Tennessee, the nearest population center downstream. Location maps of these stations are given in Figures 1 and 3.

Station 1 - White Oak Creek Station 1 monitors the effluent from the ORNL Process Waste Treatment Plant.

Station 2 - Station 2 is located on White Oak Creek a short distance upstream from Station 1 and provides data on radionuclide content from operation discharges above the Process Waste Treatment Plant.

Station 3 Water Monitoring Station 3 is located on White Oak Creek a short distance above the confluence of White Oak Creek and Melton Branch. This station measures the streamflow and radionuclides content from the ORNL plant effluents; SRWDA Nos. 1, 2, 3, 4, and portions of 5; contaminated floodplain sediments; and other potential sources.

Station 4 Station 4 is located on Melton Branch, a short distance above the confluence with White Oak Creek. This station measures streamflow and radionuclide content from SRWDA No. 5, several experimental reactor sites, and other areas.

Station 5 - Samples of White Oak Creek effluent are collected at White Oak Dam by a continuous proportional sampler which was designed and constructed at ORNL. Proportional sampling is necessary to obtain a truly representative sample, since streamflow and concentration of radioactive materials in the stream may vary independently over a relatively wide range in a relatively short period of time, depending upon weather and operating conditions. Streamflow at White Oak Dam is measured by means of a Stevens water-level recorder and stilling well in the lake pool in conjunction with the White Oak Dam gate, which serves as a rectangular weir through which the water flows. The weir system was rated by the U.S. Geological Survey and found to be accurate to within 5%.

Samples are collected weekly from White Oak Dam and analyzed for gross beta activity as a control measure and as a means of evaluating the gross concentration of radioactivity entering the Clinch River. Portions of the weekly samples are composited, proportional to the flow, into monthly composite samples that are subjected to more detailed analyses by wet chemical and gamma spectrometric techniques. The weekly samples are

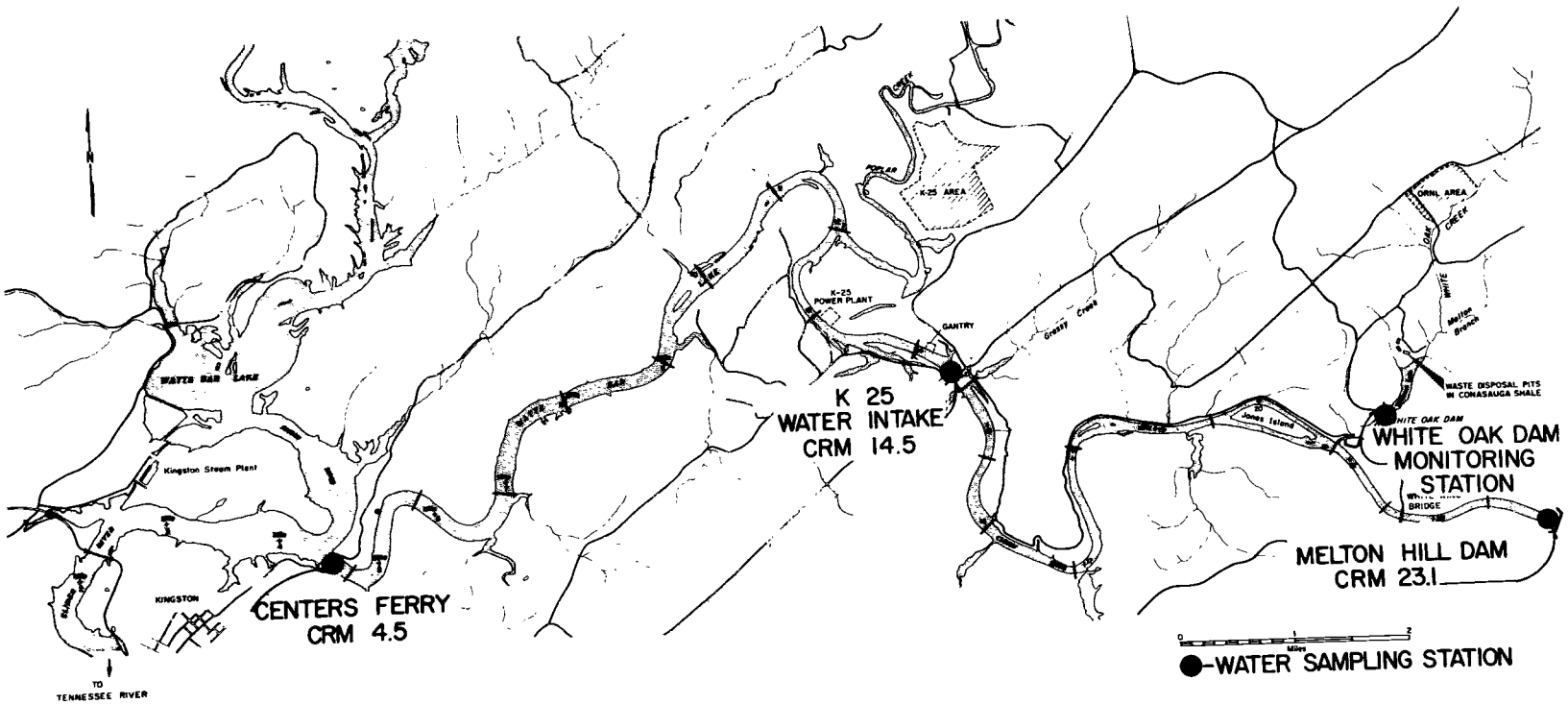


Fig. 3. Water monitoring locations in the Clinch River.

analyzed for the transuranic alpha emitters, total strontium, and ^{131}I, which represent the elements in the waste stream with the highest hazard indices.

The monthly composites are concentrated and analyzed by radiochemical and gamma spectrometric techniques, normally for the following radionuclides: ^{90}Sr, ^{137}Cs, ^{144}Ce, ^{106}Ru, ^{95}Zr-^{95}Nb, ^{60}Co, ^{3}H, trivalent rare earths, and gross beta. Analyses for other nuclides may be performed as the need arises. These analyses are performed to determine the percentage distribution and concentrations of the various nuclides in the effluent stream and to calculate the quantity of each radionuclide released to the Clinch River. More frequent analyses are made if concentration levels in White Oak Creek vary significantly from the experienced normal.

Calculations are made of the concentrations of radioactivity in the Clinch River for the point of entry of the wastes, using the concentrations measured at White Oak Dam and the dilution provided by the River. These calculations are based on uniform mixing of the two streams within a short distance downstream from the point of entry of the wastes. The calculated concentration of each radionuclide in the River is compared with its respect MPC_w value as specified by Chapter 0524 of the DOE Manual and the resulting fractions are summed to arrive at the % MPC_w in the Clinch River.

The annual discharges of radionuclides to the Clinch River as measured at White Oak Dam from 1949-1978 are given in Tables 4 and 5. The measured MPC_w at White Oak Dam and the calculated MPC_w in the Clinch River from 1974-1978 are given in Figures 4 and 5, respectively. A comparison of MPC_w at White Oak Dam and the calculated value in the River from 1974-1978 is given in Table 6. The total amount of radionuclides discharged into the Clinch River is given in Table 7. The amount of ^{3}H released to the river in three-year periods from 1965-1977 is given in Figure 6.

As a followup to this calculated concentration, two sampling stations are maintained in the Clinch River below the point of entry of the wastes: one at the ORGDP water intake, Clinch River mile (CRM) 14.5, and the other at Center's Ferry near Kingston, Tennessee (CRM 4.5). In addition, a sampling station is maintained in the Clinch River above the point of entry of the waste at Melton Hill Dam (CRM 23.1) to provide background data.

ORGDP Water Intake Sampling Station - The ORGDP water sampling station, which was designed and constructed at ORNL, collects a sample from the Clinch River that is proportional to the flow in the River near the water intake of the ORGDP plant water system (the first point of Clinch River water usage downstream from the point of entry of ORNL wastes). The samples are brought into the laboratory at weekly intervals, acidified, and combined into quarterly composite samples for analysis. The quarterly composite samples are concentrated by evaporation and analyzed by wet chemical and gamma spectrometric techniques for gross activity and for those radionuclides present in significant amounts. Average concentrations of individual radionuclides are determined from the analytical data,

Table 4. Annual discharges (Ci) of radionuclides to the Clinch River 1949-1963

Year	^{137}Cs	^{106}Ru	^{89}Sr	^{90}Sr	TRE*(-Ce)	^{144}Ce	^{95}Zr	^{95}Nb	^{131}I	^{60}Co	^{3}H	TRU
1949	77	110		150	77	18	180	22	77		NA**	0.009 (for
1950	19	23		38	30	NA	15	42	19			0.0379
1951	20	18		29	11	NA	4.5	2.2	18			0.0734
1952	9.9	15		72	26	23	19	18	20			0.0296
1953	6.4	26		130	110	6.7	7.6	3.6	2.1			0.0838
1954	22	11		140	160	24	14	9.2	3.5	NA		0.0729
1955	63	31		93	150	85	5.2	5.7	7.0	6.6		0.2491
1956	170	29		100	140	59	12	15	3.5	46		0.2840
1957	89	60		83	110	13	23	7.1	1.2	4.8		0.1454
1958	55	42	NA	150	240	30	6.0	6.0	8.2	8.7		0.0755
1959	76	520	0.28	60	94	48	27	30	0.5	77		0.6770
1960	31	1900	1.9	28	48	27	38	45	5.3	72		0.1860
1961	15	2000	2.0	22	24	4.2	20	70	3.7	31		0.0682
1962	5.6	1400	1.7	9.4	11	1.2	2.2	7.7	0.36	14		0.0627
1963	3.5	430	0.98	7.8	9.4	1.5	0.34	0.71	0.44	14		0.1660

*Total rare earths minus cerium.
**No analysis performed.

Table 5. Annual discharges (Ci) of radionuclides to the Clinch River 1964-1978

Year	^{137}Cs	^{106}Ru	^{89}Sr	^{90}Sr	TRE*(-Ce)	^{144}Ce	^{95}Zr	^{95}Nb	^{131}I	^{60}Co	^{3}H	TRU
1964	6.0	191	0.79	6.6	13	0.3	0.16	0.07	0.29	15	1929	0.0775
1965	2.1	69	0.59	3.4	5.9	0.1	0.33	0.33	0.20	12	1161	0.4979
1966	1.6	29	0.85	3.0	4.9	0.1	0.67	0.67	0.24	7.4	3090	0.1599
1967	2.7	17	0.73	5.1	8.5	0.2	0.49	0.49	0.91	3.1	13273	1.0335
1968	1.1	5	0.55	2.8	4.4	0.03	0.27	0.27	0.31	1.2	9685	0.0440
1969	1.4	1.7	0.31	3.1	4.6	0.02	0.18	0.18	0.54	1.0	12247	0.1989
1970	2.0	1.2	0.27	3.9	4.7	0.06	0.02	0.02	0.32	1.0	9473	0.4003
1971	0.9	0.50	0.20	3.4	2.9	0.05	0.01	0.01	0.21	0.8	8945	0.0456
1972	1.7	0.52	NA**	6.05	5.2	0.03	0.01	0.01	0.34	1.3	10600	0.0687
1973	2.3	0.69		6.7	NA	0.02	0.05	0.05	0.46	1.1	15000	0.0759
1974	1.2	0.22		6.0		0.02	0.02	0.02	0.23	0.60	8633	0.02
1975	0.62	0.30		7.2		NA	NA	NA	0.28	0.50	11061	0.02
1976	0.24	0.16		4.5					0.03	0.86	7422	0.01
1977	0.21	0.20		2.7					0.03	0.44	6249	0.03
1978	0.27	0.21		2.0					0.04	0.36	6292	0.03

*Total rare earths minus cerium.
**No analysis performed.

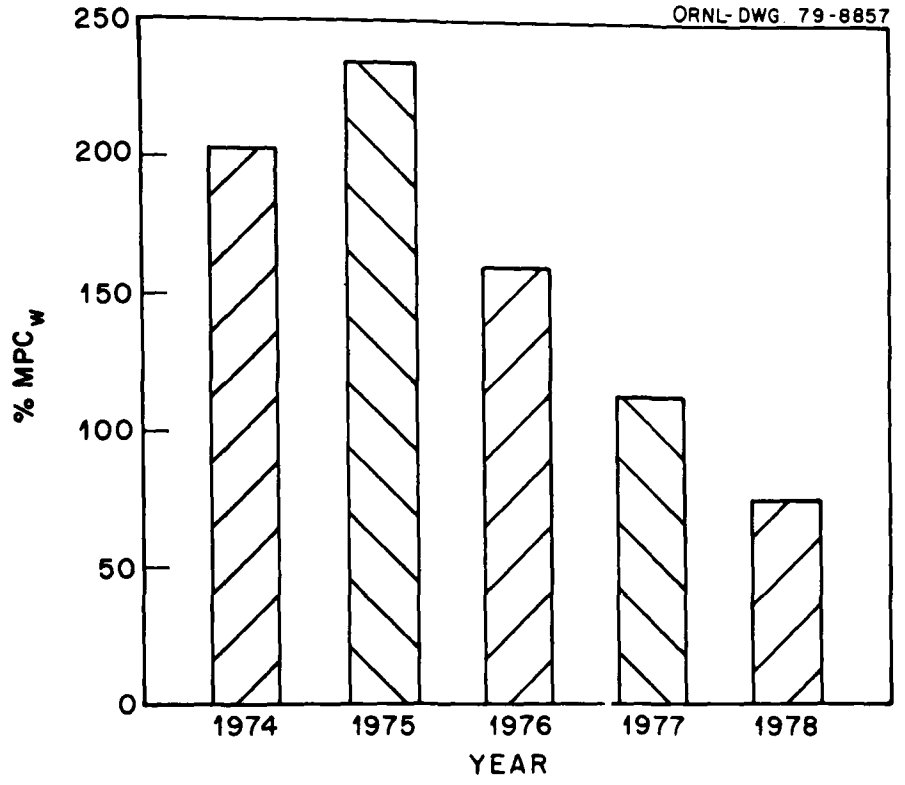


Fig. 4. Percent of MPC_w total over White Oak Dam.

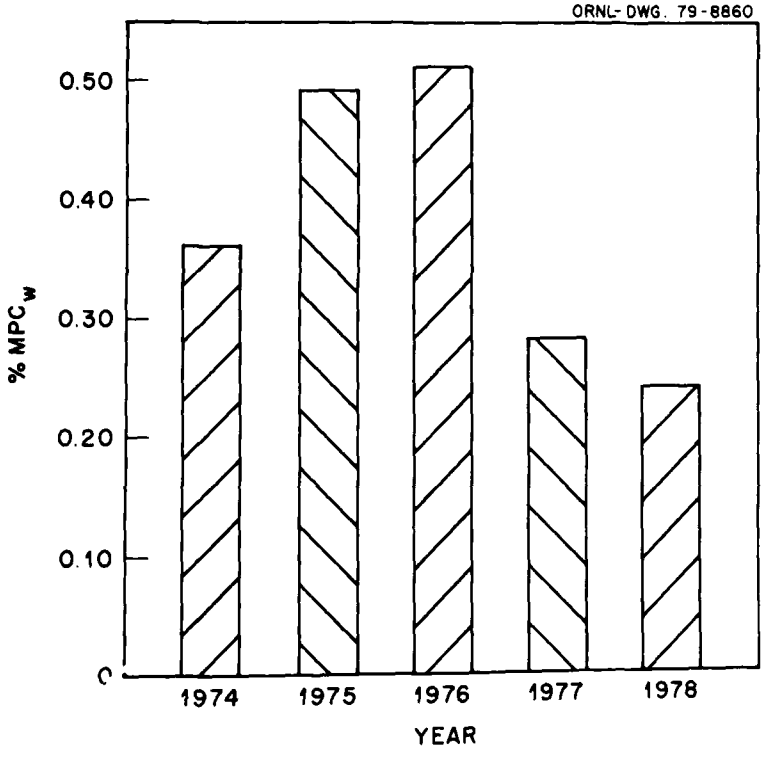


Fig. 5. Percent of MPC_w total for dilution calculation in the Clinch River.

Table 6. Total percent MPC_w for all radionuclides

1974-1978

Year	White Oak Dam	D.F.	Calc. C.R.*
1974	201%	688	0.29%
1975	234	646	0.36
1976	160	423	0.38
1977	112	537	0.21
1978	75	481	0.16

*Calculated concentrations in Clinch River using dilution factors.

Table 7. Total amount of radionuclides discharged into Clinch River

1949-1978

Time Interval	Radionuclide	Ci
1949-78	^{90}Sr	1,179
1949-78	^{95}Nb	286
1949-78	^{95}Zr	375
1949-78	^{106}Ru	6,932
1949-78	^{131}I	174
1949-78	^{137}Cs	687
1964-78	^{3}H	124,945
1955-78	^{60}Co	321
1949-74	^{144}Ce	342
1949-71	TRE(-Ce)*	1,289

*Trivalent rare earths minus cerium.

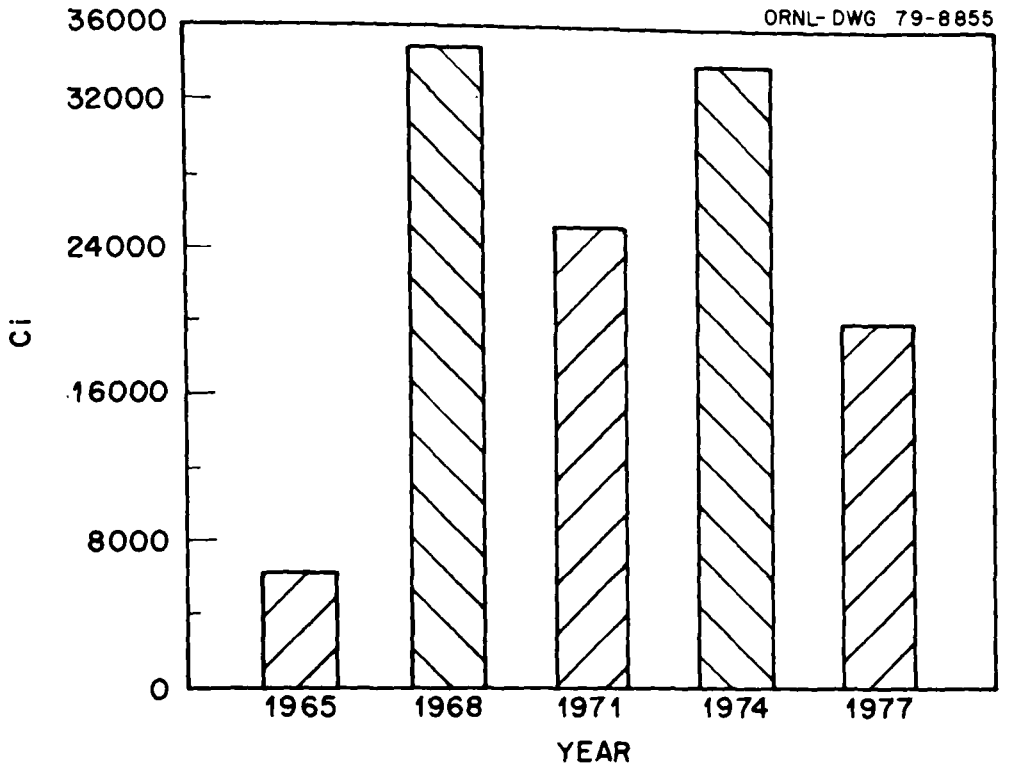


Fig. 6. Tritium releases to the Clinch River (3-yr. intervals).

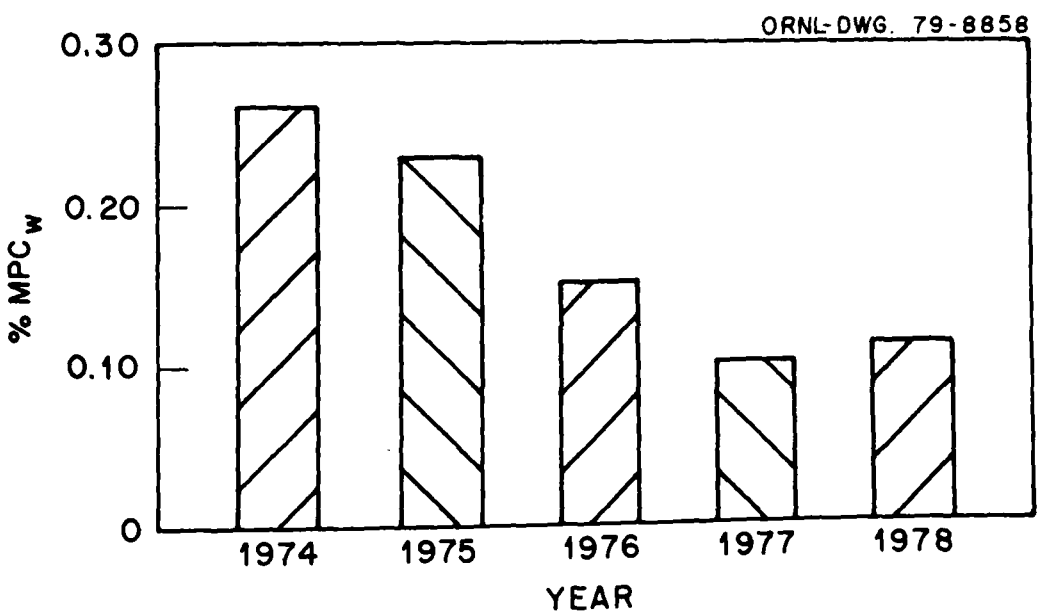


Fig. 7. Percent of MPC_w total at the ORGDP intake.

and the percent MPC_w for the mixture is calculated from the concentration values measured. The MPC_w at this point from 1974-1978 are given in Figure 7.

The ORGDP sampling station was established to provide data relative to the concentrations of radioactivity in water taken from the Clinch River for normal treatment plant usage, to provide an index of the hazard to the ORGDP plant population as the first user of Clinch River water downstream from the point of entry of the ORNL wastes, and to provide data for comparison with the concentrations in the Clinch River calculated from White Oak Dam releases and the dilution provided by the River.

Center's Ferry Sampling Station - A "grab" sample is collected daily at the Center's Ferry sampling station which is located on the Clinch River at CRM 4.5. Thermal stratification exists at this location, with the cold water of the Clinch River running under the warmer backwater of the Watts Bar Reservoir. The sample is collected 25 ft below the surface of the water to ensure the collection of Clinch River water for the sample. Fluctuations in concentration at this location are relatively small due to the distance downstream from the point of entry of the wastes, and dispersion of radioactivity in the Clinch River water is complete; thus a grab sample is considered adequate. The daily grab samples are composited, acidified, and analyzed on a quarterly basis. The preparation of the sample and the analyses performed are the same as those cited for the ORGDP water sampling station.

Samples from the Center's Ferry sampling station provide data relative to the average concentration of radioactive materials in the Clinch River at the nearest population center (Kingston, Tennessee) downstream from the point of entry of the wastes. The MPC_w from 1976-1978 is given in Figure 8. A comparison of the calculated MPC_w at ORGDP and Center's Ferry is given in Table 8.

Table 8. Total percent MPC_w i Clinch River

Year	ORGDP	C.F.*	Calc. C.R.**
1974	0.26%	0.21%	0.36%
1975	0.23	0.15	0.49
1976	0.15	0.15	0.51
1977	0.10	0.12	0.28
1978	0.11	0.12	0.24

* Center's Ferry

** Calculated concentrations in Clinch River using dilution factors

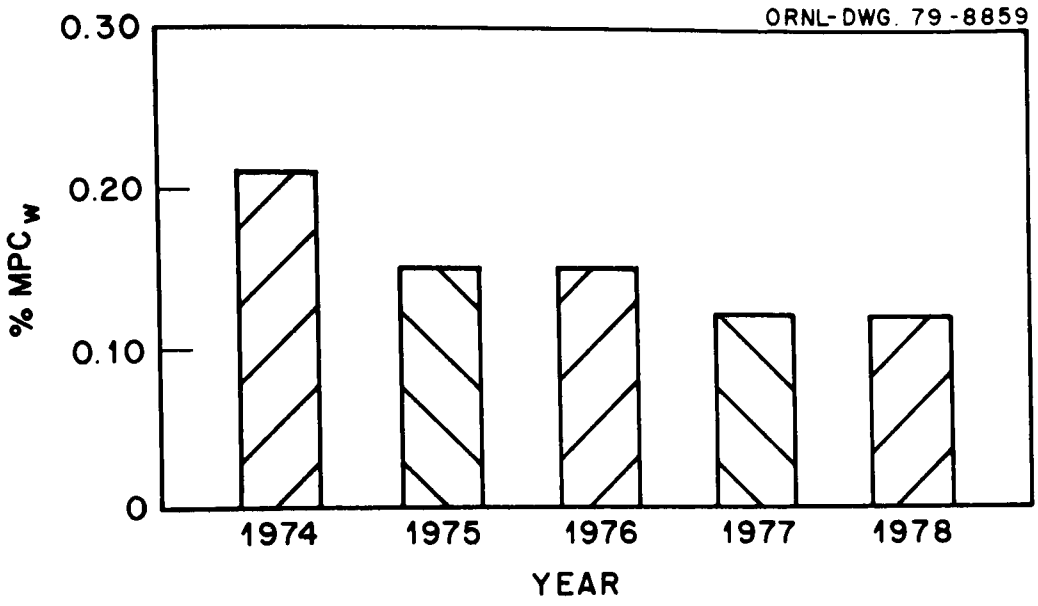


Fig. 8. Percent of MPC_w total at Kingston, Tennessee.

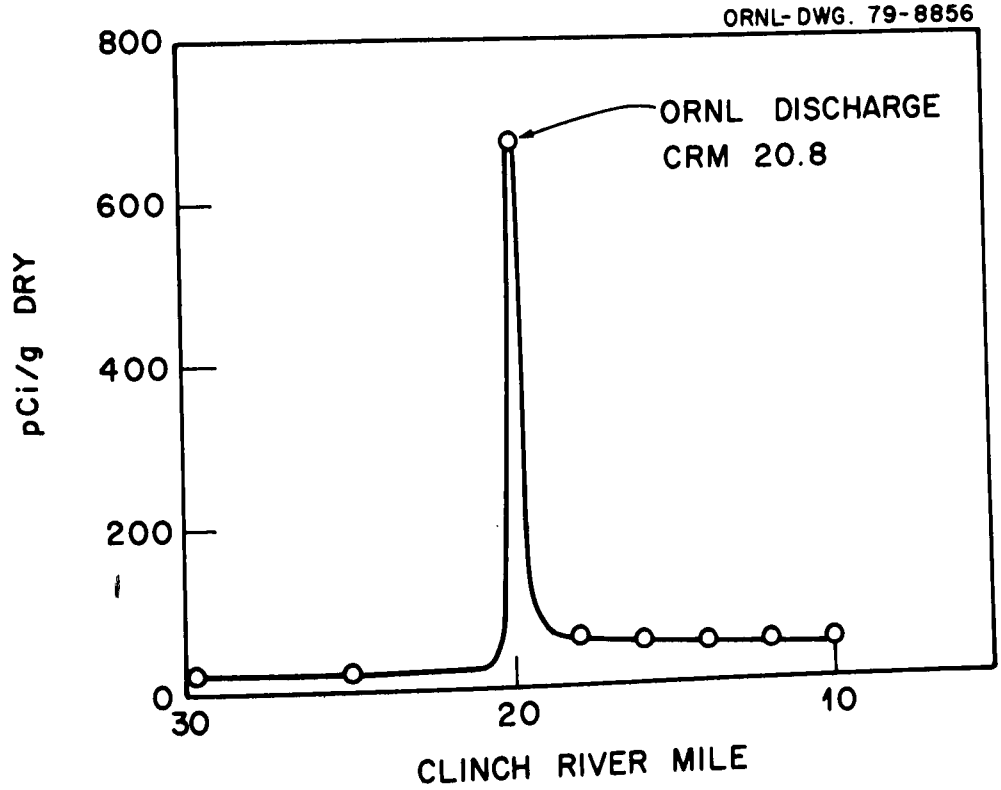


Fig. 9. Cesium-137 in river silt (1967).

Melton Hill Dam Sampling Station - The Melton Hill Dam sampling station collects a sample proportional to the flow of water through the power-generating turbine, which represents all of the discharge from the Dam other than a minor amount discharged in the operation of the lock. The sampler was designed and constructed at ORNL and is located on the lower side of the Dam, with the intake positioned at the tail race of the turbines. The sampler is keyed to the turbine operation such that a sample is collected only when the turbines are operating, even though water from the tail race is continuously pumped through the sampling lines. Samples are collected from the station at weekly intervals, acidified, and composited for a three-month period in polyethylene containers. The quarterly sample is processed and analyzed in the manner set forth (above) for the ORGDP water sampling station.

In addition to meeting routine monitoring requirements, water monitoring aids in the detection of abnormalities. For example, in 1969, high levels of ^{60}Co were detected in the water at Melton Hill (CRM 23.1). River silt was then analyzed and found to contain 16 pCi/g of ^{60}Co above Melton Hill. Concentration below ORNL discharges was 1.2 pCi/g (CRM 19.1). Investigation resulted in the finding of a waste tank leak at American Nuclear Corporation located at approximately CRM 51. Other types, of samples, such as sediment and fish, are also collected for the purpose of monitoring the discharges from the plant site.

Fish Sampling

Several species of fish from the Clinch River are sampled. Ten fish of each species are composited for each sample, and the samples are analyzed by gamma spectrometry and radiochemical techniques for the critical radionuclides that contribute to dose. An estimate of man's intake of radionuclides from eating the fish is made by assuming an annual rate of fish consumption of 37 lbs. The estimated percentage of MPI is calculated by assuming a maximum permissible intake of fish to be comparable to daily intake of 2.2ℓ of water containing the MPC_w of these radionuclides for a period of one year. The highest recorded MPI in the last 15 years for any species has been 8.1%.

Sediment Sampling

Annual average concentrations of radionuclides in the surface layers of bottom sediments in the Clinch and Tennessee Rivers were conducted from 1951-1969. This information is relevant to the dispersal and movement of radioactivity released from ORNL. Data varied from year to year with the changes in the amounts of radionuclides released. The principal radionuclide contained in the bottom sediments is ^{137}Cs with lesser amounts of ^{60}Co, ^{106}Ru, and ^{144}Ce being retained. Only a small amount of ^{90}Sr released to the river is incorporated in the bottom sediments. Water analyses have shown that nearly all of the ^{90}Sr is either in solution or associated principally with suspended solids and, thus, passes through the river system. Sediment data can indicate discharges as shown in Figure 9.

SUMMARY

I. Facts with regard to the releases from ORNL waste practices over the last twenty years.

A. Ruthenium Releases.

1. A large amount of ruthenium was released during 1959-64. This reflected the seepage of ^{106}Ru from the ILW waste pit disposal area following the transfer of a large quantity of material to the pit in 1959.

2. The Conasauga shale did not retain ruthenium as well as other radionuclides.

B. Tritium Releases.

1. Large quantities of tritium have been released to the Clinch River in recent years.

2. Origin of the bulk of the tritium is apparently the burial grounds where tritium-bearing waste from another laboratory was buried.

3. From a practical standpoint, little can be done to inhibit or control these releases.

C. There has been a downward trend in the number of curies released for all other radionuclides.

	Ci	
Radionuclides	1959	1978
^{60}Co	77	0.36
^{90}Sr	60	2.0
^{131}I	0.5	0.04
^{137}Cs	76	0.27

II. A number of corrective measures have been taken at ORNL to reduce the discharges. Some of these are:

A. The method of disposal of liquid waste into pits and trenches was discontinued in 1965, and the use of hydrofracture was begun in 1966.

B. Rerouting of White Oak Creek to bypass floodplain areas.

C. The installation of the new Process Waste Treatment Plant which became operational in April, 1976, has significantly reduced the ^{90}Sr discharges to White Oak Creek from facility operations.

D. Paved intercepter ditches have been placed in the burial ground area to reduce the surface runoff in that area.

E. Hydrological consideration is being reviewed in the selection of a new burial site.

F. Small trenches are being used to reduce the amount of water collected in them after disposal has occurred.

G. Volume reduction programs have been initiated.

Better monitoring programs are also being initiated. Other proposed corrective measures for the reduction of releases and proposed facilities for the disposal of low-level waste are being considered. These efforts have resulted in a reduction in the discharges from the site as has been shown in this paper.

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EVALUATION OF A DECOMMISSIONED RADWASTE POND

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Abstract - An eight hectare radwaste pond (216-S-17) which received cooling water effluent from a nuclear fuel reprocessing plant at Hanford from 1951 to 1954 was contaminated due to unplanned releases. Subsequently, it was decommissioned by covering the area with 45-60 cm of backfill. Soil erosion and nuisance contamination, in the form of tumbleweeds (Salsola kali), occurred between 1954 and 1972 due to the lack of a specific revegetation program. Siberian Wheatgrass (Agropyron sibericum) and Cereal Rye (Secale cereale) were planted in 1972 and allowed to grow under natural conditions. A routine evaluation of the site disclosed the presence of contaminated Siberian Wheatgrass plants. This report describes the results of a radioecological study of the site in 1978. Nondestructive methods developed for in situ evaluation to determine radionuclide inventories and transport parameters for biotic and abiotic compartments are presented concomitant with standard procedures. Results indicate that Siberian Wheatgrass is a suitable perennial for revegetation of low-level waste disposal sites in an arid environment.

INTRODUCTION

In October 1951, 216-S-17 Pond became operational by receiving process-vessel cooling water and steam condensate from the Redox nuclear fuel reprocessing plant (Fig. 1). The original pond site was formed by creating an earthen dike approximately one meter high on the north and west sides of the polygonal area indicated in Figure 2. The designated use area was approximately 23 hectares, but photographs indicate that only 8 hectares were inundated at any time. The pond averaged about 0.3 meters deep with a maximum depth of 0.6 meters.

S-17 Pond received effluent until April 1954 when it was deactivated due to unplanned releases of fission products and consequently radionuclide concentrations in the pond sediments "exceeded prescribed limits" . A determination was then made to plug the pipeline near the outlet and the pond was allowed to dry up. The contaminated pond area was reportedly covered with 45-60 cm of backfill to prevent the spread of the major contaminants, i.e.; ^{137}Cs, ^{90}Sr, and ^{238}U. Due to extensive wind erosion near the inlet at the northeast corner (Fig. 2), additional soil was added to a depth of approximately 105 cm above the contaminated sediment layer. In succeeding years, wind erosion and various other factors precluded the establishment of native vegetation in most of the pond area. However, introduced weeds such as tumbleweeds became well established in some areas and where erosion had reduced the backfill depth their roots penetrated the contaminated sediment layer. This resulted in the accumulation of "nuisance quantities" of ^{137}Cs and ^{90}Sr in plant tissues (C177) .

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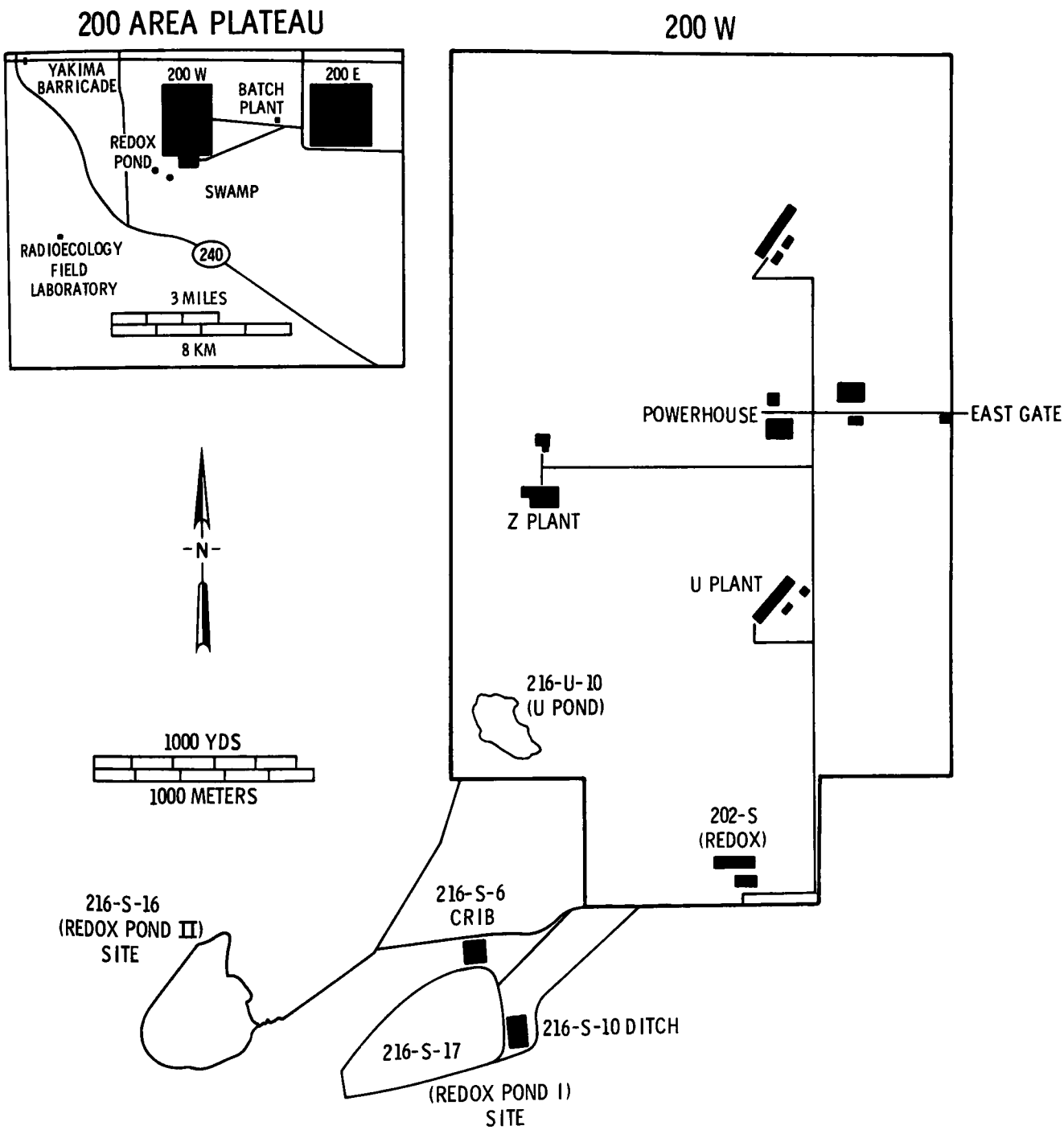


FIGURE 1. Map showing the locations of the 200 Area environs and 216-S-17 Pond.

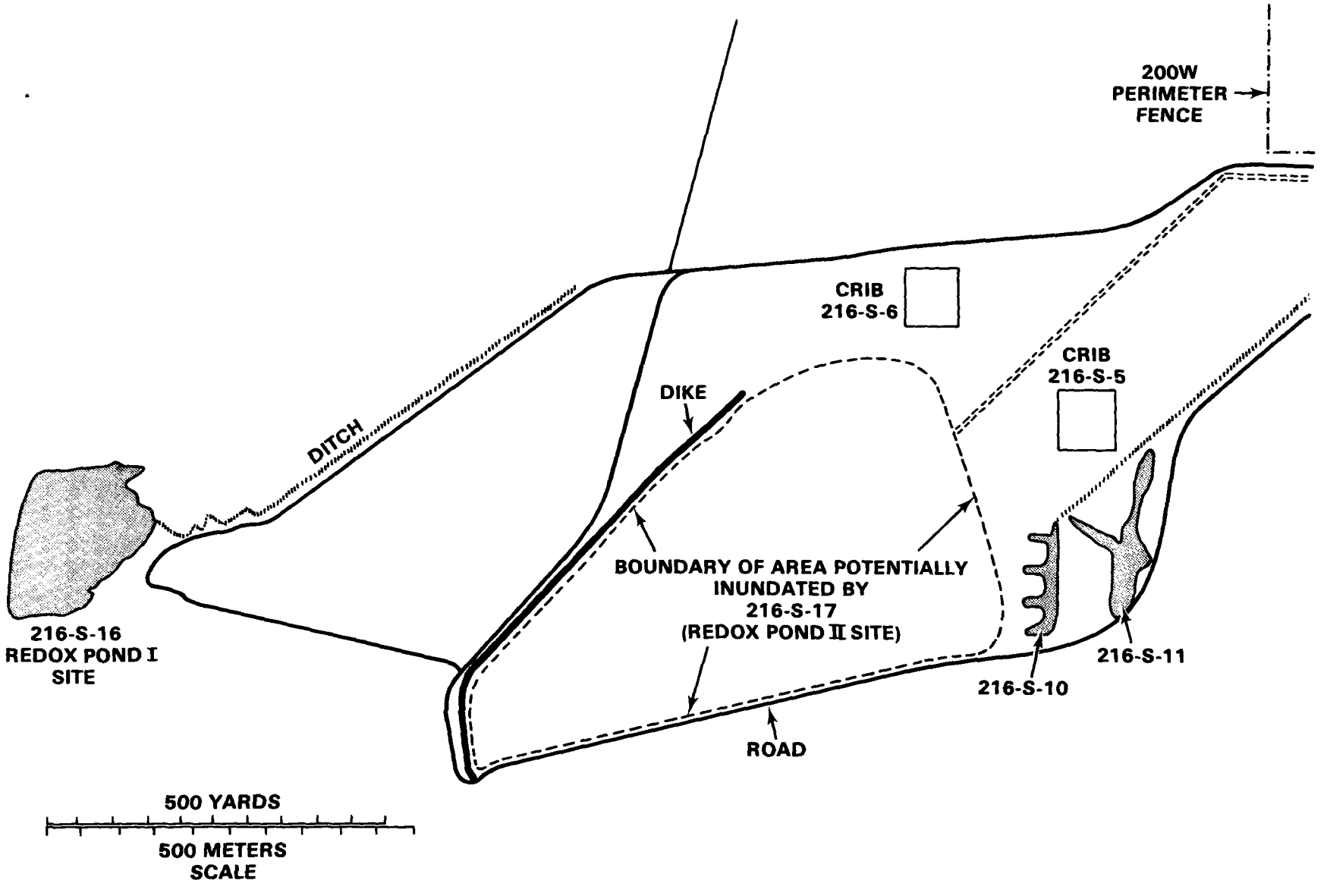


FIGURE 2. Detailed map of the 216-S-17 Pond site and surrounding environs.

In an attempt to control these problems, the western portion of the pond was seeded to Siberian Wheatgrass, an introduced range species from Eurasia. The revegetation attempt was successful and by 1975 Siberian Wheatgrass covered a large portion of the original pond area. This new plant was successful in stabilizing the surface soil and the numbers of tumbleweeds were greatly reduced through interspecific competition. However, wind erosion during the period from decommissioning in 1954 until reseeding in 1972 had reduced the original backfill in some areas to a depth less than 15 cm above the contaminated sediment layer. Subsequently, laboratory analyses of the Siberian Wheatgrass collected from these areas indicated that some of the plants had incorporated radionuclides into their leaf tissues

The objectives of this study were to determine radionuclide inventories for major biotic and abiotic compartments of the pond and evaluate nondestructive testing methods utilizing field instrumentation for in situ radionuclide analyses.

METHODS AND MATERIALS

Field sampling protocol

Sampling grid

Due to the irregular shape of the pond, a modified 30 meter quadrat grid system was utilized as a guide for vegetative sampling to assess radionuclide uptake. (Fig. 3) A theodolite was used to survey in the grid lines with metal posts placed at each 60 meter sampling location. Smaller wooden stakes marked the 30 meter intermediate locations. Peripheral areas of the actual pond site were not included in the grid. Only those areas supporting bunchgrasses with a density of at least 1 plant/100m^2 were considered. The grid comprised an area of approximately 7.6 hectares.

Vegetation sample removal

At each of the sampling locations two nearest Siberian Wheatgrass plants over 15 cm in diameter were selected for anlysis. Samples were obtained by cropping the bunchgrass at the base. Each plant was placed into a paper bag, labeled with the sample location number, and then transported to the laboratory for gamma energy analyses.

In situ analyses

Approximately five months following the vegetation sampling, a special-purpose mobile van (Dev Van I) designed to support in situ measurements of environmental radionuclide levels was utilized for additional field measurements. As a test of the unit's capability for providing credible, fast, inexpensive data concerning surface/subsurface waste, work was initiated to acquire concentration data for the vegetative sampling grid utilizing those same wheatgrass plants sampled earlier (Fig. 3). This provided an excellent opportunity to conduct a direct comparison of results, costs, and conclusions.

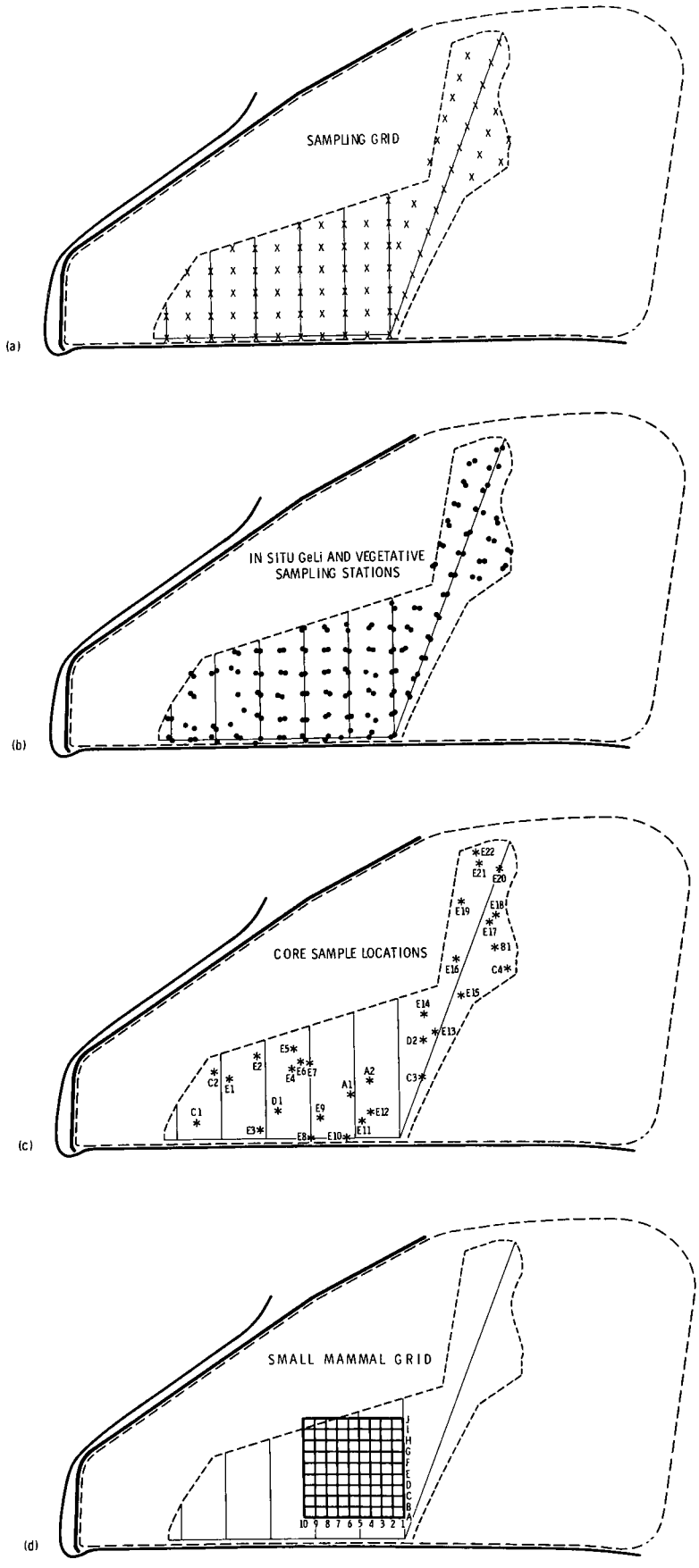


FIGURE 3. Maps showing locations of study area in relation to S-17 Pond boundaries: (a) location of field sampling locations; (b) location of the vegetation samples for lab analyses; (c) location of the soil core samples; (d) location of the small mammal trapping grid.

A collimated GeLi detector was employed to obtain in situ quantitative measurements of radionuclides contained in vegetation and soil. Signal output from the GeLi detector was interphased to a 1024 channel analyzer. Spectral data were transferred directly to magnetic tape and transported to the laboratory for further data reduction. A detailed description of the modified van unit can be found in Bruns (Br77). A ^{137}Cs source was used in the field for instrument calibration before actual sample-site counting was initiated.

Field measurements were made with the GeLi detector mounted on a conical lead shield 36 cm high and 36 cm in diameter at the base. Four hundred feet of coaxial cable connected the detector to the analyzer. All measurements were made at least 20 to 30 meters distant from the support van. Sample points were counted for five minutes.

Soil core sampling

A modified 30 meter quadrat grid system was used as a guide for site characterization work (Fig. 3c). Thirty-one cores were collected utilizing the stratified random grid sampling technique.

A tripod rig and hammer were used to drive a 6 cm diameter split-tube core sampler, utilized to reduce cross-contamination, to a depth of 125 cm. Use of the split-tube sampler permitted acquisition and transport of the samples as plastic sheathed, undisturbed sections approximately 38 cm in length. There were three such sections completed per 125 cm core sample. Drill depth and core length differed due to soil compaction by hammer impact. Core samples were transported in .5 liter cans to a laboratory hood for further subsampling. Each 125 cm core sample was divided by placing the 5 cm surface portion in one can and each ten centimeter portion thereafter in individual cans. The soil samples were oven-dried and weighed. Each soil portion was counted in the lab for ten minutes using a collimated GeLi detector.

Small mammal population

In order to sample the small mammal population of the pond area, a 10 by 10 trap grid was permanently staked. (Fig. 3d). Baited, Sherman folding live-traps were placed 15 meters apart, utilizing one trap per grid station. The grid was operated for at least four days per month from March through December.

The captured animals were examined and data were recorded concerning species, sex, reproductive condition, abnormalities, and weight. They were then toe-clipped for permanent identification and released at the point of capture. Only those animals which suffered mortality as a result of trap exposure (N=27) were collected for radionuclide analysis. Whole carcasses were counted to determine total body burden (pelt and GI tract were not removed) of ^{137}Cs. Population estimates were calculated using the Jolly-Seber method (Jo65) and density estimates were based on the boundary-strip method (St54).

Additional soil samples were collected from 20 mammal burrows and 11 ant mounds for ^{137}Cs analysis to determine if these animals were transporting subsurface contamination above ground. These samples were randomly collected from the mammal or ant burrow nearest to each sample core site (Fig. 3c).

Laboratory analytical methods

Gamma pulse height (GeLi), strontium 89-90 (wet chemistry), and uranium analyses (spectrophotometer) were performed. Samples of soil, vegetation, and mammals were analyzed for comparison with the in situ GeLi measurements to establish appropriate double-sample techniques.

RESULTS AND DISCUSSION

The estimated inventory of principal radionuclides in the S-17 Pond system calculated from discharge data is presented in Table 1 . The present study considered all major radionuclides. However, only data for ^{137}Cs have been returned from laboratory analyses and are discussed here. Table 2 presents inventory data for ^{137}Cs calculated from this study for the major abiotic and biotic components of the pond site. Figure 4 shows the calculated spatial variability of ^{137}Cs. The concentration contour map (Fig. 4a) was generated by an aerial radiological survey (Ti75) and the surface concentration GeLi detector contours (Fig. 4b) were determined with portable instrumentation associated with Dev Van I. The surface water map (Fig. 4c) shows the relationship between the sample grid and the observed surface water distribution in 1953. The final map (Fig. 4d) depicts the concentration isopleths for the Siberian wheatgrass samples collected for comparative lab analyses. The vertical distribution of ^{137}Cs in the soil column is presented in Figure 5. This was the mean depth profile based on weighted means from each of the concentration contours generated by the in situ field measurements (Fig. 4b).

Soil

The average surface soil concentration associated with the S-17 sample grid was approximately 17 pCi/gm (Table 2). The average levels of ^{137}Cs in soils of the 200 Areas from worldwide fallout and plant operations range from 1-3 pCi/gm dry weight . The overall soil inventory for the site was estimated to be 6.3 Ci (Table 2). This value agrees reasonably well with the estimated inventory of 17 Ci calculated from discharge information (Table 1), considering the fact that the study grid comprised only 30 to 40 percent of the potentially contaminated area (Fig. 4c). About .1 Ci of the soil total for ^{137}Cs is associated with the top 5 cm of soil. Therefore, assuming that the top 5 cm are potentially susceptible to wind erosion, a maximum of only .02 percent of the total soil contamination would be available for atmospheric resuspension.

The vertical profile data (Fig. 5) show a majority of the contaminated sediments to be located at the 5 to 35 cm depth. Although the pond site was reportedly backfilled with 45 to 60 cm of soil , the vertical distribution data indicate that there was insufficient backfill in some areas, or

Table 1. Inventory of major radionuclides discharged to 216-S-17 Pond

Radionuclide	Curies At Time of Discharge	Curies Decayed Through 1/1/79
Pu	0.2	0.2
^{90}Sr	40.0	21.1
^{137}Cs	30.0	16.5
U	0.05	0.05

Table 2. Cesium - 137 Inventory for Major Compartments of S-17 Pond Study Area ($\bar{x} \pm SE$)

Compartment	N	Mean pCi/gm	Mean $\mu Ci/m^2$	Total ^{137}Cs Per Organism (pCi)	Totals	Percent of Total
Total Soil	381		---	---	6.3 Ci ± 3.6 Ci	99.998%
Surface Soil*	31	17 ± 5.3	1.8 ± .6	---	$1.3 \times 10^5 \pm 4.2 \times 10^4$ µCi	.02%
Vegetation	165	4.7 ± .7	$1.5 \times 10^{-3} \pm 2 \times 10^{-4}$	2220 ± 330	110 ± 16 µCi	.002%
Small Mammals**	27	1.9 ± .7	---	26 ± 10	0.004 ± 0.001 µCi	2×10^{-8}%
					TOTAL 6.3 ± 3.6 Ci	

* Surface soil values are for only the top 5 cm of soil and are included in the total soil inventory
** All values for mammals are wet weight

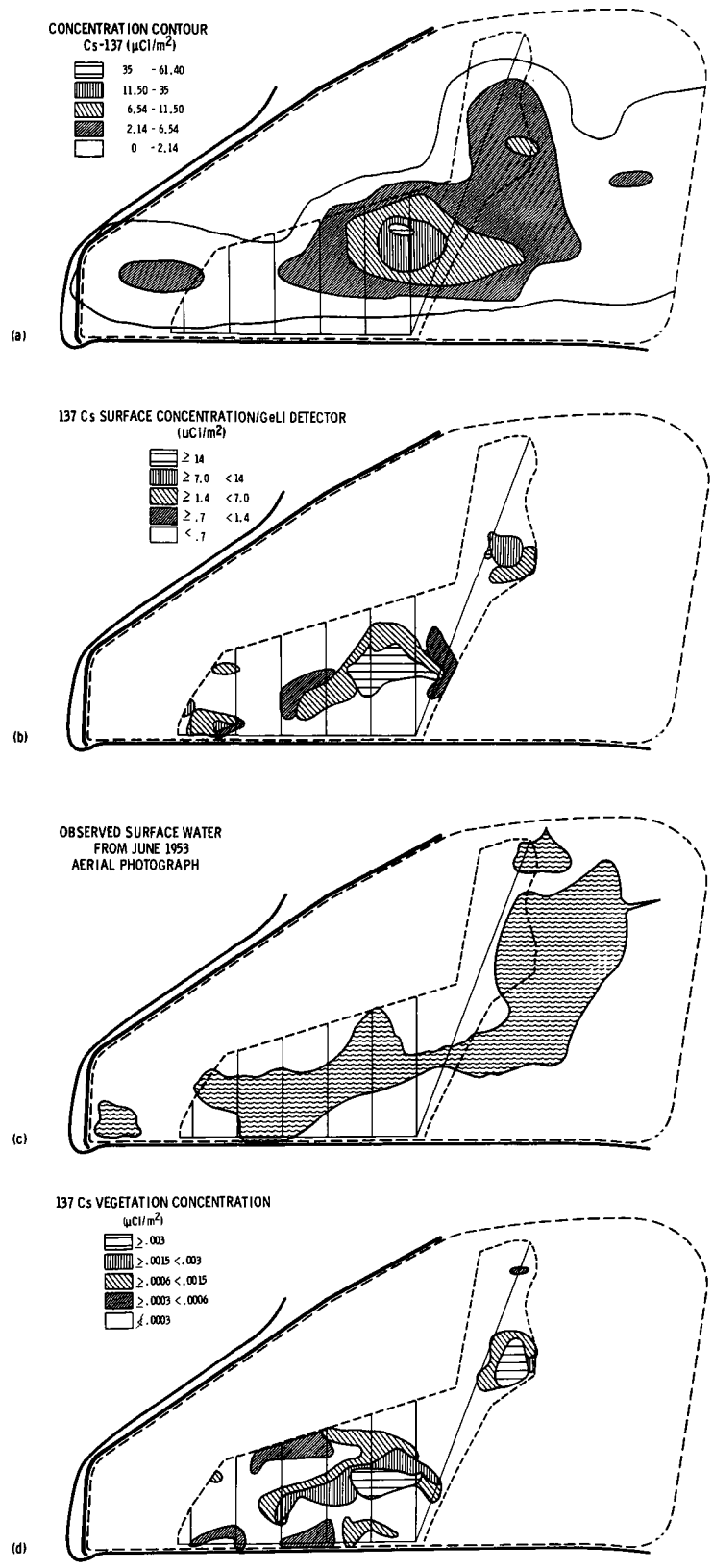
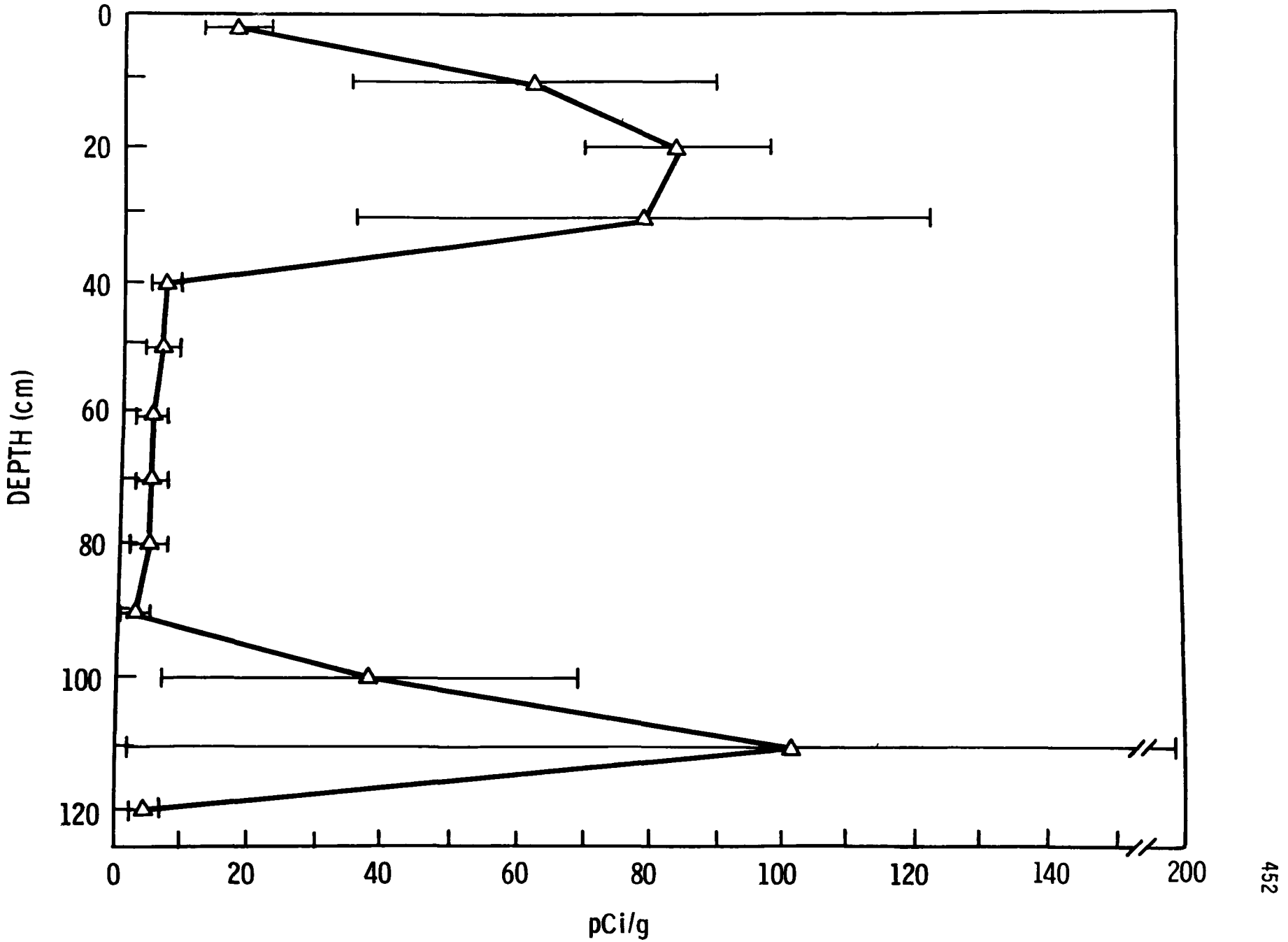


FIGURE 4 Maps relating spatial variability of ^{137}Cs associated with S-17 Pond and study grid: (a) concentration contours of ^{137}Cs distribution calculated from 1973 aerial survey data; (b) ^{137}Cs concentration contours generated from in situ GeLi Measurements; (c) observed distribution of surface water in S-17 Pond based on aerial photograph; (d) vegetation concentration contours based on field collection and laboratory analysis of Siberian Wheatgrass.


0
20
40
60
80
100
120
DEPTH (cm)
0
20
40
60
80
100
120
140
200
pCi/g

considerable wind erosion took place over the period before revegetation. The spike noted at the 95-115 cm depth was due to a single core located within an area of the pond which received a greater proportion of soil backfill (106 cm).

It should be noted that surface concentration values (Fig. 4a, b) were calculated from exposure data utilizing in situ measurements. They do not represent actual concentrations for surface soil. The inventories provided in Table 2 and Figure 5 present data on the calculated amounts of ^{137}Cs contained in the top 5 cm of soil based on soil core samples. The highest exposure rate observed on the pond site was .5 mR/hr at 1 meter.

Vegetation

The principle thrust of the vegetation sampling program was directed toward ascertaining the potential for radionuclide uptake in Siberian Wheatgrass, since the plant appeared to be an extremely successful perennial for revegetation of disturbed areas. The average background value due to fallout in vegetation is 2 ± 1.2 pCi/gm in the Hanford environs while the mean value determined for Siberian Wheatgrass was 4.7 ± .7 pCi/gm dry weight. Standing crop estimates calculated in early May 1978 provided a total biomass estimate for Siberian Wheatgrass of 0.3 kg/m^2 dry weight. This resulted in a total ^{137}Cs inventory for the wheatgrass on the study site of 110 µCi (Table 2). Visual inspection of root depth indicated that the majority of the root mass was associated with the 5-30 cm soil depth, with the longest roots being less than 60 cm long. Apparently, over a period of time the combination of wind erosion and shallow soil backfill (45-60 cm) resulted in optimum conditions for uptake of radionuclides by the Siberian Wheatgrass. However, only .002 percent of the total calculated site inventory was incorporated by the Siberian Wheatgrass. The concentration ratio observed from this study appeared to be in the order of the 10^{-1} to 10^{-2} range. This is comparable with other vegetative types, such as cheatgrass and tumbleweeds in the Hanford area (C172). With proper range management techniques, including at least one meter of backfill and erosion control until seedlings are established, Siberian Wheatgrass would be a suitable perennial for revegetation of disturbed sites in the Hanford area.

Small mammals

A total of 27 Great Basin pocket mice (Perognathus parvus) were analyzed for total body burden of ^{137}Cs. The highest value for all the samples was 13.6 pCi/gm and the sample mean was 1.9 ± .8 pCi/gm wet weight (Table 2). Control samples, affected by fallout levels alone, indicate mean values for ^{137}Cs in small mammals of approximately 2.2 ± 1.6 pCi/gm wet weight (Ga77). Therefore, although the animals on the S-17 Pond are living in a radiation zone they do not appear to be incorporating much ^{137}Cs into their systems. A density determination for the pond site indicated an average resident population of 20 mice/ha. Density estimates for the surrounding sagebrush (Artmesia tridentata) - Sandburg bluegrass (Poa sandburgii) - cheatgrass (Bromus tectorum) community are expectedly higher at 25 to 28 mice/ha (He76). Therefore, the total estimated ^{137}Cs inventory for the small mammal compartment of the study grid is .004 ± .001 µCi (Table 2).

The pocket mice could pick up ^{137}Cs by burrowing through contaminated soil and ingestion of small amounts of soil and attachment of soil particles to their fur and skin. They could also have incorporated the ^{137}Cs into their bodies by ingestion of the seeds and green tissues of the contaminated plants. At this time, it is not know whether these animals utilize Siberian Wheatgrass in their diets. If the body burden is due to ingestion of ^{137}Cs, the biological half-life for ^{137}Cs in small mammal species has been estimated to range from 2 to 8 days (St70; Ba75).

The behavior of pocket mice on the pond may be more important in terms of radionuclide availability than the small amounts contained within their bodies. The concentration values for soil samples collected from the ant mounds were within background values for surface soil with a mean of 1.1 $\pm$ .4 pCi/gm; the highest value being 5.35 pCi/gm. However, the mean value for the mouse burrow samples was 268.0 $\pm$ 175 pCi/gm, although this value was principally due to three samples with high levels (range 682-3490 pCi/gm).

CONCLUSIONS

Construction and other activities associated with radioactive waste site preparations usually result in the partial or complete denuding of the native vegetation on these sites. The restablishment of plant cover on these areas is a difficult task in an environment which receives only 6-8 inches of precipitation annually. Natural revegetation in the sagebrush steppe ecosystem is a slow and arduous process (Da70). Wind erosion of soils becomes a major problem on disturbed areas until the soil is stabilized by plant growth. As in the case of the S-17 Pond site, undesirable deep-rooted plants such as tumbleweeds become established very quickly. The use of Siberian Wheatgrass in a comprehensive revegetation program offers a solution to these types of waste management problems.

Visual inspection of Figure 4 (a and b) shows a high degree of correlation between the two in situ measurement techniques. These techniques also compare favorably with the surface distribution of ^{137}Cs in Siberian Wheatgrass based on laboratory analyses (Fig. 4d). The primary reason for this high degree of correlation is due to the homogenous vertical distribution of the contaminated sediment layer in the soil column throughout the study grid. Upon completion of additional radionuclide analyses and further utilization of the in situ GeLi technique at other waste sites with less homogeneity, appropriate double-sampling techniques will be developed. This will provide quick and relatively inexpensive techniques for estimating inventories at future waste sites.

In relation to the radionuclide inventory presented in Table 2, biotic and abiotic compartmental estimates generated by this study are indicative of the general patterns observed in previous radioecology studies on waste management sites at Hanford (C172). In general, greater than 99 percent of the total radionuclide inventory is associated with the soil or sediment column.

Acknowledgements

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RETENTION OF LOW-LEVEL RADIOACTIVE WASTE MATERIAL BY SOIL*

by

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Abstract

Low-level radioactive wastes produced by users of radionuclides are generally disposed by shallow land burial. Reliance for containment is placed on characteristics of shallow geologic formations or soils; thus, effective waste management requires a knowledge of radioactive waste/soil interactions.

Because of the wide variations in soil and waste characteristics, the degree of radionuclide retention would be expected to vary; knowledge of that variation may be of value in predicting radionuclide mobility. This report discusses results of investigations of radioactive waste/soil interactions as they relate to radionuclide retention and its variability among soils and radionuclides.

In soil column leaching studies, radioactive waste solutions were applied to four different soil types; ^{241}Am, ^{88}Y, and ^{172}Hf were retained in the top four cm of soil with better than 90% retained by a protective surface sand layer. Less then 50% of the ^{85}Sr, ^{137}Cs, and ^{83}Rb was retained by the surface sand. No ^{88}Y, ^{172}Hf, ^{85}Sr, ^{137}Cs, or ^{83}Rb was detected by gamma counting in the leachate solutions, however, using a more sensitive analytical technique small amounts of ^{238}Pu, $^{239,240}Pu$ and ^{241}Am were found in leachates from all soils. It appears that release of this small fraction of mobile radionuclide may have a significant long-term impact on the environment. It also appears that reliance for attenuation of some radionuclides can not be placed solely on characteristics of the soil matrix.

Introduction

Shallow land burial has been used for disposal of low-level radioactive waste material for more than three decades. Recently, investigators have observed small amounts of radioactivity in the environs of several of those sites, signaling the need for a more thorough evaluation of the potential and mechanisms of long-term redistriubution of buried radionuclides both on site and off site.

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*Work performed under the auspices of the Nuclear Regulatory Commission.

Field experiments to describe the physical and chemical interactions affecting radionuclide migration are very expensive and in some cases cannot yield the information required for prediction. Laboratory tests are, then, generally conducted to derive the necessary data for use in flow or migration prediction models.

Over the past three decades many studies of radionuclide migration have been conducted in soil and geologic materials to assess the degree of sorption or retention the materials have for the radionuclides. However, until recently, most studies of that type relied on single prepared radioisotope solutions in carefully controlled systems and did not represent the very complex conditions existing in radioactive waste interactions with soil or geologic media.

A study was designed to provide information on radionuclide retention and migration in soils as a function of soil type and radionuclide species. Preliminary results show the nature of low-level radioactive waste material interactions with several widely varying soil types. Based on information gained in this study, future experiments will be designed to more precisely evaluate factors affecting retention or migration and to develop a measure of the confidence one might expect in retention values. This information will be incorporated into radionuclide migration and dose assessment models used in low-level waste management and licensing.

This study is one phase of a larger study to derive some confidence in predictions of radionuclide retention by soils and is sponsored by Nuclear Regulatory Commission, Division of Safeguards, Fuel Cycle and Environmental Research. Figure 1 depicts the rationale behind the study. A great deal of variability in the degree of radionuclide retention does exist because of the wide variety of radioactive sources and species in each source and the large variability in the evironmental materials available for interaction. The variety of waste sources is indicated in the first column of Fig. 1; examples of characteristics of the waste/soil solution believed to influence retention are listed in the second column. The degree of radionuclide retention as determined by soil types for each waste/soil characteristic will define an envelope from which some confidence relative to retention can be projected. Correlation of the retention with one or a combination of more specific parameters (characteristics), such as, soil texture, pH, clay type, *etc.*, may yield a smaller degree of predicted retention variability. In other words, the more that is known about the system the better one can predict both short- and long-term radionuclide retention .

Materials and Methods

Four soils were reacted with a low-level radioactive industrial waste solution in order to evaluate the radionuclide retention capacity of the soils. Several major physical and chemical soil characteristics likely to be related to radionuclide retention are listed in Tables 1 and 2. The A_p horizon of Fuquay loamy sand represents the top 15 cm of soil collected from the Barnwell area of South Carolina. The A_p horizon of Fayette silt loam represents the top 18 cm of soil collected from Sheffield, Illinois. The B horizon of Carjo loam and the C horizon of Puye sandy loam are from Los Alamos, New Mexico and were collected from depths of 5 to 15 cm and 25 to 51 cm, respectively. The four soils range in pH (saturated paste) from 5.5 to 7.1 and have similar cation exchange capacities (CEC) ranging from 15 to 23 meq/100 g of soil. Although the clay content of the Fuquay is very low, enough clay is

present to identify the major clay types as vermiculite and kaolinite. One of the characteristics believed to be very important in mediating the chelate complexation of radionuclides is the available (extractable) iron. Large amounts of extractable iron were present in Fayette and Fuquay soils as measured by the synthetic chelating agent diethylenetriaminepentaacetic acid (DTPA).

Leaching columns, 3.8 cm in diameter were prepared in triplicate using 300 g (air dry) of each soil type. A 1-cm plug of washed silica sand (20-50 mesh) was placed at the top and bottom of each column. After packing, each column was saturated with distilled water from the bottom to minify the trapping of air and then allowed to drain to field capacity prior to commencement of leaching.

Soil column setup and leaching were conducted according to the procedure of Essington and Nishita (Es66). The waste solution was added to the top of the soil column in 10-cm irrigation increments (115 ml each) until the infiltration rate of the waste solution was seriously impaired. At that point 2.5 l of waste solution had been passed through the soil column. Each 10-cm leach was collected and a selected few were analyzed for major gamma emitting radionuclides and ^{238}Pu, $^{239,240}Pu$, and ^{241}Am. The gamma emitting radionuclides were measured with a germanium (lithium drifted) detector and pulse height analyzer. The other radionuclides were measured using radiochemical separations and alpha pulse analysis. Electrical conductivity, Eh, pH, and alkalinity were also determined on each leachate. Upon completion of the leaching experiment each column was fractioned into approximately 2-cm increments. Each increment was analyzed for major gamma emitting radionuclides and ^{238}Pu, $^{239,240}Pu$, and ^{241}Am.

The leaching solution was a two-week composite of a liquid radioactive waste material accepted by a waste treatment facility. Table 3 lists several of the many measured characteristics of a similar waste material as collected and after centrifuging to remove particulates greater than about 0.05 μm in diameter. Material remaining in the supernatant solution after centrifugation was designated as "soluble" although the supernatant probably contained small particulates, polymers, and ions. Centrifugation removed substantial quantities of phosphate, aluminum, iron and organic matter, suggesting that a predominance of these materials was associated with particulates larger than 0.05 μm. Those materials are also suspected of being involved in interactions with some of the radionuclides in the waste.

Some of the more prevalent radionuclides present in the waste are listed in Table 4. The ratio of radionuclide concentration before and after centrifuging are given to indicate their degree of partition and to suggest a degree of complexity of the waste solution. Three groupings are suggested in Table 4 based on the amount of radionuclide contained in the soluble fraction relative to the total. The group including ^{83}Rb, involves the largest amount of soluble radionuclide, whereas the group including ^{88}Y involves the largest amount of insoluble radionuclide. Note particularly that the partition of the two isotopes of plutonium is significantly different in the two fractions (standard deviation/ mean of 4 replicates = C.V. $\simeq$ 0.1).

Results and Discussion

The distributions of various waste radionuclides in soils and leachates indicate the complexity of the chemical and physical interactions between the

waste and the soil matrix. That complexity is indicated in Fig. 2 by the presence of at least two forms of ^{241}Am shown by the accumulation of a large portion in the sand layer and by the small but constant amount in the deeper soil fractions. Figure 2 also shows the ^{241}Am distribution in the three relicate soil columns of Carjo soil as well as the variability of ^{241}Am in the various soil fractions. The vertical bars represent the plus or minus one-sigma counting error for each fraction. Note that in the shallow portion of the soil columns the counting error is small and data are reproducible. In the deeper portions of the column the counting error is large, however, the ^{241}Am levels for the three replicates appears to fall well within the counting error. A non-linear least-squares procedure was used to fit the ^{241}Am values in the soil column fractions to an exponential equation of the type shown on the figure. Subsequent figures will use those fitted curves where possible for clarity of presentation; however, several of the curves were hand fitted.

Figure 3 shows the distribution of ^{241}Am in the four soils studied. The distribution of ^{241}Am in Puye, Carjo, and Fuquay, did not appear to be different, whereas ^{241}Am in Fayette appeared to be distributed somewhat differently, particularly in the shallower parts of the column. Note that in all cases the sand layer retained about 90% of the ^{241}Am in the column. Additionally, the level of ^{241}Am in the deeper column fractions approached a constant. That level of ^{241}Am was calculated to be nearly the same concentration as that found in the final leachates and therefore, can be attributed to the soil column water remaining at field capacity.

Hafnium-172, ^{241}Am, and ^{88}Y appear to be similarly distributed in all soil types as illustrated by the data for the Puye soil in Fig. 4. As with ^{241}Am, the sand layer accumulated better than 90% of the ^{172}Hf and ^{88}Y. Hafnium-172, ^{88}Y, and ^{241}Am were not detected in the leachate solutions with the gamma counting technique initially employed. However, the lower levels of ^{241}Am were easily detected by radiochemical separations and alpha pulse counting.

The distribution of ^{137}Cs in the four soils is shown in Fig. 5. In this case only about 40% of the ^{137}Cs was removed by the sand layer; however, ^{137}Cs was not detected in the leachate solutions. The differential distribution of ^{137}Cs in the 5- to 12-cm region of the Puye soil may be related to soil characteristics. In the shallower depths the ^{137}Cs had distributed in a manner similar to that for ^{241}Am, ^{172}Hf, and ^{88}Y as indicated by the slopes of the distributions in the 0- to 5-cm region.

Strontium-85 distribution in the four soils is shown in Fig. 6. Only about 40% of the ^{85}Sr was removed by the sand layer and no ^{85}Sr was detected in the leachate solution. Strontium-85 appears to distribute in the soils differently from the radionuclides discussed earlier. This conclusion is based on the observation that the slopes of the distribution curves in the upper portion of the columns are not as steep as for the other radionuclides, indicating that greater amounts of ^{85}Sr have moved deeper into the soil columns. Similar distribution patterns were reported by Essington *et al.* (Es66) and Nishita and Essington (Ni67) in columns of agricultural soils containing radioactive strontium and leached with irrigation or distilled water. Movement or retention of the strontium was believed to be governed by the sorption mechanisms of the soils and the competition of calcium ions for sorption sites. Since no ^{85}Sr was found in the leachate solutions, that portion of the ^{85}Sr not removed from the waste leaching solution by filtration appears to be attenuated by sorption mechanisms.

Figure 7 shows the distribution of ^{83}Rb in the four soils. The distribution of ^{83}Rb in Puye, Carjo and Fayette was similar to that of ^{241}Am; however, only 10% of the ^{83}Rb was retained by the sand layer and no ^{83}Rb was found in the leachates.

Rubidium-83 distribution in Fuquay was quite different from that in the other soils in that concentrations were constant with depth to about 7 cm and then steadily decreased to a non-detectable level. It appears that the ^{83}Rb was retained by Fuquay to a lesser degree than was ^{85}Sr (Fig. 6). However, based on the similarity of the ^{83}Rb distribution to that of ^{85}Sr similar sorption mechanisms may be involved. No effort was made to determine stable rubidium in the waste solution; stable rubidium could have a significant effect on ^{83}Rb retention by the soil matrix.

Plutonium-238 and $^{239,240}Pu$ were measured in the soil column leachates only. Figure 8 shows the amount of ^{238}Pu added with each increment of waste solution and the amount of ^{238}Pu found in each leachate; data are presented as averages of the leachates from the three soil columns. The appearance of ^{238}Pu in the first leach indicates a rapid breakthrough. The attainment of a relatively constant level after the second waste addition indicates that portion of ^{238}Pu in the waste solution that was not filtered or sorbed by the soil. A comparison of the ^{238}Pu in the leachate with that added to the soil columns shows that 83 to 97% of the ^{238}Pu was retained by the soil columns. Those results indicate that there was a small but highly mobile fraction of ^{238}Pu in the waste solution. Although $^{239,240}Pu$ was found in the same leachate, $^{239,240}Pu$ reacted somewhat differently from ^{238}Pu. This is shown in the ratios of $^{239,240}Pu$ to ^{238}Pu in the waste solution, centrifuged waste, and the soil leachates (Fig. 9). In general, the ratios for soil leachates fell between those of the source waste and centrifuged waste solutions. In the early stages of leaching there were rapid changes in the leachate plutonium ratios, but as leaching progressed the ratios tended to converge at about 0.1. Those data show that isotopes of the same element can react differently in the soils. Plutonium in the waste solution originates from a number of widely different operations. Those operations dealing primarily with ^{238}Pu tend to dispose of a higher percentage of soluble ^{238}Pu than soluble $^{239,240}Pu$. Those operations dealing primarily with $^{239,240}Pu$ and ^{241}Am (Table 4) tend to dispose of particulate material or material prone to be associated with particulates. Thus the nature of the source material may dominate the retention pattern in soils and the differences in plutonium isotopic distributions are not necessarily due to basic differences in behavior of the isotopes.

The waste/soil system is complex as indicated by the data presented. That complexity may be a result of a number of interactive mechanisms. The major mechanisms, in addition to filtration and ion exchange, include: precipitation and dissolution of calcium carbonate; complexation; and microbiological growth. Those mechanisms as well as evaluation of changes in the waste source upon aging and results of batch sorption studies with the same soils were reported by Fowler *et al.* (Fo78).

Waste radionuclides interacting with calcium carbonate, the primary inorganic mineral in the waste, should behave in a manner similar to that of the calcium carbonate. For example, changes in the carbonate system which result in either a precipitation or dissolution of solid carbonates should result in

a change in the radionuclide concentration in the soil solution. Such changes have been noted to occur in the waste solution upon aging, and undoubtedly occur upon contact with the soil. The changes could continue to occur during the progress of leaching as the leaching solution aerates or changes upon encountering different materials in the soil column. The extent of the carbonate effect on radionuclide retention was not specifically investigated in the experiment. However, the presence of $CaCO_3$ in the waste solutions was confirmed by X-ray diffraction. Significant decreases in the concentration of soluble plutonium and americium were observed upon formation of $CaCO_3$ in the waste material, whereas, changes in the soluble cesium and uranium concentrations did not occur. Those observations may account, in part, for the differences in the amounts of ^{241}Am and ^{137}Cs retained by the sand layer on top of the soil column.

The small but significant amount of plutonium and americium found in the leachate solutions may be due to carbonate complexing or the presence of chelated species. The charge of some soluble actinides in the waste was shown to be predominatly negative whereas the charge of the ^{137}Cs was predominantly positive (Po79). This was accomplished by passing the soluble waste fraction through cation and anion exchange resins and observing the amount of radionuclide not held by the resin (Fowler *et al.* Fo78). The work of Alberts *et al.* (A177) on the identification of the charge of plutonium in samples of Lake Michigan water indicated that the charge of the plutonium species was negative. They attributed the negative nature of the charge to the formation of a stable carbonate complex in the $CaCO_3$ super-saturated water having a pH of approximately 8. Ames *et al.* (Am76) present stability diagrams for plutonium and uranium which indicate that at pH 8.0 carbonate complexes could account for the predominantly negative charge of plutonium and uranium species.

Although the waste solution was not analyzed for the presence of chelating agents it is strongly suspected that significant quantities were present. The sources of the waste solution include decontamination fluids containing detergents and chelating agents and chemical laboratory wastes where chelating agents and other organic complexers are used and discarded. The effect of the chelating agents would be to form a very stable complex with the radioactive rare earth or actinide ions present and at pH 8 the resulting complexes are likely to be neutral or negatively charged. Those species would migrate through the soil rapidly with the possibility of exchange with metal ions in the soil solution. The degree of exchange of the radioactive ion with a metal ion would be dependent upon their concentrations in the soil solution and upon their relative stabilities with the chelate ligand.

During the conduct of the column leaching experiment an algal growth was allowed to be established in the columns. Normally the leaching experiment would be conducted with care not to allow unnatural microbiological activity; however, waste solutions allowed to impact the open environment would surely be subject to microbiological activity. The effect of the algae on radionuclide retention in the soil columns is a valid parameter. A separate experiment was conducted to test the effect of algae on their capacity to assimilate or tieup waste radionuclides. An algal bloom was allowed in the waste solution in which the pH was adjusted to 6.0; the algae were removed, washed, and analyzed. The effect of algae on retention of waste radionuclides depended on the radionuclide; a large percent of the actinides (86-95%), but only 30% of the cesium was associated with the algae. Those results are consistent with the large

accumulation of ^{241}Am, ^{172}Hf, and ^{88}Y and the somewhat smaller degree of accumulation of ^{137}Cs, ^{85}Sr, and ^{83}Rb in the sand layer on the soil column.

The information presented thus far is indicative of the complexity of the waste/soil systems. Reactions of liquid waste materials from different low level waste streams, shallow waste burial pits or trenches, or the high level waste processing streams accidentally released to the soil environment may be even more complex.

Relating the retention data to migration rates and distance is generally accomplished by use of the distribution coefficient (K_d) in various velocity models. A simple version of a model relating radionuclide velocity (V_R) to K_d is as follows:

$$V_R = \frac{V_W}{1 + K_d r}$$

where: r = bulk density/porosity,
V_W = velocity of water.

More complex models have been derived, some of which are reviewed by Travis (Tr78). Most velocity models still use K_d as the parameter that transmits the degree of radionuclide retention to the model. Actually K_d was defined by Mayer and Thompkin (Ma47) as follows:

$$K_d = \left(\frac{C_0}{C} - 1 \right) \frac{V}{M}$$

where: C_0 = tracer concentration in solution before adding mineral,
C = tracer concentration in liquid phase of mineral-water suspension,
V = volume of liquid,
M = mass of solid.

Many investigators recognize that the K_d values include a number of assumptions or qualifications which result in minimal impact when considering single source radionuclides in simple systems. The inherent assumptions and qualifications appear to not result in minimal impact when used for real radioactive wastes. Quoted K_d values might be used by licensers or policy makers without due consideration of the limitations.

The distribution coefficient should be independent of the solid to liquid ratio. In practice, however, equilibrium is never attained in an environmental system, and evidence has appeared suggesting that K_d is not independent of the solid to liquid ratio. This has profound impact when measuring K_d in the laboratory using a 4-part liquid to 1-part solid ratio then applying that data to a system of water flow in a similar soil or geologic medium where the effective water to solid ratio may be 1 to 100. Further, a more important concern in the concept of K_d for predicting radionuclide migration relates to the complexity of the waste/soil system. A

number of different forms of a given radionuclide were shown to exist *e.g.*, filterable, sorbable, and highly mobile. Each form may consist of a number of species; each specie could be described by a separate K_d value. Using K_d values generated on a combined system will not predict the degree of retention of all of the species present. Some species will migrate more rapidly and some more slowly than average. Those species migrating rapidly, even if in low concentrations, may constitute a significant impact on the environment. Even if a K_d value is generated for a particular specie, as that specie migrates to a new matrix material or is intercepted by a leach solution of different characteristics, the degree of retention will likely change thus negating that K_d as a predictor.

Summary

Four soils of widely differing characteristics were treated with a radioactive liquid waste solution in order to evaluate the degree and pattern of retention of various long lived radionuclides. The experimental system was designed to provide preliminary information on various waste/soil systems representative of accidental releases to the environment and possible releases from shallow land burial.

Results of the studies indicated that the wastes were very complex in form and species of each radionuclide. Some isotopes of the same element appear in different forms, each acting differently with respect to retention by soils. The results also suggest that extreme care must be exercised when generating and using the standard distribution coefficient (K_d) in migration and dose assessment models. The K_d values generated in laboratory tests may not predict adequately the true degree of radionuclide interaction with the soil. Migration rates of some portion of the radionuclide sources may change with time and distance complicating even further the predictive value of K_d.

Results of the laboratory studies are significant in the interpretation of the migration of waste radionuclides from shallow land burial pits. Although wastes buried in a shallow land disposal pit are considered solids, a certain fraction is converted to a liquid phase through decomposition processes and the action of seepage water and becomes available for transport. Physical removal of a filterable fraction by a surface soil in a biochemically dynamic environment will result in eventual degradation of organic matter and the release of associated radionuclides to the soil solution.

The fate of all forms of a radionuclide in the environment may ultimately be similar. However, initially some radionuclides may migrate great distances because of their nature. Thus, a prediction of the movement or changes in the movement of waste radionuclides from a point of release necessitates some knowledge of biochemical, chemical, and physical interactions of waste radionuclides with their environment.

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	TOTAL	SOLUBLE
pH	7.8	8.1
Eh (mV)	+410	+400
PO_4 (*)	29	2
Ca (*)	75	51
Al (*)	2	0.2
Fe (*)	12	0.4
COD (*)	170	60

(*)=(ppm)

TABLE 1. SELECTED MAJOR PHYSICAL CHARACTERISTICS OF FOUR SOILS USED IN LEACHING STUDIES.

	Puye	Carjo	Fayette	Fuquay
	(ppm)	(ppm)	(ppm)	(ppm)
Total Ca	10 000	10 000	10 000	1000
Ammonium Acetate Extractable				
Ca	1800	1000	1400	230
Mg	170	200	330	22
Fe	4.8	5.4	36	1
Mn	24	54	24	16
Al	6	9	24	4
DPTA Extr. Fe	6	7	28	50

TABLE 2. SELECTED MAJOR CHEMICAL CHARACTERISTICS OF FOUR SOILS USED IN LEACHING STUDIES.

	Puye	Carjo	Fayette	Fuquay
P_w at F.C.[1]	43	40	40	30
CEC	23	18	15	16
pH	7.1	6.4	6.6	5.5
SAND %	45	53	19	86
SILT %	48	37	73	14
CLAY %	7.5	10	8.5	~0
CLAY TYPE[2]	KM	KM	KMV	VK

[1]Percent Moisture at Field Capacity

[2]V=Vermiculite; K=Kaolinite; M=Montmorillonite

TABLE 3. SELECTED PHYSICAL AND CHEMICAL PROPERTIES OF SOURCE WASTE (TOTAL) AND SOLUTION PHASE (SOLUBLE) AFTER CENTRIFUGATION.

	$T_{1/2}$	S/T[1]
^{83}Rb	83d	.67
^{85}Sr	64d	.54
^{137}Cs	30y	.60
^{238}U	4.51×10^{9}y	.46
^{88}Zr	85d	.15
^{60}Co	5.26y	.11
^{88}Y	108d	.018
^{172}Hf	5y	.001
^{241}Am	458y	.012
^{238}Pu	86.4y	.031
$^{239,240}Pu$	24390y, 6580y	.011

[1]T=Source waste, S=Soluble fraction

TABLE 4. MAJOR RADIONUCLIDES IDENTIFIED IN SOURCE WASTE, THE RADIOACTIVE DECAY HALF-LIFE, $T_{½}$, AND THE PARTITION UPON CENTRIFUGATION.

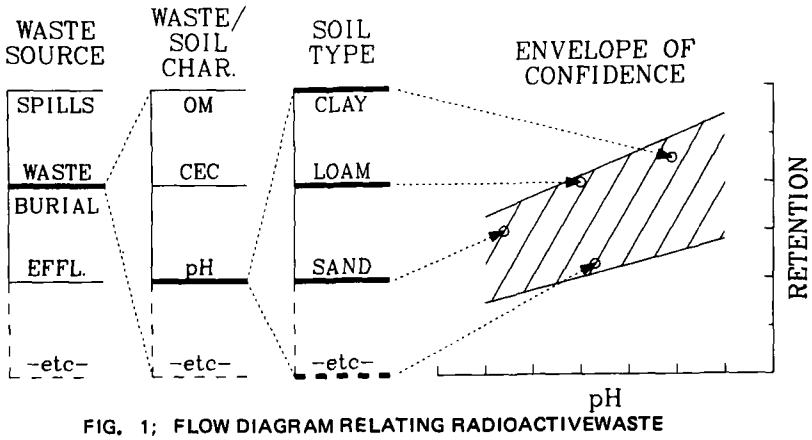

FIG. 1; FLOW DIAGRAM RELATING RADIOACTIVEWASTE SOURCE AND SOIL CHARACTERISTICS TO RADIONUCLIDE RETENTION AND THE DEVELOPMENT OF STATISTICALLY BASED CORRELATIONS

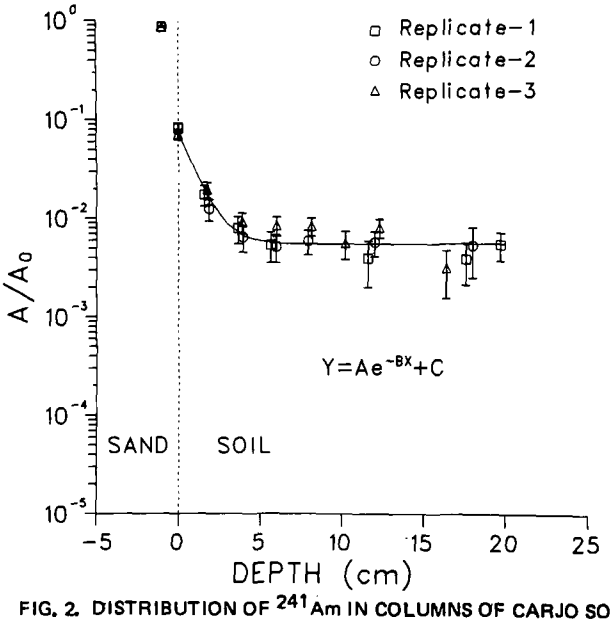

FIG. 2. DISTRIBUTION OF ^{241}Am IN COLUMNS OF CARJO SOIL AFTER LEACHING WITH RADIOACTIVE WASTE. A/A_0 REPRESENTS FRACTION OF TOTAL COLUMN ^{241}Am FOUND IN EACH INCREMENT.

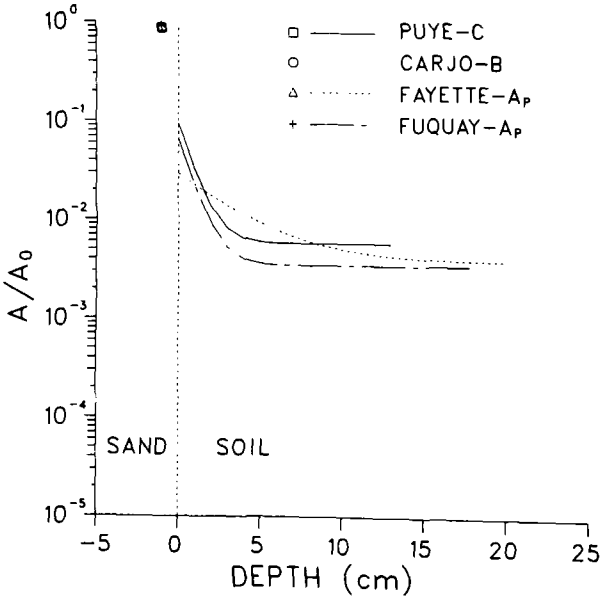

FIG. 3. DISTRIBUTION OF ^{241}Am IN FOUR SOILS.

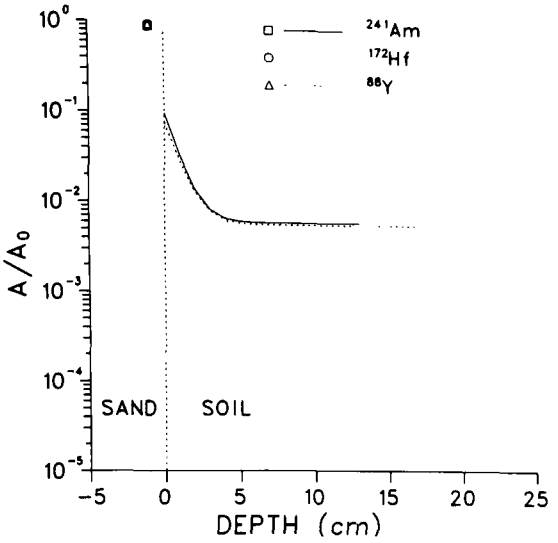

FIG. 4. COMPARISON OF THE DISTRIBUTION OF ^{241}Am, ^{241}Am, ^{172}Hf AND ^{88}Y IN PUYE SOIL.

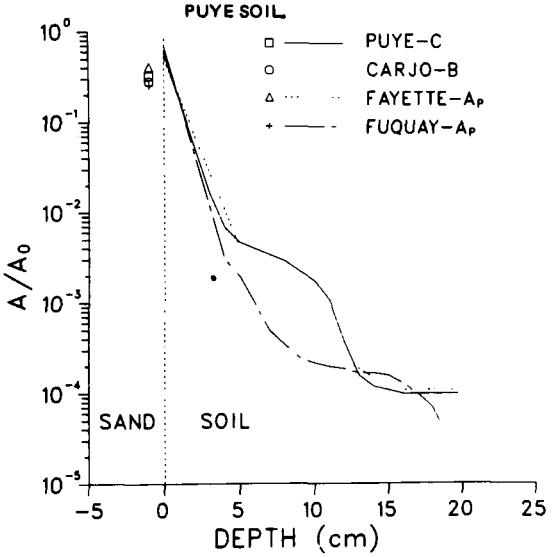

FIG. 5. DISTRIBUTION OF ^{137}Cs IN FOUR SOILS.

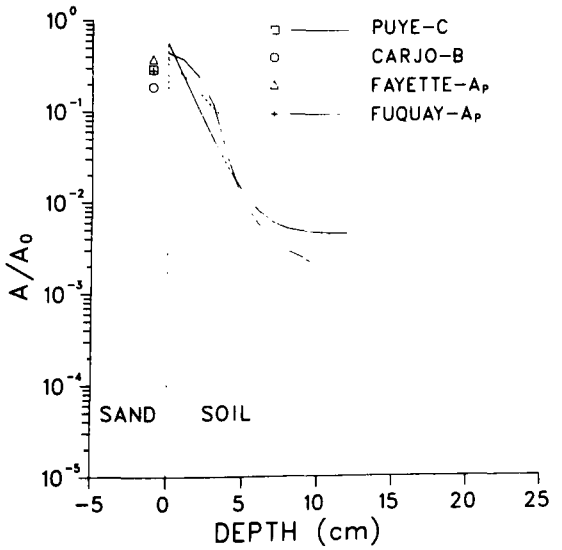

FIG. 6. DISTRIBUTION OF ^{85}Sr IN FOUR SOILS.

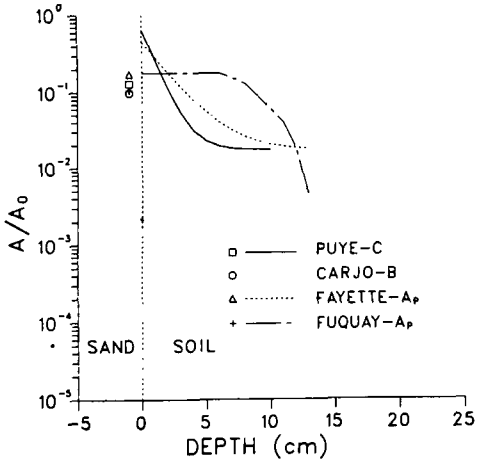

FIG. 7 DISTRIBUTION OF ^{83}Rb IN FOUR SOILS.

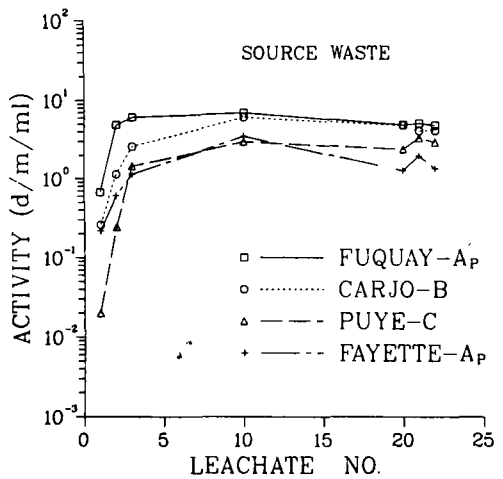

FIG. 8. PLUTONIUM-238 IN SOURCE WASTE AND IN LEACHATES FROM FOUR SOILS.

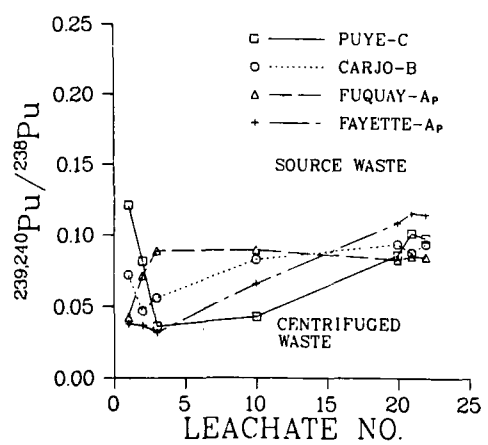

FIG. 9. RATIO OF $^{239, 240}Pu$ TO ^{238}Pu IN SOURCE WASTE, IN CENTRIFUGED WASTE, AND IN LEACHATES FROM FOUR SOILS.

THE USE OF HANFORD WASTE WATER PONDS BY WATERFOWL

by

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Abstract

Census and environmental surveillance information on waterfowl that use the Hanford Site 200 Area waste water ponds are described and evaluated. Physical features of the ponds are discussed in relation to their use and suitability for waterfowl. Seasonal distributions observed for the years 1971 through 1974 indicate that the highest use by waterfowl occurs during the spring and fall migratory periods. Base population estimates are 300 to 400 resident waterfowl with a few tens of pairs nesting during the summer. Environmental surveillance data on ^{137}Cs in muscle tissue are presented for the years 1971 through 1977. Comparisons are made between Columbia River and waste water pond waterfowl, between waterfowl groups, and among ponds. Waterfowl collected from ponds frequently have easily detected levels of ^{137}Cs in muscle tissue. However, those waterfowl collected from the Columbia River seldom show a ^{137}Cs level above that expected from worldwide fallout. Waterfowl collected from the pond with the smallest ^{137}Cs inventory and the poorest waterfowl habitat contained the lowest levels of ^{137}Cs in muscle tissue.

Introduction

The purposes of this study were to:

- identify the waterfowl species that use the Hanford Site waste water ponds
- establish the abundance of waterfowl during all seasons of the year
- evaluate features of the ponds (vegetation, size, amount of human activity) for their attractiveness to waterfowl
- compare concentrations and variations of ^{137}Cs in muscle tissue between dabbling ducks and diving ducks and among ducks using different ponds and the Columbia River.

Study Areas

The study areas consisted of three shallow ponds located in the 200 Area of the Hanford Site. The waste water ponds are commonly referred to as U Pond, Gable Mountain Pond, and B Pond. A diagram of vegetation zonation around the ponds is shown in Figure 1.

U Pond is the smallest of the ponds studied (6 ha), and has received coolant water and other waste water since July 1944. In July 1972, a chemical herbicide was applied and immediately defoliated approximately 80% of the cottonwoods (*Populus* spp.) and willows (*Salix* spp.). Other species of vegetation, both terrestrial and aquatic, were less affected. Some of the trees, mostly cottonwoods, showed new growth by the next spring, and now about one-half of the

original live canopy has returned. The pond is characteristic of most ponds of the region in later stages of ecologic development. In addition to the usual patches of cattails (Typha latifolia) and reeds (Scirpus spp.), large willow trees and cottonwoods (10 m tall) grow along the shoreline. A dense cover of weedy vegetation ranging from 0.5 to 1.0 m tall and composed mostly of Russian knapweed (Centurea repens) and cudweed (Gnaphalium margaritacea) grows in the moist soil not occupied by trees. Human activity and disturbance from nearby heavy construction has been extensive.

Gable Mountain Pond is the largest of the ponds studied (29 ha) and has been used since December 1957. The water source is primarily coolant water that is discharged continuously. The pond has only a partial dike and has not been treated with herbicides to control riparian vegetation. Human disturbance is limited. This pond, in time, is expected to show the same advanced stages of vegetation zonation as is currently found at U Pond. Cattails and patches of reeds are extensive, and peachleaf willow trees (Salix amygdaloides) are widely scattered but smaller and younger than trees at U Pond.

B Pond, the second largest pond (19 ha), has been active since April 1945 and receives water from a coolant source and a chemical sewer ditch. Shoreline dike construction and herbicide applications in 1971 and 1972 destroyed most riparian and emergent aquatic plants. Reestablishment of shoreline vegetation has been slow. Some isolated reed patches occur in the pond shallows, and the terrestrial shoreline is ringed by a narrow zone of barnyard grass (Echinochloa crusgallii). Human activity at the pond is limited; however, the close proximity to the 200-E Area (about 1 km) adds a noise factor that is absent at Gable Mountain Pond.

Methods Employed

Weekly observations at the ponds were conducted from September 1971 to March 1974. Generally, waterfowl were observed during the morning hours while driving slowly around each pond, stopping at prescribed points, and scanning open water and border vegetation with binoculars. This technique, although not a complete census, revealed trends in abundance and species diversity. Also, human disturbance near each pond and meteorological conditions were noted in order to assess their possible effects on observed bird counts. Observation records for each census date were compiled for computer storage and retrieval.

Waterfowl are routinely collected as part of the Hanford environmental surveillance program. A 500-g sample of breast muscle is taken from each bird and a gamma spectroscopic analysis is performed on the fresh tissue. Various comparisons of the levels of ^{137}Cs in duck muscle tissue were made with the use of computer generated log-normal probability plots (Wa76).

Results and Discussion

Waterfowl belong to the order Anseriformes and are represented by the ducks, geese and swans. Ducks are often subdivided into dabbling ducks, diving ducks and mergansers. The dabbling ducks dabble and tip as they feed mostly on vegetation in shallow water. Diving ducks dive underwater when they feed on vegetation and varied forms of animal life. Mergansers dive for their food like the diving ducks but feed primarily on invertebrates and fish. For purposes of this discussion, the American Coot, order Cruiformes, has been included as

a waterfowl species. Coots may feed like dabbling ducks, but also will dive for submerged aquatic plants and invertebrates. All of these waterfowl are legally hunted, but geese and dabbling ducks are preferred game.

Seasonal Distribution and Abundance of Waterfowl

During the 33 months of weekly waterfowl censuses, a total of 20 duck species, the Canada goose, the Whistling Swan and American Coot were observed to use the Hanford waste ponds (Table 1).

The American Coot was the most abundant species observed to use the waste ponds; 19,141 of these birds were counted during the study. The Mallard was the most abundant dabbling duck (5,858 total observed), while the Ringnecked Duck was the most common diving duck (4,255 total observed). The total numbers of waterfowl observed were: 9,853 dabbling ducks, 12,437 diving ducks, 538 mergansers, 4,446 Canada geese, and 25 Whistling Swans.

Figures 2 and 3 show the weekly totals of dabbling and diving ducks, respectively. Figures 4 and 5 show weekly totals of Canada geese and coots, respectively. Note that the numbers of waterfowl in all groups display seasonal ups and downs typical of migratory waterfowl. Weekly fluctuations are also apparent and indicate that ducks, geese and coots frequently move on and off the ponds. The seasonal and weekly changes in duck and coot numbers indicate that few remain on the ponds as permanent residents.

Large numbers of dabbling ducks, geese and coots visit the Hanford waste ponds in the fall and midwinter (October through December) and in the spring (March through May). The Common Merganser is a visitor only in the fall and winter. During these migration periods, most individual waterfowl are likely to spend less than a few days or weeks associated with the ponds. It is equally likely, however, that some birds spend over a month associated with waste ponds. As shown by Figures 2, 3 and 5, a base population of about 100 dabbling ducks, 100 diving ducks and 150 to 200 coots occur on the waste ponds during the fall-early winter and spring migration periods. Nesting surveys each year indicate that between 10 and 30 pairs of dabbling ducks (Mallard, Pintail, Cinnamon Teal), no more than 10 pairs of diving ducks (Ruddy Duck, Redhead, and Lesser Scaup), and no more than 40 pairs of coots nested on the ponds.

The numbers of waterfowl that used the individual waste ponds can be estimated from Table 1. Gable Mountain Pond received the greatest use by all waterfowl groups. However, some species were observed more frequently at other waste ponds. Of the dabbling ducks for instance, shovelers preferred B Pond. In the diving duck group, goldeneyes and Bufflehead were most abundant on B Pond.

The reasons for these differences in pond use are unclear, but may be related to available food types, physical features, and human activity at each pond. The openness of B Pond and scarcity of emergent aquatic plants may be attractive to the three diving duck species. Canada geese may prefer Gable Mountain Pond because of nearby cheatgrass (Bromus tectorum) fields in which the geese were observed to forage. Coots and many of the dabbling ducks and diving ducks may have been more abundant on Gable Mountain Pond because of the large water surface area and the low degree of human activity. The Common Merganser's nearly total association with Gable Mountain Pond may be directly related to a relatively abundant food supply. This merganser feeds on fish, and Gable Mountain and U Ponds were the only ponds containing fish. Goldfish were somehow introduced

TABLE 1. Total Count of Waterfowl Observed on Hanford Waste Ponds, September 1971 Through March 1974

	B Pond	U Pond	Gable Mountain Pond
Dabbling Ducks			
Mallard	1390 (26)*	1373 (26)	2494 (48)
Gadwall	8 (1)	32 (5)	578 (94)
American Wigeon	80 (5)	482 (27)	1214 (68)
Green-Winged Teal	271 (48)	131 (23)	164 (29)
Blue-Winged Teal	24 (28)	39 (45)	23 (27)
Cinnamon Teal	29 (18)	37 (23)	93 (59)
Shoveler	326 (54)	120 (20)	159 (26)
Pintail	137 (17)	70 (9)	579 (74)
Total Dabbling Ducks	2265 (23)	2284 (23)	5304 (54)
Diving Ducks			
Redhead	199 (25)	25 (3)	568 (72)
Canvasback	0	4 (1)	570 (99)
Greater Scaup	293 (26)	120 (11)	694 (63)
Lesser Scaup	429 (42)	12 (1)	572 (57)
Ring-Necked Duck	951 (23)	113 (3)	3107 (74)
Common Goldeneye	626 (65)	24 (3)	299 (32)
Barrow's Goldeneye	39 (91)	4 (9)	0
Bufflehead	1870 (62)	61 (2)	1097 (36)
Old Squaw	1 (9)	0	10 (91)
Ruddy Duck	108 (14)	62 (8)	579 (77)
Total Diving Ducks	4516 (36)	425 (3)	7496 (61)
Mergansers			
Hooded	0	2 (67)	1 (33)
American	1 (1)	3 (1)	531 (98)
Canada Goose	926 (21)	0	3520 (79)
Whistling Swan	0	0	25(100)
American Coot	1257 (7)	330 (2)	17352 (91)

*Percent distribution among ponds

into these two ponds and a large supply is seasonally available to fish-eating waterfowl.

The rank of puddle duck and diving duck abundance observed at all ponds is shown in Table 2. A similar relative abundance has been reported in several studies of nearby ponds located off of the Hanford Site (Je48, Ha54, Jo56 and Yo60).

TABLE 2. Relative Abundance (Rank) of Ducks Observed on Hanford Waste Water Ponds

Puddle Ducks	Diving Ducks
1. Mallard	1. Ring-Necked Duck
2. Wigeon	2. Bufflehead
3. Pintail	3. Greater and Lesser Scaup
4. Gadwall	4. Common and Barrow's Goldeneye
5. Shoveler	5. Redhead
6. Green-Winged Teal	6. Ruddy Duck
7. Blue-Winged and Cinnamon Teal	7. Canvasback

Cesium-137 in Pond Environments

The concentration of ^{137}Cs in pond water at each of the study sites was estimated from averaging results of environmental surveillance samples collected quarterly from 1971 through 1977.[a] These values are shown in Table 3. Analytical standard errors for individual water samples may have been as large as ±100% and typical detection levels ranged from 30 to 50 pCi/ℓ through 1973. Detection levels were reduced to 3 to 5 pCi/ℓ, and standard errors were improved for later samples because of improved analytical techniques.

TABLE 3. Average Annual Concentrations of ^{137}Cs at Hanford Waste Water Ponds, pCi/ℓ

Year	B Pond	Gable Mountain Pond	U Pond
1971	36	51	79
1972	BDL*	42	220
1973	BDL	56	73
1974	16	31	BDL
1975	6.0	52	42
1976	BDL	31	8.0
1977	3.5	14	13

*BDL - Below Detection Limit

(a) See Ho78 for the most recent Hanford environmental surveillance report.

The majority of the ^{137}Cs inventory present at each pond is in the upper 5 cm of bottom sediments. Values reported for pond sediments are summarized in Table 4. A great deal of spatial variability was reported as noted in the range of concentrations. Very little information has been reported on the concentrations of ^{137}Cs in waterfowl food sources available from pond environments, but the levels are considered to be low.

TABLE 4. Sediment Concentrations of ^{137}Cs at Hanford Waste Water Ponds, pCi/g Dry-Wt

Pond	Average Concentration (Range)
U (Em74)	12000 (6000 - 21000)
Gable Mountain (Cu74)	30000 (13000 - 59000)
B	42*

*Single value

Cesium-137 in Waterfowl

Cesium-137 normally occurs in the flesh of waterfowl collected from Hanford waste water ponds. A small amount can be attributed to worldwide fallout, but some is undoubtedly contributed by the waste pond environment. The data presented in Figures 6 and 7 show that dabbling ducks on Hanford waste ponds (n = 53) had higher body burdens of ^{137}Cs than similar ducks on the Columbia River (n = 290). Dabbling ducks from waste ponds had body burdens an order of magnitude greater than dabbling ducks collected from the Columbia River. The solid-line curves are computer-generated least squares regression lines and 0.1 pCi/g is the approximate analytical detection level (ADL). The geometric mean (50%) for ducks collected on the Columbia River is below the ADL. However, the similarity in slopes of the regression curves for pond and river ducks indicate a similar degree of variability in each population.

Surveillance data for waste water ponds were not extensive enough to compare the ^{137}Cs levels in mergansers, geese or coots. However, ducks collected from the waste ponds were compared according to the ponds they used and according to dabblers versus divers. The most frequently collected dabbling duck was the Mallard, whereas Buffleheads and goldeneyes were the most common diving ducks. Figures 8, 9 and 10 are log-normal plots of ^{137}Cs concentrations for all ducks collected on Gable Mountain Pond (n = 32), B Pond (n = 24) and U Pond (n = 25). Again, 0.1 pCi represents the approximate analytical detection level. The slopes of the regression curves are virtually the same and indicate similar variability from pond to pond. The geometric means were approximately 39 pCi/g for Gable Mountain Pond, 4.6 pCi/g for B Pond, and 36 pCi/g for U Pond. Clearly, ducks collected from B Pond were less radioactive than ducks collected from Gable Mountain or U Ponds. The maximum ^{137}Cs levels reported for a duck from each pond were 210 pCi/g at Gable Mountain Pond, 260 pCi/g at B Pond, and 130 pCi/g at U Pond. The three high values (Figure 9) for B Pond do not follow the pattern for other collected ducks and may represent ducks that came from another waste pond. The two low values for Gable Mountain Pond (Figure 8) also are unusual and may represent recent immigrants from offsite.

Differences of ^{137}Cs in dabbling ducks and diving ducks for all ponds combined can be compared using Figures 7 and 11. The steep slope of the regression curve for diving ducks (Figure 11) indicates large variability (large spread) in the data and may be the result of a small sample size (n = 14). However, the maximum concentration levels for ^{137}Cs in dabbling ducks (≃130 pCi/g) is virtually the same as for the divers (≃120 pCi/g).

Conclusions

The abundance of surface water in the region of the Hanford Site attracts waterfowl. In addition, the isolation and lack of hunting pressure is conducive to waterfowl use of the Hanford waste water ponds. However, the ponds probably provide a better food source for diving ducks and coots than for dabbling ducks or Canada geese. Census data show that pond use by diving ducks and coots was greater than use by any other category of waterfowl. The reason for this greater use may be that dabbling ducks and geese have the option of feeding on terrestrial plants. Diving ducks and coots may use other local bodies of water, but the amount of interplay is unknown. Census data taken at the waste ponds indicate seasonally high use by waterfowl coinciding with fall and spring migratory periods. Base population estimates indicate 300 to 400 resident waterfowl, but nesting surveys noted only several tens of pairs.

Surveillance data on ^{137}Cs concentrations in dabbling ducks show a difference between those collected from the Columbia River and those collected from waste ponds. For example, a dabbling duck would be much more likely to contain a higher than normal ^{137}Cs concentration if it were collected from a waste pond. Seasonal differences in ^{137}Cs concentrations for any waterfowl were not evident.

Waterfowl use of ponds and ^{137}Cs concentrations in muscle tissue seem to be related to pond size, degree of human disturbance, ecological stage, and sediment concentration of ^{137}Cs. The concentration of ^{137}Cs in waterfowl flesh was greatest at U Pond and Gable Mountain Pond where ^{137}Cs sediment concentrations are highest. The high degree of human disturbance at U Pond may offset the advantage of an advanced stage of ecological development and result in lower waterfowl use than at other ponds. B Pond represents a poor habitat, but its large size seems to attract waterfowl, especially diving ducks. The fact that there was little difference in the maximum level of ^{137}Cs in birds collected at each pond indicates that waterfowl probably fly between ponds.

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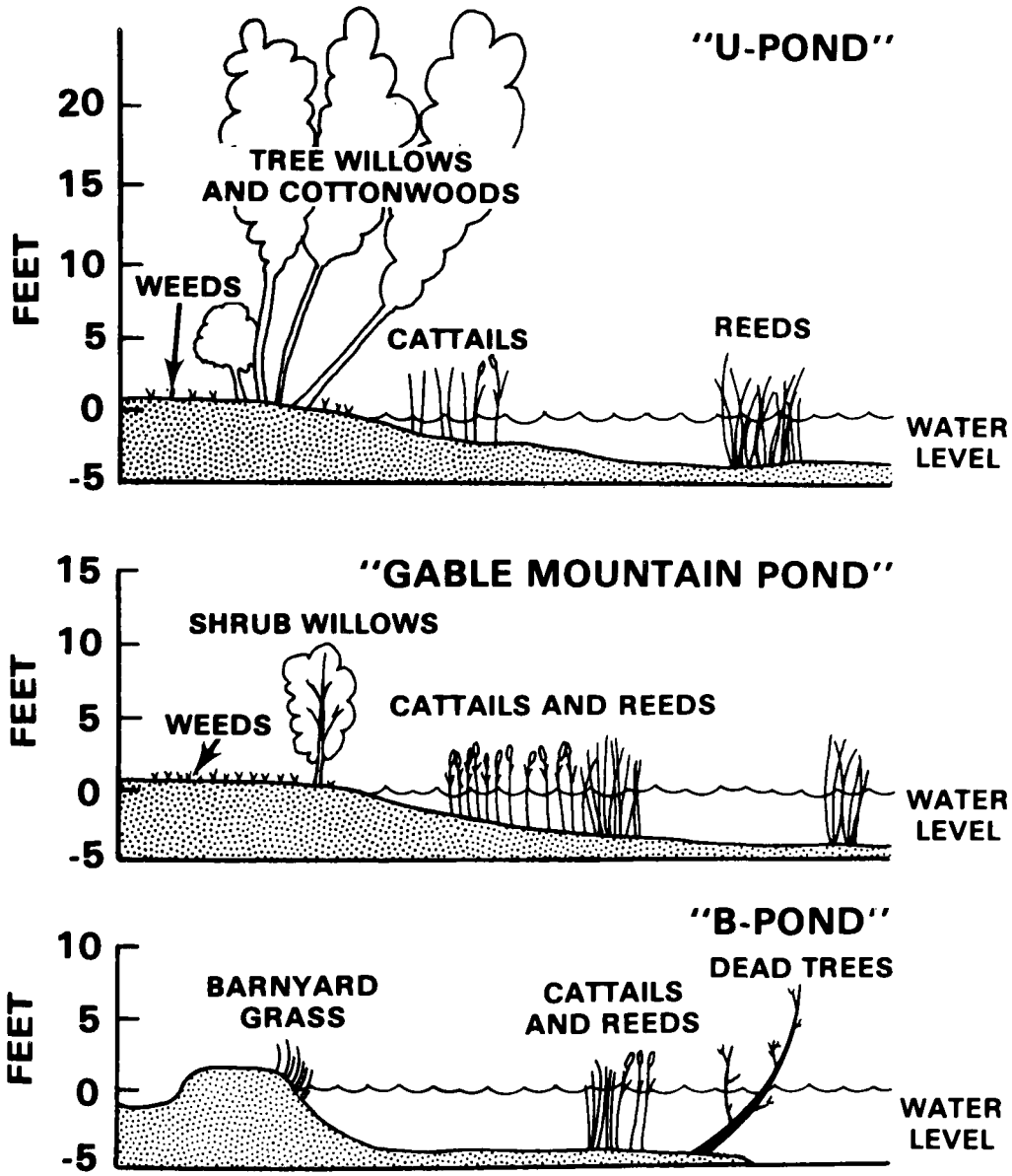

FIGURE 1. Vegetation Zonation Around Hanford Waste Water Ponds

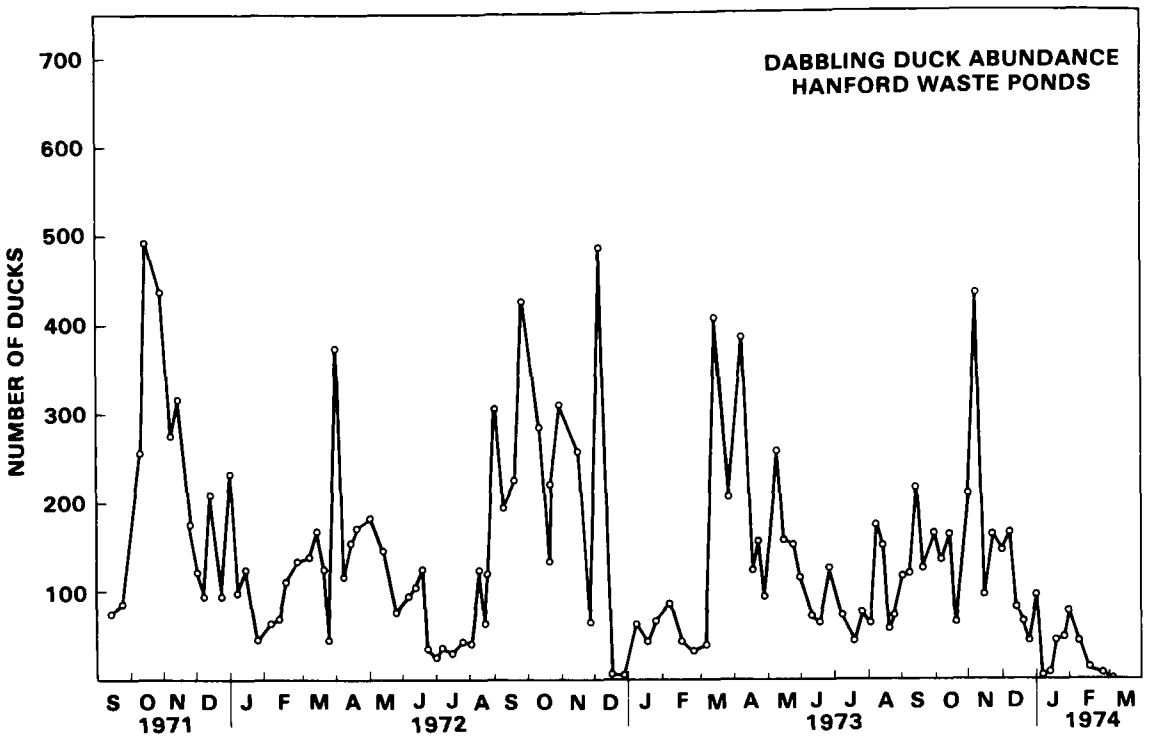

FIGURE 2. Weekly Totals of Dabbling Ducks Observed at Hanford Waste Water Ponds

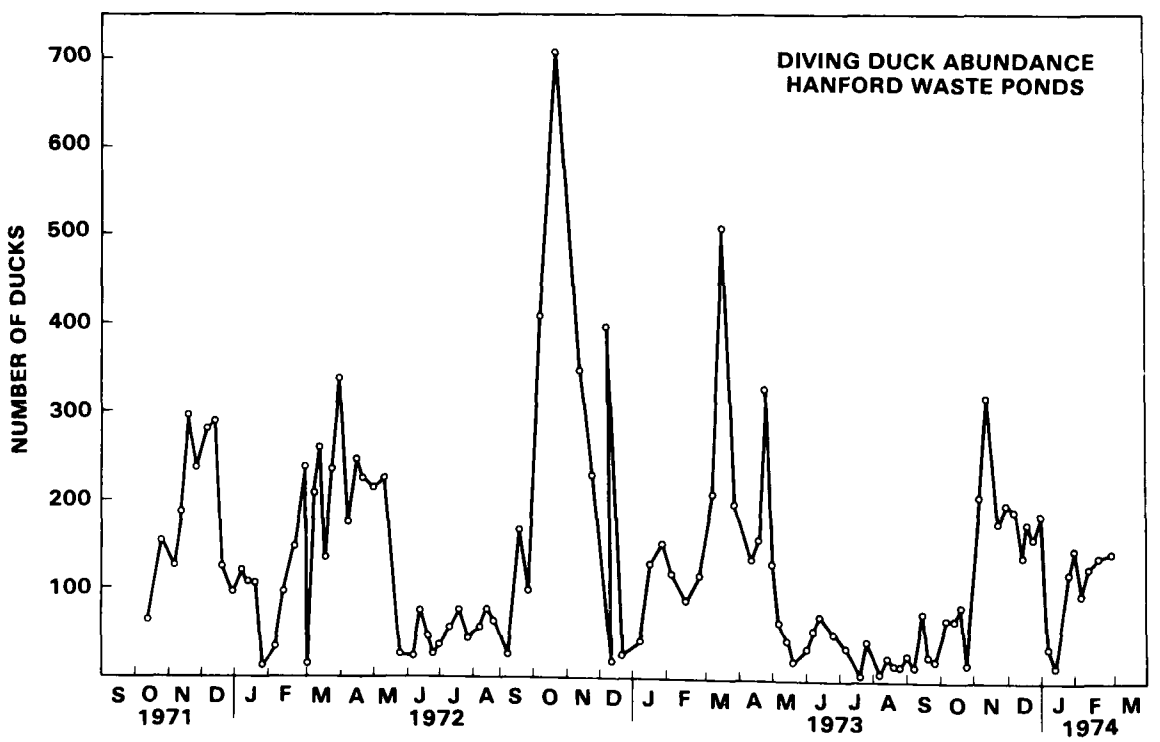

FIGURE 3. Weekly Totals of Diving Ducks Observed at Hanford Waste Water Ponds

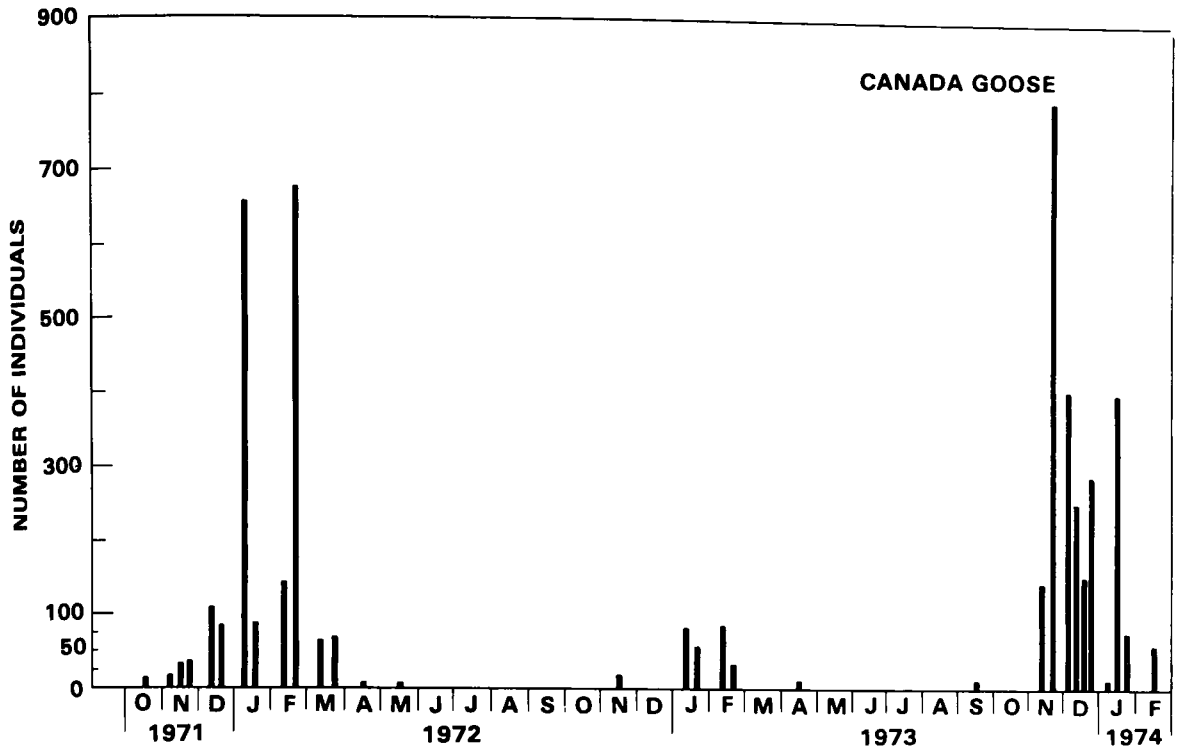

FIGURE 4. Weekly Totals of Canada Geese Observed at Hanford Waste Water Ponds

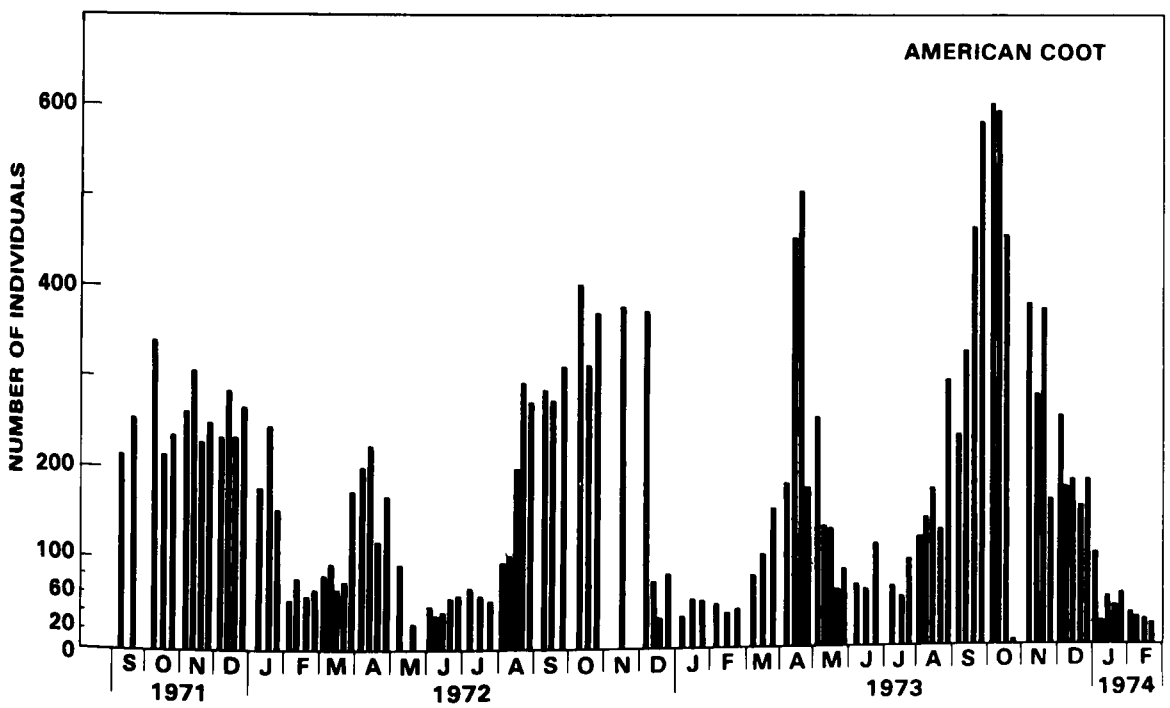

FIGURE 5. Weekly Totals of Coots Observed at Hanford Waste Water Ponds

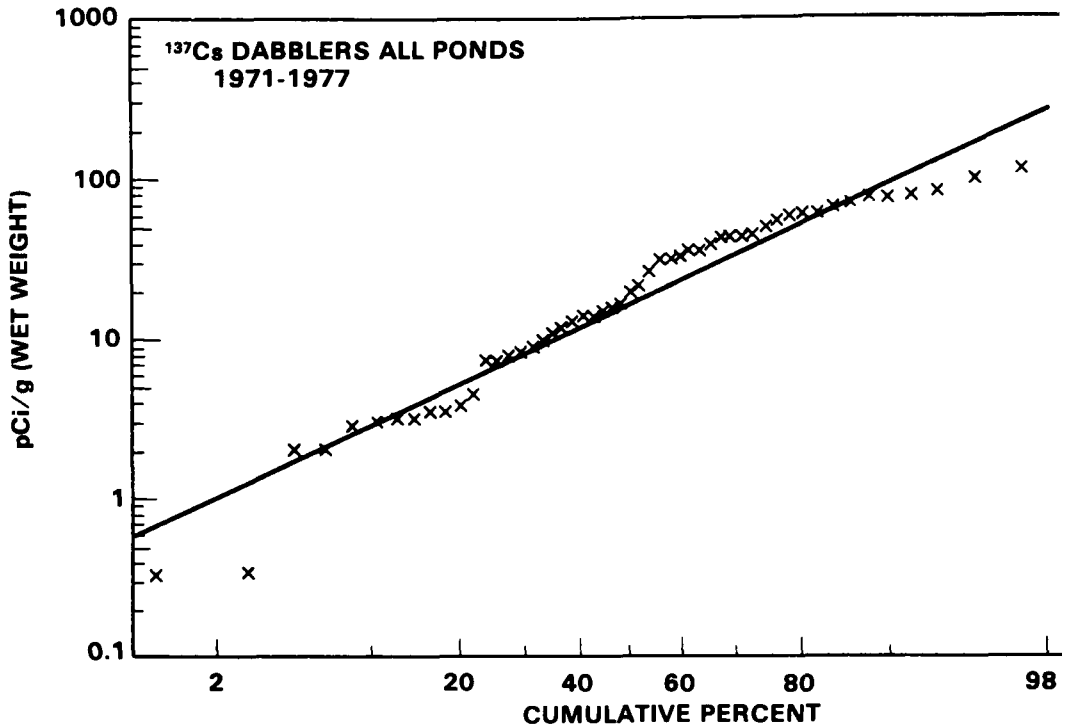

FIGURE 6. Cesium-137 Concentration in Dabbling Ducks Collected from Hanford Waste Water Ponds

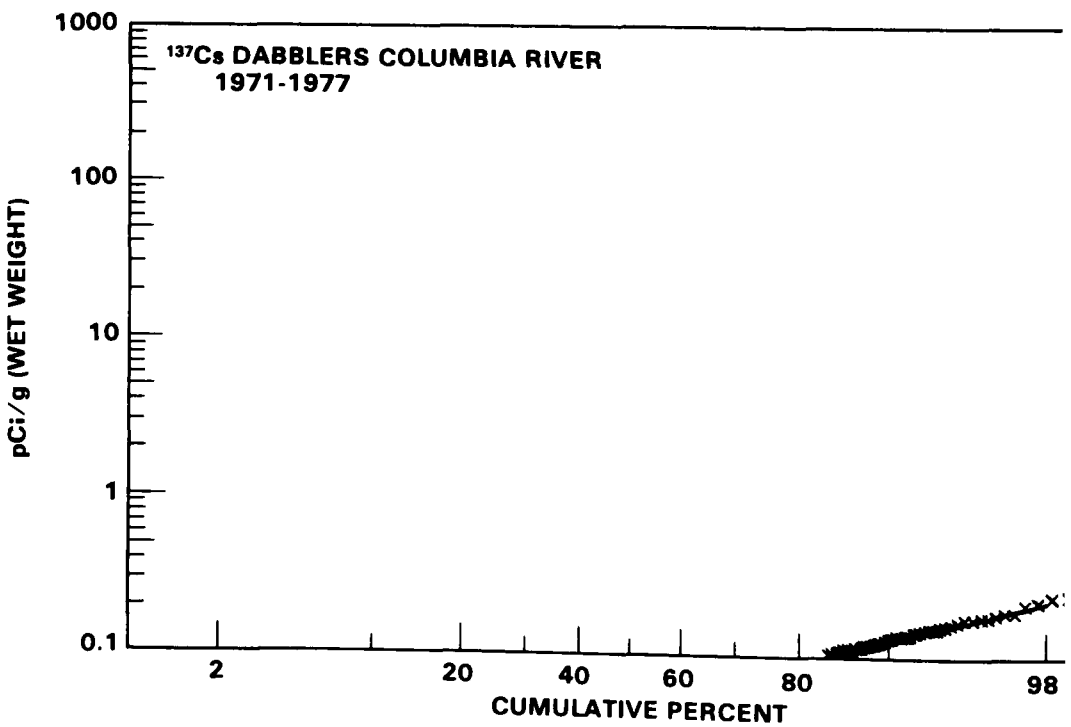

FIGURE 7. Cesium-137 Concentration in Dabbling Ducks Collected from the Columbia River

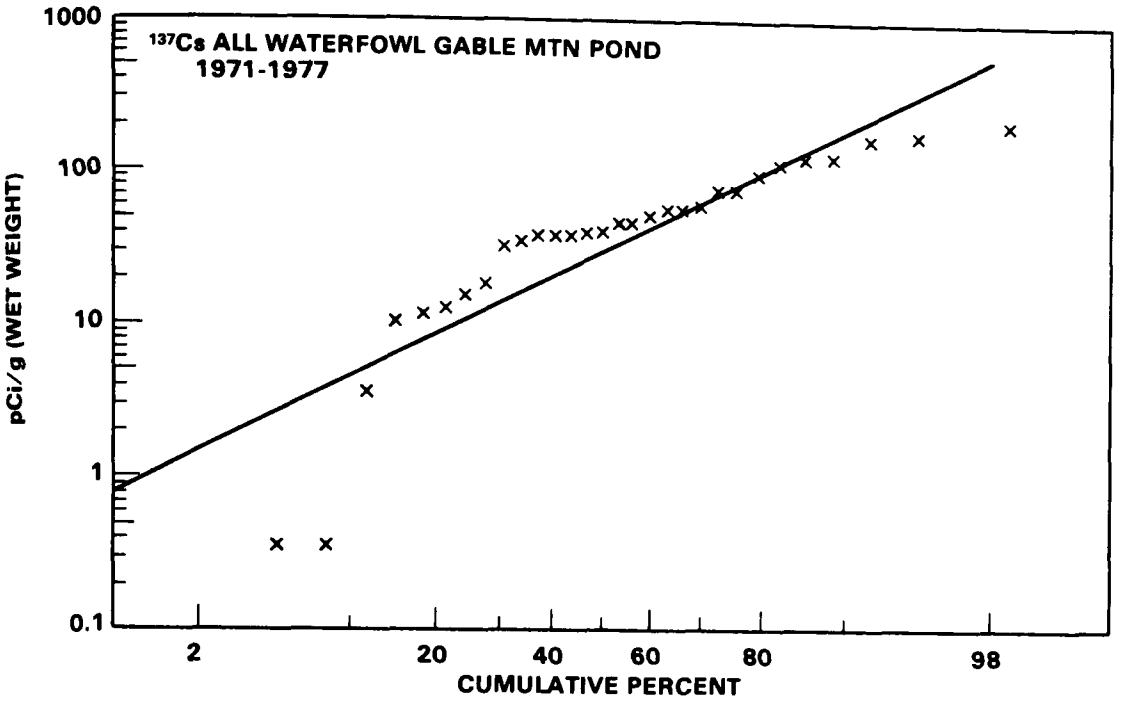

FIGURE 8. Cesium-137 Concentration in All Ducks Collected from Gable Mountain Pond

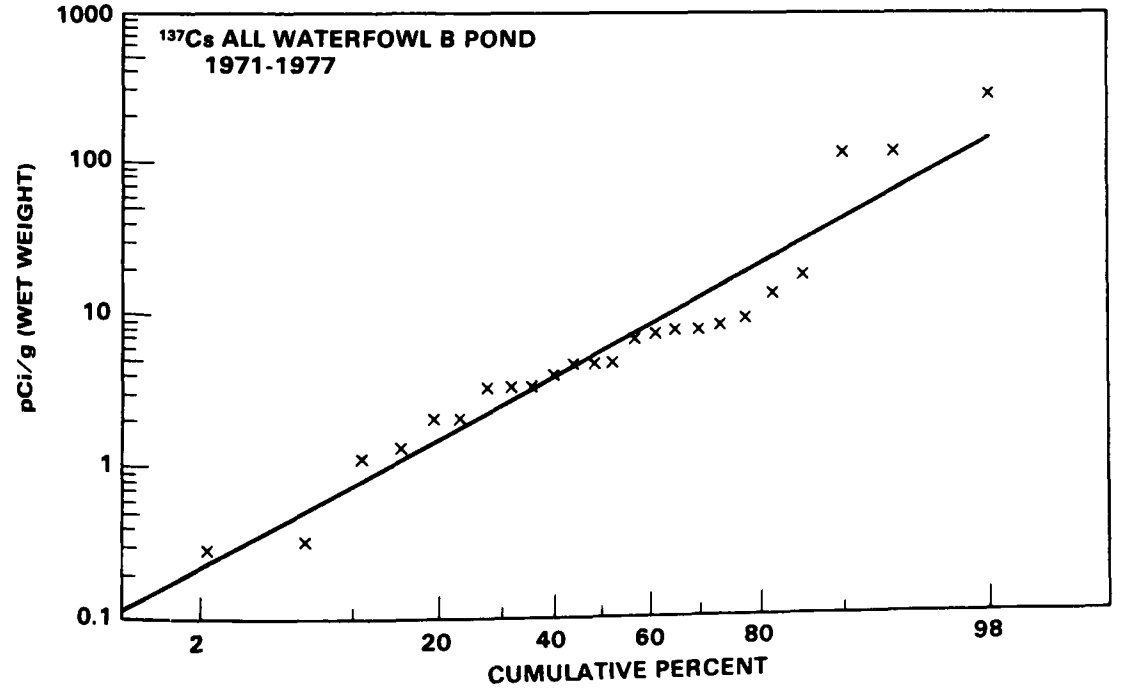

FIGURE 9. Cesium-137 Concentration in All Ducks Collected from B Pond

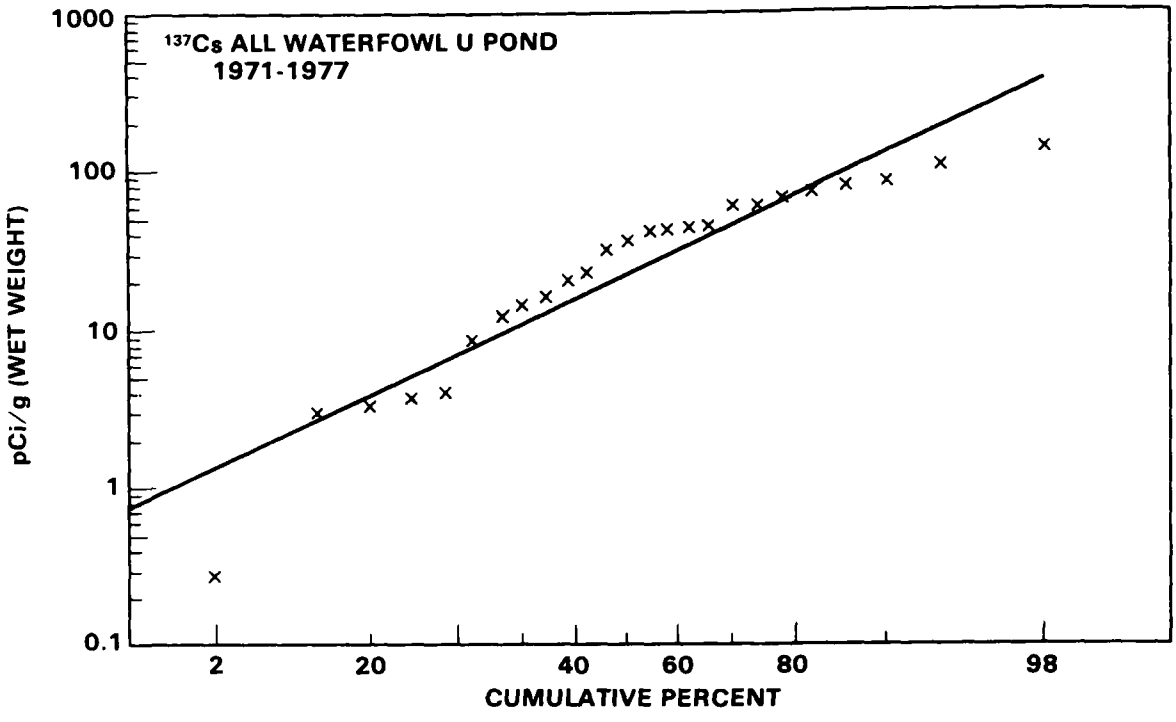

FIGURE 10. Cesium-137 Concentration in All Ducks Collected from U Pond

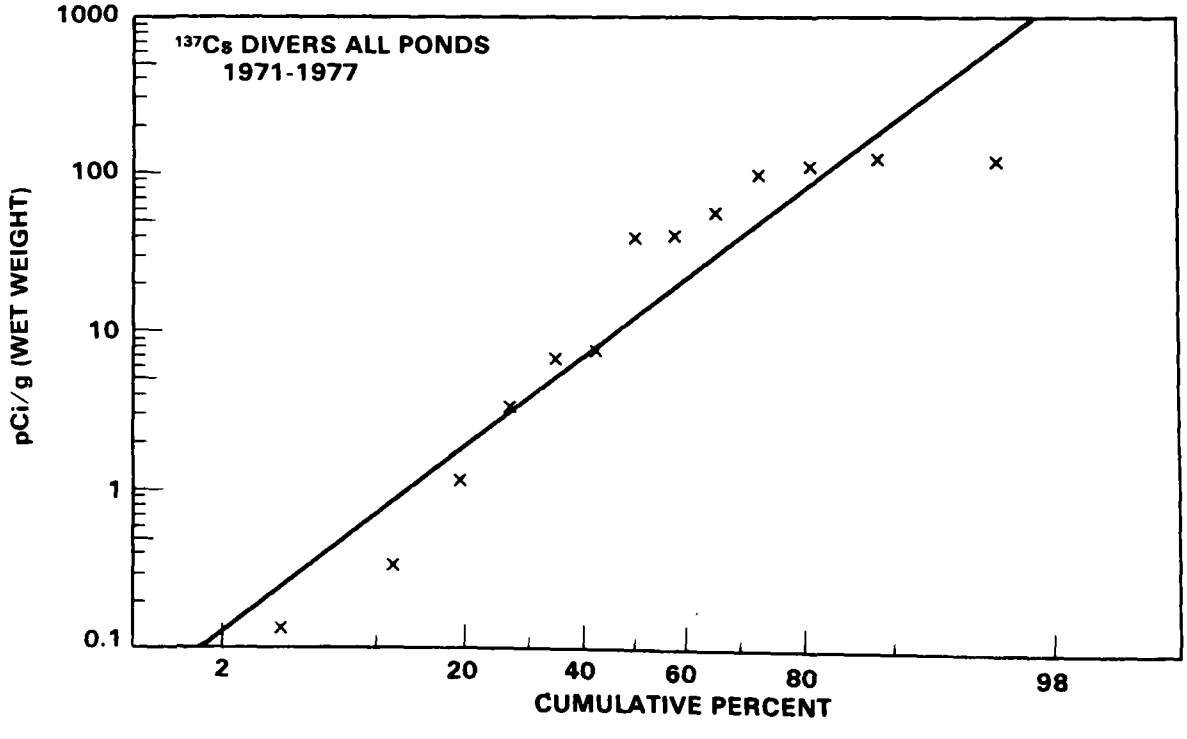

FIGURE 11. Cesium-137 Concentration in Diving Ducks Collected from All Ponds

CESIUM-137 IN COOTS *(Fulica americana)* ON HANFORD WASTE PONDS: CONTRIBUTION TO POPULATION DOSE AND OFFSITE TRANSPORT ESTIMATES

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Abstract

American coots *(Fulica americana)* were periodically collected from ponds receiving low-level radioactive waste on the Hanford Site and from ponds on a control area. Gut contents and selected tissues were removed and analyzed for ^{137}Cs, ^{90}Sr and gross Pu. The concentration of ^{137}Cs in coot flesh was the highest of the radioelements measured. Coots from the pond receiving the greatest quantity of fission products had the highest ^{137}Cs concentrations. One- and fifty-year ^{137}Cs total-body dose commitments to an individual from ingesting all edible tissues of one Gable Mountain Pond coot were calculated at 1.9 and 2.1 mrem, respectively. The 50-year population dose from the projected harvest of Gable Mountain Pond coots during one year was estimated to be 0.13 person-rem, which is likely high because the coots must leave the Site to be harvested. Total ^{137}Cs export from Hanford Site ponds via coots, having an average body burden of 0.092 μCi, was estimated to be 46 μCi per year.

Introduction

Process ponds located on the Department of Energy's Hanford Site in southcentral Washington contain low levels of radioactive wastes. Three ponds currently in use contain mixtures of fission and activation products including transuranic elements. The ponds presently support a variety of plant and animal life. Waterfowl use the ponds extensively and are capable of transporting radioactive materials offsite. Should waterfowl leave the waste ponds and be harvested by hunters, they may become part of the regional human food supply.

This paper evaluates the role of coots in exporting radionuclides from the ponds and for the dose to man resulting from ingestion. Although a number of waterfowl species use the ponds, the American coot *(Fulica americana)* is the most abundant (Fi75). The breeding population was estimated to hatch as many as 200 young per year, 40 of which may reach flight age (Fi79). In addition to the breeding population, migrant coots use the ponds during fall and winter. Peak numbers of coots on the ponds have reached nearly 600. Beyond being the most abundant waterfowl species on the ponds, concentrations of ^{137}Cs in coot flesh are consistently higher than in other waterfowl (Cu74, Fi77).

Although the coot is not necessarily a preferred game species, it is relatively abundant, easy to bag, and a part of the waterfowl hunters' harvest. During the 1973 hunting season, the "retrieved kill" data for Washington State waterfowl hunters included 8,104 coots, which comprised 1.3% of the total bag for all waterfowl species (US78).

The Study Area

The ponds are used for disposing of process waste water by percolation and evaporation. They have undergone ecological succession since their formation and have shoreline vegetation that provides suitable nesting habitat and food supply for waterfowl (Fi75).

Gable Mountain Pond, with a 29 hectare surface area, is the largest of the three ponds. Constructed in 1957, this pond contains the greatest quantity of fission products, primarily as a result of an accidental release in 1964 (An74). B-Pond, in existence since 1945, has a surface area of approximately 19 hectares. It contains the smallest quantities of radioactive materials as the result of minor unplanned releases. U-Pond is the smallest with an area of about 6 hectares. It was formed in 1944 and receives waste water from a laundry facility that contains low levels of radionuclides. Of the three ponds it contains the smallest quantity of fission products but the greatest amount of plutonium.

Methods

One hundred three coots were collected from Gable Mountain Pond, 18 from U-Pond and 31 from B-Pond. An additional 13 coots were collected from the Columbia Wildlife Refuge in central Washington to serve as controls to determine fallout concentrations of radionuclides. Collections were made at 1 to 2 month intervals from June, 1974 through March, 1976. An additional collection was made in January, 1977. Five to nine birds were collected during each sampling period.

The birds were whole-body counted to determine ^{137}Cs content, then they were weighed, dissected and selected samples were removed for radiochemical analysis. The samples included muscle, liver, bone and contents of the intestinal tract. Samples were analyzed for gamma-emitting radionuclides with a sodium iodide detector and multichannel analyzer. Selected samples were also sent to a commercial laboratory where they were analyzed for ^{90}Sr and gross Pu.

Results and Discussion

The concentrations of ^{90}Sr, ^{137}Cs and gross Pu for selected coot sample types from birds collected at Gable Mountain Pond are summarized in Table 1. These data show clearly that ^{137}Cs occurred in greatest concentration in tissue from Gable Mountain Pond coots. Additional plutonium analyses from U-Pond coot samples were made because of the high Pu content in U-Pond relative to the other two ponds. These analyses showed the Pu concentrations in muscle from U-Pond coots were only slightly greater than for Gable Mountain Pond birds, but still insignificant in comparison with ^{137}Cs (Table 1).

Table 1. Average Concentration of ^{90}Sr, and ^{137}Cs and Gross Pu in Selected Tissues of Coots Collected at Gable Mountain Pond

Sample Type	Average Dry Weight per Coot (g)	Concentration (pCi/g dry weight)					
		^{90}Sr (n = 12)		^{137}Cs (n = 103)		Gross Pu (n = 24)	
		x	SE	x	SE	x	SE
Bone	24.0	2.6	0.6	200.	30.	0.023	0.006
Liver	4.3*	0.53	0.20	440.	40.	0.052	0.031
Muscle	59.0*	0.28	0.12	570.	40.	0.019	0.015
Gut Contents	3.0	4.3	1.5	3,400.	200.	0.14	0.03

*Multiply by 2.9 to convert to wet weight.
n = 16 for muscle only.

Table 2. Average Concentration of ^{137}Cs in Samples of Coots from the Study Areas

Sample Type	Concentration (pCi/g dry weight)							
	Gable Mountain Pond (n = 103)		U-Pond (n = 18)		B-Pond (n = 31)		Columbia Wildlife Refuge (n = 13)*	
	x	SE	x	SE	x	SE	x	SE
Bone	200.	30.	70.	10.	5.7	0.8	1.0	0.3
Liver	440.	40.	220.	20.	16.	2.	0.7	0.2
Muscle	570.	40.	360.	30.	30.	4.	0.02	0.05
Gut Contents	3,400.	200.	1,300.	200.	85.	11.	0.8	0.5

*Columbia Wildlife Refuge concentrations were near or below detection limits (∿0.5 pCi/g), which varied with sample size.

Concentrations of ^{137}Cs in samples from all three ponds are shown in Table 2. Gable Mountain Pond coots were highest for all sample types. The significance of the radionuclide inventory in birds from the other two ponds is reduced when one also considers that over 90% of the total number of coots observed on the Site were on Gable Mountain Pond (Fi75).

Coots from Gable Mountain Pond were sampled periodically to determine whether there were any seasonal trends in their ^{137}Cs content. Figure 1 shows no clear-cut pattern for ^{137}Cs concentration in muscle, but there was also a high degree of variability in concentrations among samples for any one collection date. Part of the reason for the variability is probably due to the fact that some coots sampled were recent arrivals and thus had not attained muscle concentrations near equilibrium. A ratio (^{137}Cs concentration in the gut/^{137}Cs concentration in the muscle) was calculated as an index of whether or not the birds sampled were recent arrivals. Since the gut content reflects the concentration of ^{137}Cs in the food, it readily equilibrates and will show a high concentration within two or three days. Muscle, on the other hand, requires longer to reach an equilibrium concentration. The time required is proportional to the biological half-time for ^{137}Cs in muscle. Although the muscle half-time for ^{137}Cs in coots is unknown, it is probably similar to that reported for other waterfowl. Fendley et al. (Fe77) reported a ^{137}Cs half-time of 5.6 days for wood ducks, and Halford et al., (Ha78) reported a

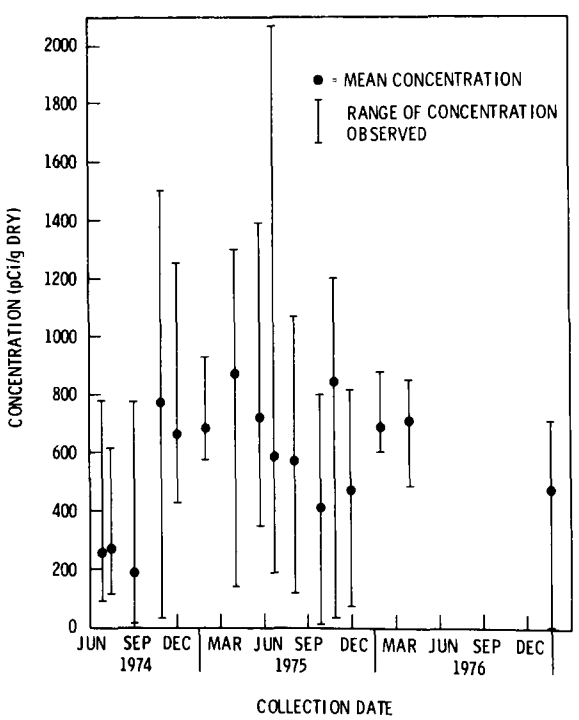

Figure 1. ^{137}Cs concentration in muscle of coots from Gable Mountain Pond (June 1974 - January 1977)

value of 11.7 days for mallards. Therefore, one would expect that it would take from 1 to 2 months, or approximately 5 half-times, for the concentration of ^{137}Cs in coot muscle to equilibrate. Newly arrived birds, with high gut to muscle ratios can be distinguished from birds which have been on the pond long enough for muscle concentrations to approach equilibrium. A plot of the gut to muscle ratio for all Gable Mountain Pond coots (Figure 2) shows that the ratios for most of the birds was clustered in the range of 3 to 10. The ratios from about 10% of the coots was considerably higher (above 20). Our interpretation is that those birds were relatively new arrivals. The highest ratios occurred during the months of September and October which correspond with the expected influx of migrant birds. These months also overlap the hunting season for waterfowl in Washington. Thus, average muscle concentrations for the year, (used here for dose to man calculations) should provide upper-limit dose estimates since part of the population harvested during the hunting season may not have muscle concentrations at equilibrium.

The total-body 50-year dose commitment (D) from ^{137}Cs to an individual ingesting all edible tissue from one coot (0.03 μCi) was calculated according to Morgan and Turner (Mo67) using parameters from ICRP #10 (In68):

$$D = \frac{(51\ \frac{\text{rem}\cdot\text{dis}\cdot\text{g}}{\text{Mev}\cdot\text{day}\cdot\mu\text{Ci}})(E)(f_w)(0.03\ \mu\text{Ci})}{M} \int_0^{18250} e^{-\left[\frac{0.693(18250\ \text{days})}{T_E}\right]} dt$$

$$= 2.1 \text{ mrem}$$

where, E = effective energy for ^{137}Cs (0.59 Mev/dis);
M = total body mass (7 x 10^4 g);
f_w = fraction of ingested ^{137}Cs reaching organ of reference (1.0); and,
T_E = effective half-time of ^{137}Cs in total body (114 days).

A similar calculation of a one-year dose gives approximately 1.9 mrem. Thus, a member of the general public consuming one coot would receive about 2% of the dose an individual receives from natural background sources of approximately 100 mrem/year (Ei73). This would also be equal to 1.1% of the U.S. Department of Energy's radiation protection standard of 170 mrem/year for individuals and population groups in uncontrolled areas (US77).

Harvest data for coots in the State of Washington for the 1973 hunting season show that hunters bagged approximately 8,100 birds (US78). Breeding season population estimates for the same year put the state coot population at 37,240 birds. The ratio of retrieved harvest to population estimates, suggest that approximately 22% of the state population may have been harvested. An average of 300 coots were observed on the Hanford area waste ponds during the 1973 hunting season (Fi75). If one assumes that 22% were bagged, then approximately 70 coots might have been harvested. Since about 91% of the Hanford area coots were on Gable Mountain Pond (Fi75), then the harvest

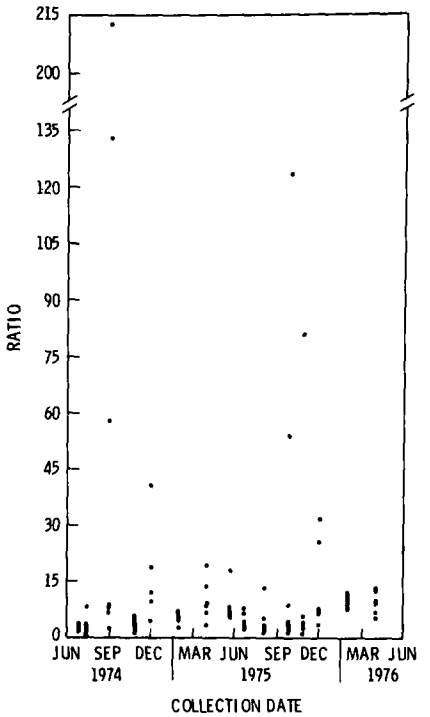

Figure 2. Ratio (gut content/muscle) for ^{137}Cs concentration in coots from Gable Mountain Pond

estimate for that pond for 1973 is 63 birds. The 50-year dose commitment to the general public (waterfowl hunters and their families) from consuming the edible portion of 63 Gable Mountain coots (1973 data) was 0.13 person-rem. The harvest estimate and subsequent dose calculations from Gable Mountain Pond coots is very likely high. It is unlikely that coots using the Hanford area ponds will be harvested at a rate as high as the state average. Coots must leave the confines of the Hanford Site to be available to hunters.

The movements of coots from Hanford area waste ponds have not been investigated extensively enough to accurately determine the quantity of ^{137}Cs transported offsite. However, the amount of ^{137}Cs available for annual export is the product of the coot whole-body burden and the number of coots that leave the ponds each year. If one examines the data reported by Fitzner and Rickard (Fi75) and assumes that the reduction in birds observed at succeeding observations reflects the movement of birds away from the ponds, then the number of birds leaving the ponds averaged about 500 coots/year during 1972 and 1973. The coot body burdens were 0.10 ($\pm$ 0.01), 0.05 ($\pm$ 0.01) and 0.004 ($\pm$ 0.001) μCi for Gable Mountain, U-, and B-Ponds, respectively. The average body burden, weighted according to relative abundance of coots for the three ponds, was 0.092 μCi. Therefore, the annual export of ^{137}Cs from the ponds could be about 46 μCi/year (0.092 μCi/coot x 500 coots/year).

Acknowledgements

We thank Mr. H. A. Sweany for his technical assistance during the study. The study was supported in part by Rockwell Hanford Operations' Long-Term Management of Low-Level Waste Program and in part by the Department of Energy Office of Health and Environmental Research.

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RESPONSE TO A WIDESPREAD, UNAUTHORIZED DISPERSAL OF RADIOACTIVE WASTE IN THE PUBLIC DOMAIN

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Abstract

In March 1976 State of Nevada radiological health officials became aware that radioactive items destined for disposal at a radioactive waste burial facility near Beatty, Nevada had instead been distributed to wide segments of the public domain. Because the facility was jointly licensed by the State of Nevada and the federal Nuclear Regulatory Commission, both agencies quickly responded. It was learned that over a period of several years a practice existed at the disposal facility of opening containers, removing contents and allowing employees to take items of worth or fancy. Numerous items such as hand tools, electric motors, laboratory instruments, shipping containers, etc., had received widespread and uncontrolled distribution in the town of Beatty as well as lesser distributions to other locations. Because the situation might have had the potential for a significant health and safety impact, a comprehensive recovery operation was conducted. During the course of seven days of intense effort, thirty-five individuals became involved in a comprehensive door by door survey and search of the town. Aerial surveys were performed using a helicopter equipped with sensitive radiation detectors, while ground level scans were conducted using a van containing similar instrumentation. Aerial reconnaissance photographs were taken, a special town meeting was held and numerous persons were interviewed. The recovery effort resulted in a retrieval of an estimated 20-25 pickup truck loads of radioactively contaminated equipment as well as several loads of large items returned on a 40-foot flatbed trailer.

Discussion

In February 1976, the Radiation Safety Officer (RSO) at a low-level radioactive waste land disposal area near Beatty, Nevada noticed that the company's stockpile of bagged cement was diminishing disproportionately to the quantity of liquid waste being solidified. The RSO, who was relatively new to the site, found on investigation that some company employees had been using the facility's cement mixing truck and materials to pour concrete on several different jobs in the nearby town of Beatty. The truck was routinely used to solidify liquid radioactive waste onsite and was internally contaminated. The company reported this matter to the State of Nevada, radiological health officials.

It was not uncommon for uncontaminated site equipment, road graders, etc., to be used in assisting area residents and on civic projects. To better understand the events which occurred at and around the disposal site, it is essential that one understand the somewhat unique characteristics of the locale. Beatty is a small, remote desert town of approximately 500 residents located about 125 miles north of Las Vegas, Nevada. The area is bordered on the west by the

Death Valley National Monument and on the east by the U.S. government's Nevada Test Site. Employment in the area generally comes from the Nevada Test Site, a few tourist-related businesses, ranching, mining and the waste burial facility. Many residents of the town were employees or former employees of the waste facility or were relatives or friends of these people. Because of the difficulty and cost of obtaining materials in such a remote location, the people of Beatty appeared reluctant to part with any item that could have salvage value. Many residences were observed to have accumulated stockpiles of salvageable items including lumber, tires, automobiles, appliances and a myriad of other materials. It was in this isolated realm of self-sufficiency, that items destined for disposal at the waste facility found their way into the public domain.

The waste facility itself is a low-level radioactive waste burial operation, which in 1976 was jointly licensed by the State of Nevada and the U.S. Nuclear Regulatory Commission (NRC). The State regulated the disposal of source, by-product and natural occurring materials while the NRC's regulatory activities were limited to special nuclear materials (SNM). Materials shipped for disposal came from a wide variety of users of radioactive materials including military, medical, industrial and research facilities. A large quantity of material arriving at the site for disposal consisted of equipment that became contaminated during usage. Typical items included protective clothing, tools of all kind, laboratory equipment and instruments, electric motors, piping, wiring and other paraphernalia. In addition, a large quantity of radium dial clocks, watches, compasses, assorted gauges and other similar obsolete military equipment were received at the site for disposal over the years.

During their investigation of the cement mixer occurrence, State officials discovered that other possibly contaminated materials had been removed from the disposal site in past years and were distributed in the town of Beatty. The State quickly responded by suspending the company's license on March 8, 1976 and halting all State licensed operations at the site (similar action was subsequently taken by NRC). The State also requested radiological assistance from the Environmental Protection Agency's (EPA) Las Vegas facilities to help locate contaminated materials in the town of Beatty. The request was made to EPA because of their proximity, their monitoring capabilities and because of the existence of professional acquaintances. The State also notified NRC which promptly dispatched an inspector to the site.

NRC's initial role was in an investigative capacity only, because there had been no direct evidence of the release from the site of articles contaminated with SNM. However, early survey efforts did identify the presence of a plutonium contaminated triple beam balance and weights in a private residence. It was disclosed that the scale and weights had come from the disposal site, thereby involving the SNM license and creating a situation under joint State and NRC jurisdiction. The State and NRC subsequently joined their resources in a cooperative effort.

Through detailed investigation it was disclosed that during the period of 1967 to 1973 it was common practice at the site to open containers of radioactive waste, remove the contents and allow employees to take what they wanted. It was disclosed that over a period of time, mostly through trial and error, employees were able to determine which containers held desirable items. This could be determined in large measure by the location from which the containers were received. Another method employed to determine whether a container

was to be opened was to roll it over and if it rattled, it was a good indication that the contents were of value. Such containers came to be referred to as "rattlers". From information gathered, it was apparent that a myriad of items destined for disposal had actually been diverted into the public domain. Plywood shipping containers were routinely disassembled to salvage the wood. Numerous radium dial objects were taken. Contaminated tools of all kinds were diverted. It was apparent that any item of practical use or of just human interest had the potential for being diverted. Interrogation of involved individuals revealed that only minimal radiation surveys were performed on materials removed from the site.

One item of particular interest which had been routinely diverted was a large (about 200 cubic ft.) cylindrical steel shipping container referred to as a "Bennett Bucket." These containers were fabricated and used for waste shipments by a large laboratory involved in the use of large quantities of radioactive materials including transuranics. Upon receipt at the disposal site, the contents of these containers were usually dumped into a disposal trench and the containers were diverted for such uses as septic tanks, animal grain and water storage and as storage tanks for both potable and nonpotable water. From detailed investigation it was determined that approximately 141 Bennett Buckets were shipped for disposal during the 1967 to 1973 time period. It was estimated that about 95% of these were opened and diverted from disposal.

On the afternoon of March 10, EPA personnel began to conduct radiation surveys in the town of Beatty and on outlying ranches. Systematic assignments were made to assure complete survey coverage. Because the majority of the town's streets were unnamed and maps did not exist, aerial photographs were used to identify locations. A "Statement Of Authority For Investigation" was issued by the Nevada State Health Officer. The Statement delineated applicable Nevada statutes that authorized the entrance upon private property and authorized the impoundment of materials found to be radioactive. Copies of the Statement were carried by personnel performing surveys. However, the legal authority was not frequently imposed as the vast majority of residents were cooperative in allowing surveyors to enter their property and, if applicable, remove contaminated or radioactive items.

The principal survey instrument used was a sensitive, fast response gamma scintillation instrument (He76). Other survey instruments, including ones with alpha detection capability were available and used as circumstances dictated. Surveyors worked on an individual basis, carefully surveying each room of residences, as well as yards, garages, storage sheds, business establishments and the piles of salvable items prevalent throughout the town. General ground rules were to perform a thorough gamma survey, make note of any suspicious looking items for followup alpha surveys, only pick up items exhibiting positive indications of radioactivity, and make note of identified contaminated items which were not picked up in order to allow later pick up. By the evening of March 10, it became apparent that large quantities of contaminated material were present in the town as pickup truck loads of items were identified and recovered.

On March 11, 1976, NRC issued an order to the disposal company suspending NRC licensed activities at the facility. In addition, under terms of the Interagency Radiological Assistance Plan (IRAP), NRC requested radiological assistance from the Energy Research and Development Administration (ERDA, now

Department of Energy DOE) to assist in the comprehensive radiation surveys already initiated in the town of Beatty. Additional NRC personnel were dispatched to the site to assist. NRC inspectors were also dispatched to three other disposal sites to determine whether a similar problem might exist at those sites. The State of Kentucky was notified because of a site there under sole State jurisdiction. It was found that the situation was unique to the Beatty site.

On the afternoon of March 11, an NRC representative in Beatty met with ERDA representatives to brief them on the scope of the problem. ERDA responded with a team consisting of ERDA employees and personnel from three ERDA contractors (EG&G, Inc., Lawrence Livermore Laboratory (LLL), and Reynolds Electrical and Engineering Co. (REECO)). On the morning of March 12, EPA and ERDA survey efforts were quickly intergrated with overall responsibility being jointly shared by the State of Nevada and NRC. Three-man survey teams were formed consisting of two personnel performing alpha surveys and one performing gamma surveys. In addition, an EG&G helicopter and van, both equipped with a large array of sodium iodide gamma detectors, were used to survey outlying ranches and dispersed areas as well as locations in town (Jo76). A LLL van equipped with a portable GeLi detector and multichannel analyzer was used for isotopic identification of samples and to perform in situ analyses where necessary (An76). A REECO sodium iodide gamma detector with a multichannel analyzer and a portable gas proportional beta/alpha detector with a scaler were set up in a Beatty motel being used as a "command post" (Re76). A base of radio communications was established at the "command post" and walkie-talkies were used for field communications. Overall, approximately 35 individuals had become involved in the survey effort.

By Friday, March 12, survey teams began to hear rumors that some material was being buried, disposed of in the desert or otherwise hidden by residents, apparently out of fear of being "caught with materials." To counter this and to establish better public relations, State officials called a town meeting that evening. Notice of the meeting was spread by the county sheriff's office. About 130 residents attended the meeting which was held in the school gymnasium. The local residents were informed of the situation and told that the agencies were principally interested in recovering the materials and assuring that there was no health hazard. The townspeople were assured that no action would be taken against them. Residents were afforded the opportunity to ask questions. Generally, the tone of questions from townspeople was serious and concerned. The atmosphere was good natured and friendly. The meeting lasted about one hour. Results of the town meeting were evident on the ensuing weekend as people voluntarily returned miscellaneous items.

Survey efforts continued on March 13 and 14 and by the evening of March 14, the bulk of work had been completed and the survey teams were disbanded. Approximately 20 to 25 pickup truck loads of materials were recovered. In addition, several loads of large, bulky and heavy items were returned on a 40-ft. flatbed truck. A skeleton crew of personnel remained in Beatty through Friday March 19 performing followup surveys and generally wrapping up loose ends. On Monday, March 15, representatives from NRC, EPA and the waste disposal company met with the Governor of Nevada and other State officials to hold a debriefing. The Governor had maintained a keen interest and had requested frequent updates during the course of the survey effort. A joint State/NRC press conference was held following the meeting with the Governor. The press conference was attended by several Las Vegas radio and television stations as well as the printed press. News coverage waned quickly after the initial coverage.

Isolated survey efforts continued through March 26. Previously unavailable locations in and around Beatty were surveyed. It was necessary from information gathered during investigations in Beatty to followup leads and conduct surveys in Indian Springs, Pahrump and Fallon, Nevada as well as Richland Washington and a few locations in California. A few additional items were recovered as a result of these surveys.

During the course of surveying in Beatty and the vicinity, about two dozen Bennett Buckets were located. By best estimates it was considered that a little over one hundred unaccounted for Bennett Buckets were still in the public domain. Since many of these containers were used to ship significant quantities of the more hazardous radionuclides, NRC concluded that further effort was necessary to locate the containers. On March 30, the NRC Region V office issued 1500 postal patron addressed letters covering the geographical areas which could reasonably be affected. The letter briefly described the circumstances, provided a sketch of a Bennett Bucket and provided a convenient reply form with a self-addressed postage paid envelope. The letter assured residents that NRC's interest was to locate and survey the containers or other materials believed to have come from the disposal site.

Thirty responses were received to the letters (only responses with positive information were requested). Of the thirty, the majority reported negative information, a few responded with uncomplimentary remarks and a few reported containers or other materials which may have originated at the disposal site. Followup surveys were made on these items and, as a result, a few more items were recovered.

An attempt was made to quantify the cost of responding to this radiological incident. It was estimated that approximately $50,000 combined cost was incurred by the various organizations involved in investigating the incident, conducting surveys and generally evaluating the health and safety significance. The specific impact of this occurrence on the public health and safety is the subject of another paper entitled "Health and Safety Implication of a Widespread, Unauthorized Dispersal of Radioactive Waste in the Public Domain" (No79).

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HEALTH AND SAFETY IMPLICATION OF A WIDESPREAD, UNAUTHORIZED DISPERSAL OF RADIOACTIVE WASTE IN THE PUBLIC DOMAIN

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Abstract

As a result of the distribution of contaminated and potentially contaminated articles and containers in the public domain, assessment of the possible threat to public health and safety was required. An extensive effort was made to identify radioisotopes and assess the hazard associated with the large number of items recovered during surveys. Exposure by direct radiation appeared minimal, and the majority of effort was devoted toward identifying potential internal exposures. Many of the recovered articles were intact obsolete military equipment with luminous radium facings. Other typical items included a plutonium contaminated balance scale and weights, a strontium-90 contaminated stainless steel plate, a centrifuge with carbon-14 and cobalt-60 contamination and a hoist assembly with plutonium contamination. A large number of specially fabricated waste containers which had previously been used to ship significant quantities of americium, plutonium and other nuclides were found in use for grain storage, water supply tanks and other applications. Sensitive gamma radiation surveys and smear and water samples identified no significant contamination in those tanks which had been located. Based on available information of isotopic identification and quantification, and location and use of contaminated items individuals with the highest potential for internal deposition were identified for whole body counting and bioassay. The results of whole body counting, including phoswich counts for plutonium and bioassays for tritium, carbon-14, strontium 89-90 and plutonium-238 and 239 did not yield any evidence of internal deposition. Two employees of the waste disposal facility indicated higher than usual tritium urine levels which returned to normal on resampling. Based on consideration of all factors, this uncontrolled release of contaminated waste in the public domain had no identifiable effect on the exposed population.

Discussion

The disclosure of the distribution of contaminated or potentially contaminated articles or radioactive materials discussed in the "Response to a Widespread, Unauthorized Dispersal of Radioactive Waste in the Public Domain," (WE79), made necessary a prompt evaluation of the resulting threat to the health and safety of the public. The purpose of this paper is to identify the factors which resulted in the conclusion that this uncontrolled release of contaminated waste in the public domain had no identifiable effect on the population at risk.

The early phase of the investigation was conducted concurrently at three levels. The first involved field and laboratory measurements of radiation and contamination levels in Beatty and the surrounding area. Second, during

discussions with employees and former employees of the disposal company, efforts were made to identify items or types of items which had been diverted and, where possible, the recipients of such items. As information concerning the recipients or the types of items diverted became available, the information was provided to the field survey teams in order to permit the teams to be more specific and alert for certain types of items. The third level involved inquiries with certain organizations which had sent waste for disposal. The latter was done in an effort to identify the level of hazard represented by certain of the waste shipments that were routinely opened. Following the early phase, individuals were selected for whole body counting and bioassay based on the findings during the early phase.

The first field surveys were associated with slabs poured with concrete from the transit-mix truck used at the site to solidify liquid waste. Interviews identified three, and a possible fourth, locations in Beatty where slabs had been poured. The slabs were to be used as the foundation of a residence, a patio behind a saloon, the foundation and floor of the town jail and courthouse building, and possibly the floor of a chicken coop. Portable instruments used for these surveys included Baird Atomic NE-148A, Gamma Scintillators, with ranges to 3 mR per hour and a low range of 0 to 30 uR per hour. This instrument was capable of detecting a 5 uCi radium-226 source at six feet in air. The six foot measurement was approximately 2 uR per hour above background in a 4 uR per hour background field. The response of this instrument was sufficiently rapid to make it a good "fast search" gamma detector. Eberline Instrument Corporation (EIC) PAC-1SA, Alpha Scintillators, were used to monitor suspect locations and items that had no detectable gamma levels, and EIC E-500 B, Beta/Gamma Meters were used to survey gamma radiation levels above the range of the gamma scintillation instruments (HE76).

Surveys of the slabs identified three localized areas on the saloon slab which ranged from 8 to 45 mrad per hour, beta plus gamma, and a relatively uniform, 3 to 6 uR per hour, above background for the residence slab. These measurements were made with the detectors in contact with the slabs. Contamination in the residence slab appeared to be relatively homogeneous. The other two slabs were found to be background. Gamma spectrum analysis of concrete samples from the saloon identified the presence of localized concentrations of cobalt-60 with trace quantities of cobalt-58 and manganese-54. Samples of left over concrete from the residence slab pour showed trace levels of cobalt-58 and 60 and manganese-54. Concrete samples from the courthouse pour showed only background on analysis. Samples from the chicken coop pour were not analyzed. The saloon patio slab was removed and returned to the waste disposal site for burial. No action was taken with respect to the residence slab.

The continuing investigation identified an apparent widespread dispersal of potentially contaminated or radioactive items in the community and the surrounding area. In order to evaluate the threat to health and safety and to recover contaminated or radioactive items for safe disposal, a door-to-door survey including all rooms in each residence or business, all out-buildings, and all piles of salvaged or construction materials was conducted.

The monitoring teams were provided with instruments of the type previously described or with Ludlum Measurements, Inc., Model 125, Count Rate Meter (Micro-R-Meter), a gamma scintillation survey instrument with three

scale ranges to 3 mR per hour full scale. Also available were Reuter-Stokes, RSS-111, Environmental Radiation Monitors with ranges to 200 uR per hour; Ludlum Measurements, Inc., Model 2200 Scaler and Alpha Scintillator for swipe counting (HE76); EIC, E-520 Beta/Gamma Survey Meters;and PAC-4G, Gas Proportional Alpha Counters. A counting laboratory was set up in a local motel. Gamma spectroscopy and proportional counting for swipe analysis was provided at that location. Portable EIC, Model MS-3, Miniscalers with Ludlum Measurement, Inc., Gas Proportional Chambers were used for swipe evaluation by some field monitoring teams. Swipe samples collected during the surveys which showed contamination were in most cases analyzed promptly. Certain samples were retained for later more detailed analysis. The majority of samples were analyzed by Reynolds Electrical and Engineering Company, Inc. at the Nevada Test Site Laboratories (Re76). Additional analyses were performed by Lawrence Livermore Laboratory's (LLL) portable Ge(Li) system (An76); EPA, Environmental Monitoring Support Laboratory, Las Vegas (He76); and Health Safety Laboratory, Idaho.

Contaminated or radioactive articles identified by the monitoring teams were either collected during the surveys or, where large numbers of items were involved, were marked for later collection. Cans of spray paint were used for marking large items. It should be noted that background radiation in this area is variable and quite high, considerably above that found at Mercury, Nevada (An76). In addition, some of the residents are "rock hounds" and collect interesting and occasionally naturally radioactive rocks. Some of these materials had been incorporated into buildings (e.g., a rock fireplace) which did not simplify the monitors' work.

In response to NRC's request for assistance under the provisions of the Interagency Radiological Monitoring Assistance Plan (IRAP), ERDA made available three mobile detector systems. Two of the systems operated by EG&G were mounted in a helicopter and a van, respectively. The third system operated by Lawrence Livermore Laboratory (LLL) was also mounted in a van. This equipment was of particular value in the survey effort in that it was found that material from the waste disposal site had been transported substantial distances and distributed over wide areas. One of the ranches where many items were found covered 72,000 acres. Much of the area is broken and cut with dry washes. Roads other than highways are rudimentary if they exist.

The EG&G van and helicopter were used to evaluate ranches approximately 15 miles north of Beatty and along the roads and highways near Beatty. The van was also used in surveys along the streets and near piles of construction materials and debris in Beatty. The helicopter was also used for visual examination of remote or otherwise inaccessible areas.

The van was equipped with both neutron and gamma detector pods. The neutron pod contained three Harshaw Type B4-72S/50 neutron detector tubes. Each tube has a thermal neutron sensitivity of 1550 cps/n/cm^2 sec. The gamma pod contained twenty 12.7 cm diameter by 5 cm thick NaI (Tl) crystals. The gamma pod was oriented so that it was most sensitive to radiation from the side of the van. The minimum detectable point source gamma activity ranged from 10 uCi of cobalt-60 at 20 feet to 90 uCi of cesium-137 at 50 feet. The helicopter was equipped with gamma detectors similar in size and number to those contained in the van but in two separate pods. The minimum detectable point source gamma activity covered the same range as the van for direct fly overs. Lateral

displacement of the detectors equal to the altitude in the range of 20 to 50 feet increased the minimum detectable point source activities by factors of approximately 2.5 to 3 (Jo76). The LLL van was equipped with a portable Ge(Li) spectrometer and analyzer. The Ge(Li) system was used to analyze specific samples and was mounted on a one meter tripod for measurements in certain buildings or near large structures (An76).

The surveys performed by the monitoring teams, vans and helicopter established that there were no significant unshielded sources of gamma or neutron radiation in Beatty or on the surrounding ranches (Jo76). The maximum radiation level observed during these surveys was 250 mrad per hour (beta plus gamma) at contact with a box containing 40 radium luminized D.C. ammeters (Re76). Measurements at about six feet, while detectable with the instruments used, were insignificant when compared with the normal background of 10 to 15 uR per hour. This was true for all radiation levels observed.

Although direct radiation did not represent a hazard, the potential for internal exposure did appear to exist. The type of items identified and recovered during the surveys established the possibility for ingestion of radioactive materials due, in some cases, to direct and continuing contact with members of the public. While it was generally true that the contaminated items recovered did not appear to present a significant hazard, there were exceptions. During the surveys, an alpha contaminated, laboratory type, triple beam balance and weights were located in a private residence. The balance and weights were used as decorations which minimized the potential for exposure to the contaminant which was subsequently identified as plutonium-239. Swipe analysis identified as readily removable 3.8×10^{-3} uCi of plutonium-239 and 3.8×10^{-2} uCi of carbon-14 and lesser quantities of uranium-235, 238 and radium-226. Investigation identified the waste disposal facility as the original local source of the balance. At another residence, 74 miles from Beatty in Pahrump, Nevada, a stainless steel balance pan was found. The pan was in daily use by a former disposal site employee as a catch-all for keys, change and other small items. An open-window GM survey of the pan indicated 70 mrad per hour, beta plus gamma, on contact with the pan. Analysis of a swipe of the pan showed 1.3×10^{-1} uCi of strontium-90 and lesser quantities of cesium-134, 137, europium-154, 155 and cobalt-60. Two other items recovered at one ranch were of special interest. One was a hoist and support structure which was identified by NRC inspectors as having come from a source fabrication facility which had experienced a large spill of plutonium-238. The hoist had been used during the decontamination of that facility. Swipes of the hoist showed 2×10^{-3} uCi of plutonium-238 or americium-241; 10^{-3} uCi of cesium-137; and 4.5×10^{-7} uCi of plutonium-239. The other item was a disassembled metal room, the door still showing the radiation symbol and the words "Anti C's Required Beyond This Point" (Re76). The door and walls indicated up to 3 mrad per hour, beta plus gamma, and 10^{-5} uCi beta and 10^{-7} uCi alpha on swipes.

Approximately 400 swipe samples were analyzed by REECo. Only swipes which indicated contamination in the field were submitted for analysis. Nuclides identified included plutonium-238 and 239, uranium-235 and 238, neptunium-239, radium-226, strontium-89 and 90, cobalt-60, cerium-139 and 144, europium-154 and 155, ruthenium-rubidium-106, cesium-134 and 137, and antimony-125. The samples were collected at approximately 30 different locations; however, approximately 280 samples came from the five locations at which most of the contaminated items were found.

The previous paper, "Response to a Widespread, Unauthorized Dispersal of Radioactive Waste in the Public Domain," (We79), briefly discussed "Bennett Buckets" and mentioned their routine diversion from the site. Out of the 134 that were estimated to have been diverted only 22½ were found, half of them on one ranch. Not all of these containers were returned to the disposal site. If the containers in use were uncontaminated and the present owner wished, they were left in place. It is believed that many of the missing containers were used as septic tanks and were therefore effectively removed from contact with the public. Of particular concern, were two "Bennett Buckets" used as water tanks at residences, one as a potable water supply and the other for showers and watering pets. Samples of water from these two "Bennett Buckets" were analyzed by the EPA Las Vegas laboratory which reported no plutonium, tritium or gamma emitting nuclides in either sample (He76). Direct surveys and swipes of the two water tanks showed only background activity (Re76).

Information provided by the original shipper disclosed that some "Bennett Buckets" had contained significant quantities of hazardous materials. For example, three of the "Bennett Buckets" had contained, when originally shipped, 600 millicuries of californium-252 and americium-241, 30 curies of promethium-147, and 900 millicuries of plutonium-239, respectively. Original container serial numbers and content records were available; however, most of the "Bennett Buckets" recovered showed no identifying markings. It should be noted that none of the recovered "Bennett Buckets" exhibited any significant activity. However, the soil near the lip of one of the "Buckets" contained a 350 uR per hour "hot spot." Analysis of the soil "hot spot" identified cobalt-60, antimony-125, chromium-51 and silver-108m and 110m (He76).

During the investigation, it was learned that one of the major items salvaged from waste shipped to the disposal site was plywood from disassembled waste packing boxes. The salvaged plywood or portions of boxes had been used fairly widely for construction or were stockpiled at various homes and ranches. Some of the wood still retained labels or markings used to identify radioactive materials (e.g., "LSA") or the name of the consignee. Most of the wood was found to be at background levels; however, contact readings up to 3 mrad per hour were identified on some piled plywood. Monitors were alerted to the use of plywood in construction and surveyed plywood wherever it was observed. No significant use of contaminated plywood was found.

Many of the items recovered were military instruments, compasses and clocks with radium luminized markings. These items were originally disposed by the military services as a part of the military's radium removal programs. The highest radiation level associated with such items was 250 mrad per hour from the 40 D.C. ammeters previously mentioned. Ships' clocks or chronometers were probably the most widely distributed single item of salvage. These items, as well as any others that were found to be radioactive were returned for disposal. The monitoring teams' efforts also identified several old clocks which contained radium but which obviously had not come from the disposal site. In several cases, the owners requested that the clocks be disposed when informed that they were radioactive.

The NRC identified a total of eight representative individuals for whole body counting and bioassay sampling. The individuals were selected on the basis of the highest probability of internal exposure due to the possession of contaminated articles or the use of "Bennett Buckets" for potable and nonpotable

water supplies. Those selected included representatives of the families that possessed the contaminated balance, the stainless steel pan, hoist and contaminated disassembled "hot lab." Those selected also represented the locations that were the origin of most of the contaminated swipe samples.

The State of Nevada identified seven present and former waste disposal site employees for whom evaluation of internal deposition was desirable. The analyses were performed by the Environmental Monitoring and Support Laboratory (EM&SL), US EPA, Las Vegas, Nevada. The EM&SL had engaged in the whole body counting and bioassay evaluation of residents from communities and ranches surrounding the test site for a number of years, in support of test site activities. Selected families and individuals from these localities have been examined on a continuing basis. Coincidentally, three of the eight individuals identified by NRC for evaluation were former or current participants in the test site evaluation program.

Whole body counts were performed using two systems. One system consisted of a single 11 inch diameter by 4 inch thick sodium iodide crystal coupled to seven photomultiplier tubes. The detector output was supplied to 200 channels of a 400 channel TMC analyzer. This system had a sensitivity for cesium-137 of approximately 5 pCi per kg. This system could identify approximately 1 nCi of most gamma emitters.

EPA reported that for the 15 individuals counted cesium-137 burdens ranged from less than 5 to 32 pCi per kg. The normal range for cesium-137 in area residents is from less than 5 to 50 pCi per kg with an average of approximately 20 pCi per kg. No other gamma emitting nuclides were identified (Ka76-77).

The second system, for plutonium-239 screening, used a cesium iodide-sodium iodide phoswich (each segment is one inch by three inches in diameter) coupled to a single photomultiplier. The signal was supplied to 200 channels of the 400 channel TMC analyzer. The detector was held in the armpit and one count was made on each side of the chest. The phoswich was used for screening for bioassay since EM&SL had no method for evaluating and correcting for chest wall thickness and had not established the sensitivity of the system. The individuals counted were compared with counts of 100 EPA employees, none of whom had known plutonium exposure. None of the 15 individuals indicated a statistical probability of internal deposition (Ka76-77).

Urine samples were collected from the 15 individuals and analyzed for tritium. Thirteen of those sampled showed tritium levels of 350 to 6400 pCi per liter. The two others had levels of 24 and 29 nCi per liter. When resampled in several months, these two individuals showed tritium levels of 2100 and 2900 pCi per liter. It should be noted that the individuals with increased tritium levels were current employees at the disposal site and had been involved in the processing and disposal of large quantities of tritium contaminated water from a reactor. Strontium-89 and 90 analysis was performed on a urine sample obtained from the user of the stainless steel balance pan. Results were less than the minimum detectable activity. Composite analysis on the urine samples from all 15 individuals for plutonium-238 and 239 were negative (Ka76-77).

In summary, radioactive or contaminated items were recovered from approximately 45 locations in Beatty or from surrounding ranches or other

communities. The bulk of the recovered materials came from approximately four or five locations, each of which was associated with a present or former employee of the waste disposal site. The great majority of the recovered items consisted of radium luminized military surplus equipment. Direct radiation was excluded as a problem as a result of extensive surveys at many locations. Whole body counting and bioassay provided reasonable assurance that ingestion had not occurred in the population with the greatest exposure potential. Based on consideration of the factors discussed in this paper, it was concluded that there had been no significant effect on the population at risk.

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AN ASSESSMENT OF THE ENVIRONMENTAL TRANSPORT OF RADIOIODINE IN THE AIR-GRASS-COW-MILK PATHWAY USING REPORTED ENVIRONMENTAL MONITORING DATA

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ABSTRACT - The environmental transport of radioiodine has been a much studied and sometimes controversial subject. This paper presents an attempt to analyze the air-grass-cow-milk pathway by using data routinely reported to the U.S. Nuclear Regulatory Commission from the effluent and environmental monitoring programs maintained at nuclear power stations. This data is used in currently accepted transport models to make a comparison of computer predicted versus actual measured values of the resultant concentration of I-131 in milk samples.

Introduction

With the advent of nuclear power and its use to generate electrical energy, there has been much concern about the effects of radioactivity on the environment and man. Thus, the U.S. Nuclear Regulatory Commission (NRC) has, in Title 10 of the Code of Federal Regulations, placed constraints on the amount of radioactivity that can be routinely released by a nuclear-powered generating station. Perhaps the most limiting pathway for radio-nuclides through the environment to man, is the air-grass-cow-milk path taken by radioiodine. Iodine in man is concentrated in the thyroid gland, and thus exiguous amounts of radioiodine can give a disproportionately large dosage of radiation to this small organ.

This report is an attempt to utilize routine environmental monitoring data to assess the reasonableness of the U.S. Nuclear Regulatory Commission's models for the environmental transport of radioiodine in the milk-food chain by comparing predicted versus actual measured levels of radioiodine in milk.

The method chosen to evaluate the NRC's predictive models was to identify particular cases where radioactivity had been detected during the operation of routine environmental monitoring programs conducted by the utilities which operate the nuclear power stations. Once such cases had been found, the applicable NRC models could be used to determine if there was a relationship between the predicted levels of radioactivity and the actual measured levels. Because of atmospheric testing of nuclear weapons, many instances where radioactivity was detected had to be eliminated because the activity could not be directly attributed to the operation of the nuclear plant.

Iodine-131 in milk was chosen as the measurement to consider in this report because it offers several advantages over other environmental measurements. First, the I-131 can be readily measured at the end of the pathway, just before man; furthermore, the analytical procedures for the detection and measurement of I-131 in milk are sensitive and usually yield good results. Another advantage is the relatively long half-life of iodine-131 (8 days) as compared to the time required to travel through the pathway. Iodine-133 and iodine-135 are also released by nuclear plants, but their half-lives of 21 hours and 6.7 hours respectively, are not long enough to allow measureable amounts of these isotopes to reach the milk stage of the pathway. In addition, the iodine-131 pathway is recognized as a primary mode by which radioactivity released to the environment can reach and affect man. The particular pathway investigated was that of gaseous iodine-131 in the reactor-air-grass-cow-milk chain.

The Meteorological and Cow-Milk Models

The modeling of the environmental transport of radioiodine is accomplished in two separate parts. The first part consists of using the meteorological data (i.e., wind speed, wind direction, atmospheric stability class, etc.) during the period of concern to predict atmospheric dispersion and relative deposition. That is, the model predicts the amount of radioiodine that will be deposited on the ground at some point of interest due to the release of iodine by a nuclear plant. The second part is the cow-milk portion of the pathway which uses the relative deposition value predicted by the meteorological model to determine the concentration of radioiodine in the milk of a cow grazing on the contaminated pasture. A detailed discussion of the meteorological and cow-milk pathway models follows.

The meteorological model used by the U.S. Nuclear Regulatory Commission in its evaluation of the atmospheric transport and dispersion of radioactive material is the XOQDOQ model and is entitled "Program for the Meteorological Evaluation of Routine Effluent Releases at Nuclear Power Stations" (Ref. 5). This model is of the constant-mean-wind-direction type, and uses "straight-line" airflow to calculate average relative air concentrations (X/Q's) and average relative depositions (D/Q's) for effluent material. The basic equation given in the XOQDOQ model for calculating the relative air concentration for elevated releases is:

$$\frac{\overline{X}(x,k)}{Q} = \frac{2.032 \cdot RF_k(x)}{x} \sum_{ij} \frac{DEPL_{ijk}(x) \cdot DEC_i(x) \cdot f_{ijk} \exp\left[-\frac{1}{2}\frac{h_e^2}{2_{zj}(x)}\right]}{\overline{u}_i \, \sigma_{zj}(x)} \quad \text{(Eq. 1)}$$

where

$\frac{\overline{X}(x,k)}{Q}$ = average effluent concentration normalized by source strength at distance x and direction k;

$\overline{u}_i$ = mid-point values of the ith wind speed class;

$\sigma_{zj}(x)$ = vertical (z) spread of effluent at distance x for the jth stability class;

f_{ijk} = joint probability of the ith wind speed class, jth stability class, and kth wind direction;

x = downwind distance from release point or building;

h_e = effective plume height;

$DEC_i(x)$ = reduction factor due to radioactive decay at distance x for the ith wind speed class;

$DEPL_{ijk}(x)$ = reduction factor due to plume depletion at distance x for the ith wind speed class, jth stability class, and kth wind direction; and

$RF_k(x)$ = correction factor for air recirculation and stagnation at distance x and kth wind direction.

This equation requires that a three-way, joint distribution be calculated for the wind speed, wind direction, and atmospheric stability class.

The wind speed can be distributed into a maximum of fourteen velocity categories. Various strategies can be employed for determining the breakdown of the categories. For example, the wind speed divisions could be equally spaced (e.g., 2 mph, 4 mph, 6 mph, etc.), or each division could contain an equal number of values. Also a finer breakdown of the lower wind speed categories could be used since the effluent does not travel as far in one increment of time for the lower velocity winds.

Wind direction is distributed into the sixteen cardinal compass sectors of 22.5 degrees each. These divisions of wind direction are shown in Figure 1.

The meteorology is also distributed by atmospheric stability. In this report the stability of the atmosphere was classified according to the temperature lapse rate (i.e., the change in the ambient temperature of the air with a change in altitude); specifically, Pasquill stability categories were used (refer to Table 1).

The meteorological data, once distributed into the proper form, is used by the XOQDOQ computer code, along with other necessary information, to calculate the atmospheric dispersion and the subsequent deposition of the effluent.

TABLE 1

PASQUILL STABILITY CLASSES

Lapse Rate (^{o}c/100m)	Class	Stability
< - 1.9	A	Very unstable
-1.9 to -1.7	B	Unstable
-1.7 to -1.5	C	Slightly unstable
-1.5 to -0.5	D	Neutral
-0.5 to +1.5	E	Slightly stable
+1.5 to +4.0	F	Stable
> 4.0	G	Very stable

Other parameters taken into consideration by the XOQDOQ model are: terrain (allowance being made for effluent recirculation), height and velocity at which effluent was released, height at which wind data was measured (for possible extrapolation to release height), plume decay and depletion; as well as allowing for possible building wake correction.

There are several inherent limitations in the XOQDOQ computer code, as well as some imposed by the methodology of this study. The model can use meteorological data for a single station only, and thus cannot take into consideration spatial variation in wind speed, direction, etc. Wind directions near the border of two sectors are credited to only one and is added into the total average of that sector; thus, any contribution of the effluent to the neighboring sector is neglected. This effect could be enhanced where local topography may bias the wind direction; for example, the wind may preferentially blow in the direction of 75 degrees, and therefore be credited to the ENE sector, and not to the E sector lying 3.75 degrees away. Also, the meteorological data

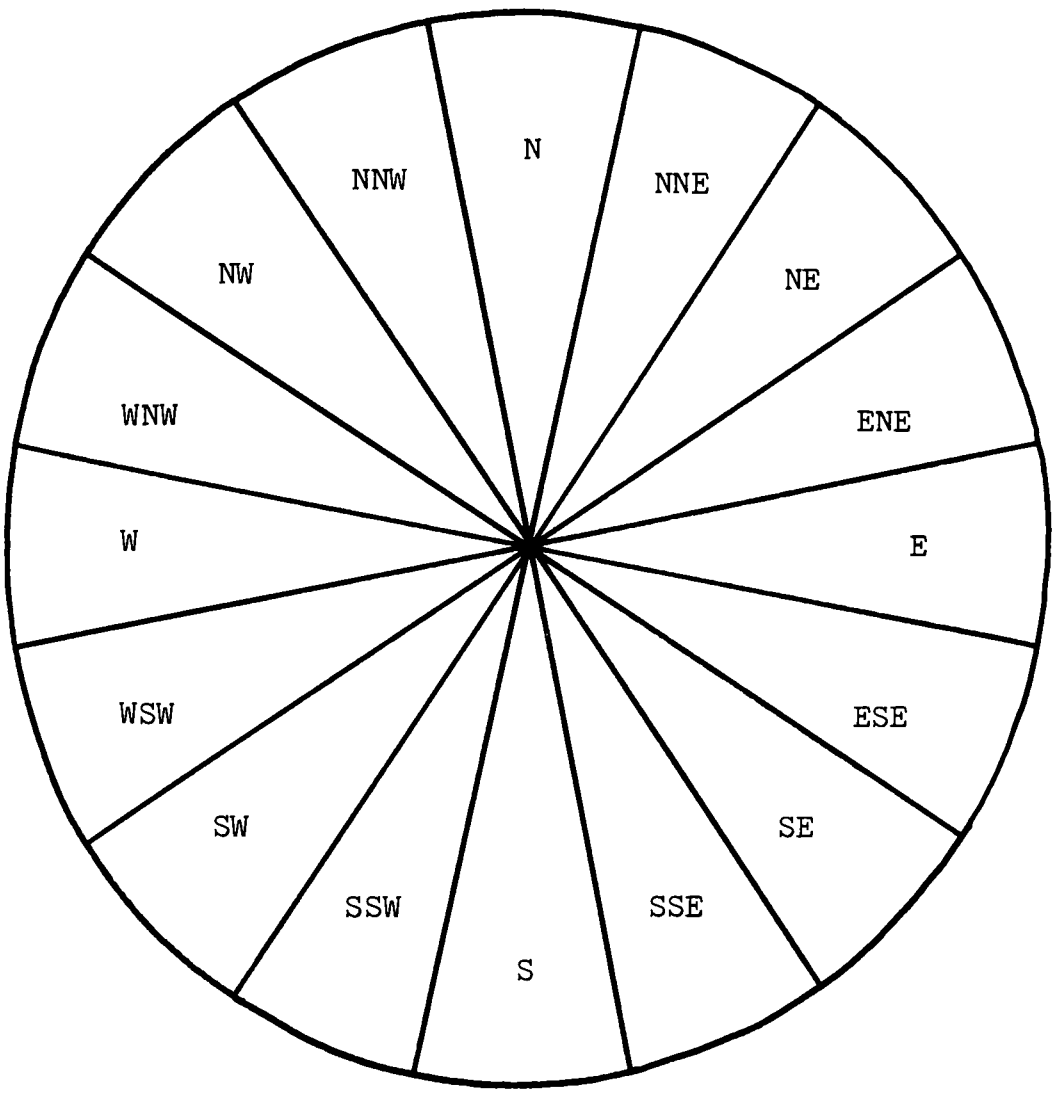

FIGURE 1.

WIND DIRECTION DISTRIBUTION

input to this model usually consists of hourly values that are calculated from fifteen-minute averages, as opposed to the average being taken over the whole hour.

As previously mentioned, the methodology of this study necessarily imposes some additional limitations to accuracy of the XOQDOQ model. With the purpose of the study being to compare model predictions with actual measured cases of I-131 appearing in milk samples it was necessary to use only a short time period of data to describe the meteorological conditions concomitant with the milk sample. The XOQDOQ model, however, is structured primarily to calculate annual averages of relative deposition and air concentration values. Thus instead of the usual 8760 hourly units of data for a year, only approximately a week's worth of data, which generally contains 168 hourly units, is used. This tends to amplify the effect of any wind direction biasing that would probably cause much less of an effect for a full year of data. In addition, it should be emphasized that the effects of the wind blowing into a particular sector were averaged over the whole sector.

Although the XOQDOQ computer code was not specifically designed to compute effluent air concentration and deposition values for short time periods, it can still be expected to yield reasonable results for this study.

After the radioiodine had been deposited on the pasture, it was assumed that the animal would consume the contaminated grass, metabolize the iodine-131 and secrete it into the milk. The mathematical representation for this model was taken from Section C.3.b of the NRC Regulatory Guide 1.109, "Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I" (Ref. 4).

To calculate the amount of I-131 appearing in the animal's milk from a given amount of I-131 deposited on the pasture grass, the following relationship was used:

$$C_m(t) = \frac{(D/Q)(Q)(f_s)(Q_f)(F_m)}{Y_u} \exp(-\lambda t) \qquad \text{(Eq. 2)}$$

where

$C_m(t)$ = Concentration in milk of I-131 (pCi/ℓ), at time t;

D/Q = Normalized deposition on pasture grass for I-131 (m^{-2});

Q = Amount of I-131 released by the nuclear plant (pCi);

f_s = Fraction of animal's daily intake of food which consists of pasture grass (default = 1);

Q_f = Animal's daily food intake (50 kg/day for cows, 6 kg/day for goats;

F_m = The average fraction of the animals daily intake of I-131 which appears in each liter of milk (0.006 days/liter for cows, 0.06 days/liter for goats);

Y_u = Crop yield -- the amount of pasture grass per unit area (0.75 kg/m^2);

λ = Radioactive decay constant for I-131 (0.086 day^{-1});

t = Average transfer time between the intake of I-131 and its appearance in the milk (approximately six hours, normally taken as zero), (Ref. 6).

The normalized deposition (D/Q) was calculated using the XOQDOQ model for the location where the animal grazes. This normalized deposition was then multiplied by the amount of I-131 released (Q), giving the amount of iodine per unit area of pasture grass. The significance of this calculation was that the total iodine released from the nuclear plant, for the week prior to the occurrence of the I-131 contaminated milk sample, was assumed to be deposited on the pasture grass. No radioactive or weathering decay corrections were considered in the deposition calculation. In the absence of site-specific data, the values of f_s, Q_f, and Y_u were assumed to have the values indicated above. The values for the transfer factor (F_m) were taken from NRC Regulatory Guide 1.109.

As with the meteorological model, there are several assumptions that must be made which limit the accuracy of the cow-milk model. The principal uncertainty of this model is the amount of I-131 consumed by the cow. This iodine value is based on estimates of food intake by the animal (Q_f = 50 kg/day for cows; 6 kg/day for goats), and the areal grass density (0.75 kg/m^2); both of these estimates are subject to wide variations.

How The Study Was Carried Out

As stated earlier, the purpose of this study was to attempt to utilize the data gathered by the environmental monitoring programs at nuclear power stations to evaluate the reasonableness of the NRC's predictive models for the environmental transport of radioiodine. To effect this study, it was necessary to identify cases where measurable quantities of iodine-131 appeared in milk samples, and then obtain the pertinent meteorological and source term data.

As part of the environmental monitoring program required of the licensees by the U.S. Nuclear Regulatory Commission, periodic collection and analysis of milk samples from nearby dairy farms is performed. Milk sampling is usually done on a weekly basis during the pasturing season. (In the northern states the pasturing season covers the period of May through September, whereas in the southern states it may extend throughout the whole year.) Although the major part of the analysis of the milk is for the detection and measurement of iodine-131, some licensees analyze for radiostrontiums and gamma-emitting nuclides. Because gamma spectroscopy techniques are not sensitive enough to achieve the lower limits of detection required by the NRC for iodine-131 activity, radiochemistry procedures are utilized to concentrate the iodine-131 for beta or beta-gamma coincidence counting. The results of the milk sample testing are reported to the NRC in the licensees' periodic environmental reports.

Because boiling water reactors (BWR's) release effluents continuously, a review of the environmental reports of all BWR's was performed. The criteria

for identifying a specific case of interest was that the measured iodine-131 value in milk (pCi/ℓ) was larger than the associated error at the 95% confidence level (2); however, in certain instances exceptions to this criterion were made. After obtaining a list of cases, a determination was made as to whether or not the source of the iodine-131 was fallout from the atmospheric testing of nuclear weapons. Several "clear periods" were established when the appearance of iodine-131 in the milk samples was attributable to the operation of the nuclear plants, and not to the nuclear weapons testing. The summer months of 1975, and the whole of 1976, were determined to be two such "clear periods". Following these allowances, approximately thirty cases were identified for further analysis.

Using The Data In The Models

Because of the large uncertainties inherent in the XOQDOQ computer code, it is generally used to compute annual averages. Therefore, it should be recognized that the use of the XOQDOQ model in this study, with the limited data available, has significant deficiencies. However, the model provides a basis for "normalizing" the evaluations of the cases, that is, all the cases (with the exception of case #17 where a puff model was used) were analyzed by the same procedure.

With the milk sampling having been done on a weekly schedule, and since most of the cases were singular, that is, there was usually no iodine-131 detected in the previous week's milk samples, the meteorological data for the week prior to the iodine-131 containing milk sample was used to calculate the iodine-131 concentrations. Furthermore, the iodine-131 source term measurements were also generally made at weekly intervals. Thus, the normalized deposition values predicted by the XOQDOQ model were based on a week of hourly meteorological data. The total iodine-131 released during the week was deposited at the rate predicted by the XOQDOQ model at the point of interest. This deposition was then used in the cow-milk pathway model to predict the amount of iodine-131 content in the milk. The predicted iodine-131 value was then compared with the actual measured value to determine the amount of agreement between the two values.

Summary and Conclusions

The results of the analyses are sumarized in Table 2 and from these data there is no simple and easily distinguishable correlation between the predicted milk values and the actual measured values. However, even though the analyses of the twenty-eight cases cannot either substantiate or invalidate the meteorological and/or cow-milk models used in the study, certain interesting observations can be made which may provide possible answers to anomalies previously discussed in the iodine pathway literature, as well as some insights into the processes involved.

If cases 10 through 16 are considered separately from the others, the models values are in reasonable agreement with the measured levels. These predicted-to-measured ratios are in the range of 0.11 to 17 (excluding case #12). During the time periods associated with these cases, the reactor was operating normally (56% to 94% full power) with no elevated release rates of iodine-131. Although these cases were unique because the milk samples were obtained from goats, and not cows as in all the other cases, there is no evidence to indicate that such

TABLE 2

RESULTS OF THE ANALYSES

CASE #	MILK VALUES (pCi/l)		PREDICTED TO MEASURED RATIO	PRECIPITATION*
	PREDICTED	MEASURED		
1	0.038	1.89±0.30	0.02	1.75+
2	26.7	5.26±0.84	5.1	0.59+
3	3.7	1.47±0.24	2.5	1.50
4	0	0.78±0.38	0	1.09+
5	1.28	1.08±0.19	1.2	1.09+
6	0.15	2.61±0.29	0.06	1.09+
7	58	1.70±0.14	34	0.91
8	39	0.04±0.11	970	0.91
9	430	2.99±0.20	143	0.91
10	0.26	2.3	0.11	0.79+
11	0.79	0.1	7.9	0.79+
12	0	0.53	0	0
13	0.61	1.40	0.44	0.38
14	0.24	0.18	1.3	0.38
15	18.2	3.4	5.4	0.26+
16	4.8	0.29	17	0.26+
17	1.7	10±1	0.17	0.15+
18	85	4.7±0.2	18	0.52+
19	0	0.58±0.19	0	0.52
20	120	0.97±0.12	124	0.52+
21	20	2.8±0.2	7.1	0.42
22	36	1.2±0.2	30	0.42+
23	246	0.70±0.17	350	0.42+
24	20	0.68±0.16	29	0.42
25	47	0.81±0.16	58	0.38+
26	120	4.2±0.2	29	0.38
27	69	0.64±0.16	108	0.38+
28	16	1.4±0.2	11	0.38+

* TOTAL INCHES FOR THE WEEK PRIOR TO THE SAMPLE

"+" INDICATES WASHOUT WAS POSSIBLY A FACTOR IN DEPOSITION

a difference is significant once the food intake and lactation parameters are adjusted appropriately.

For cases 18 through 28 the predicted values were very high, having an average predicted-to-measured ratio of seventy. The major portion of the food intake of the cows at the local dairy farms was from stored feed and not pasture grass since the cows were pastured for exercise only and on an irregular basis. This would provide part of the reason for the high ratios, but was probably not the entire cause. (Note: the food intake parameters, f_s and Q_f, of Equation 2 were not adjusted from their normal values of one and fifty respectively.

Also, it can be noted from Table 2 that high values for the predicted-to-measured ratios were usually associated with high release rates. In thirteen cases (7, 8, 9, 18, and 20 through 28) high ratio values were concomitant with high release rates of iodine-131, although the cause-effect relationship was not obvious.

As discussed earlier, the effects of rain on deposition are not considered in the meteorological model used in this study. However, whereas precipitation may occur for a very small fraction of a period of interest, precipitation scavenging appeared to be one of the more critical factors in determining whether or not radioiodine was detected in milk. In only one of the twenty-eight cases analyzed there was no rainfall associated with the milk sample. In seventeen cases washout by rain seemed to be a major cause. (These cases are indicated by a "+" on Table 2). This determination was made by considering the wind's direction during the rainfall. If the wind was blowing in the general direction of the farm (within 100°), while the rain was falling, then washout was considered to be a probable major factor in deposition. The effect of washout could be to either increase or decrease the amount of deposition at the dairy farm. If the washout occurred before the effluent reached the farm, the amount of iodine-131 consumed by the cows (or goats) would be significantly less than if the washout occurred directly over the farm.

In cases one and seventeen the washout effect was particularly critical. For case #1, 0.38 inches of rain fell while the wind was blowing toward the dairy farm involved. Another 1.07 inches of rain occurred while the wind was blowing in the approximate direction of the farm (circa 30° relative to the location of the farm). Washout also was a major factor in case No. 17. The scenario for this case was 0.15 inches of rain concomitant with a wind in the general direction of the farm (circa 50° relative to the farm). The amount of iodine-131, 10 ± 1 pCi/ℓ, measured in the milk sample would have undoubtedly been significantly less if not for the effect of precipitation scavenging.

The analyses of the twenty-eight cases provided a basis on which several general observations were made: 1) no good correlation was found between the predicted and measured values; 2) short term, high iodine-131 effluent release rates associated with reactor refueling did not significantly affect the radioiodine concentration in milk at local dairies unless there were mitigating circumstances such as rain; 3) precipitation scavenging appeared to have played a major role in the deposition of the iodine-131 in the air-grass portion of the pathway.

These observations indicate that precipitation scavenging is a primary mechanism in the environmental transport of radioiodine and should be considered

in any modeling of the air-grass-cow-milk pathway. However, it should be noted that precipitation scavenging is a function of many factors, such as: the amount of rain, the size of the droplets, cloud height, temperature, etc. These and other factors are discussed fully in Meteorology and Atomic Energy (Ref. 2).

The results of this study have strongly supported the feasibility of using data from effluent release and environmental monitoring programs for analyzing environmental transport mechanisms for radionuclides, and further studies should be accomplished using this available data.

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