U.S. DEPARTMENT OF COMMERCE
National Technical Information Service

PB-281 041

Natural Radioactivity Contamination Problems

Conference of Radiation Control Program Directors, Inc

Prepared for

Office of Radiation Programs, Washington, D C

Feb 78

NATURAL RADIOACTIVITY CONTAMINATION PROBLEMS

Published by
U.S. Environmental Protection Agency
Office of Radiation Programs
Washington, D.C. 20460

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520/1-77-009	Radiological Quality of the Environment In The United States 1977

BIBLIOGRAPHIC DATA SHEET	1. Report No. EPA 520/4-77-15	2.	3. Recipient's Accession No.
4. Title and Subtitle NATURAL RADIOACTIVITY CONTAMINATION PROBLEMS			5. Report Date February 1978 6.
7. Author(s)			8. Performing Organization Rept. No.
9. Performing Organization Name and Address A REPORT OF THE TASK FORCE Prepared by Conference of Radiation Control Program Directors, Inc.			10. Project/Task/Work Unit No. 11. Contract/Grant No. PHS 223-76-6018 EPA D7-0968
12. Sponsoring Organization Name and Address U.S Environmental Protection Agency Office of Radiation Programs (AW-458) Washington, D.C. 20460			13. Type of Report & Period Covered 14.

15. Supplementary Notes

16. Abstracts

Naturally-occurring radionuclides are ubiquitous in the environment. Under various circumstances these radionuclides primarily from the uranium and thorium decay series can contaminate the environment to the extent that they pose real or potential public health risks. The investigation and regulatory control of the impacts of most of these sources have been greatly overlooked by Fedederal and State agencies in the past.

This report provides an initial assessment of the scope of the contamination problems, the priorities for radiation control efforts, and recommendations for problem resolution and implementation of effective control measures. This report is intended to assist those persons or agencies interested in the protection of public health from naturally-occurring radionuclide contamination.

17. Key Words and Document Analysis. 17a. Descriptors

17b. Identifiers/Open-Ended Terms

Natural radioactivity	Mineral processing	Radon
Radiological problems	Consumer products	Lead
Radiation contamination	Construction materials	Polonium
Fossil fuels	Phosphate industry	Zirconium
Ground water	Uranium	
Mineral extraction	Radium	

17c. COSATI Field/Group

18. Availability Statement	19. Security Class (This Report) UNCLASSIFIED	21. No. of Pages 110
	20. Security Class (This Page) UNCLASSIFIED	22. Price PC A06/MF A01

EPA-520/4-77-015

NATURAL RADIOACTIVITY CONTAMINATION PROBLEMS

A REPORT OF THE TASK FORCE

Prepared pursuant to
PHS Contract Number 223-76-6018
which is partially funded through
EPA Interagency Agreement D7-0968

Printed February 1978

Prepared by

Conference of Radiation Control Program Directors, Inc.

With the cooperation of

U.S. Nuclear Regulatory Commission

U.S. Department of Health , Education and Welfare
Bureau of Radiological Health

and

U.S. Environmental Protection Agency
Office of Radiation Programs
Washington, D.C. 20460

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FOREWORD

The Conference of Radiation Control Program Directors is an organization whose membership is comprised of all directors of radiation control programs in the 50 States, the Territories, and some large municipal agencies. The Conference was formed to serve as a mechanism for providing a more functional means of exchanging information between State and Federal agencies as well as between States themselves in areas of mutual concern or interest. Additional objectives and purposes of this Conference are to:

(1) Promote radiological health in all aspects and phases.

(2) Encourage and promote cooperative enforcement programs with Federal agencies and between related enforcement agencies within each State.

(3) Collect and make accessible to all radiation control program directors such information and data as might be of assistance to them in the proper fulfillment of their duties.

(4) Foster uniformity of radiation control laws and regulations.

(5) Support programs which will contribute to radiation control.

(6) Assist members in their technical work and development.

(7) Exercise leadership with radiation control professionals and consumers in radiation control development and action.

The Office of Radiation Programs of the Environmental Protection Agency carries out a National program designed to evaluate public health impact from ionizing and nonionizing radiation, and to promote development of control necessary to protect the public health and ensure environmental quality. In this regard, the Environmental Protection Agency (1) develops

radiation protection standards, criteria, and guidance, (2) conduct special environmental studies, (3) evaluates radiation exposure trends, (4) assesses radiation control technology, and (5) provides technical assistance to State and local agencies responsible for radiation control.

The Office of Radiation Programs, through funding and direct technical assistance, supports the Conference of Radiation Control Program Directors, Inc., in its objectives and activities to assure an effective Federal/State partnership in limiting unnecessary environmental and public radiation exposure. Selected Conference reports are published by the Environmental Protection Agency and are distributed to Federal, State and local radiation protection personnel, industry, libraries, laboratories, and other concerned groups and individuals. These publications are for sale by the Government Printing Office and/or the National Technical Information Service.

Readers are encouraged to report errors or omissions to the Conference or the Office of Radiation Programs.

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PREFACE

Naturally-occurring radionuclides are ubiquitous in the environment. Under various circumstances these radionuclides primarily from the uranium and thorium decay series can contaminate the environment to the extent that they pose real or potential public health risks. The investigation and regulatory control of the impacts of most of these sources have been greatly overlooked by Federal and State agencies in the past. In order to initiate effective control measures in this radiation protection problem area, the Conference of Radiation Control Program Directors, Inc., established a Task Force to assess contamination by naturally-occurring radionuclides and assist the States and Federal agencies in devloping appropriate radiation protection guidance and criteria. The Task Force consisted of representatives from several State radiation control programs with resource persons from the Environmental Protection Agency.

This report provides an initial assessment of the scope of the contamination problems, the priorities for radiation control efforts, and the Task Force's recommendations for problem resolution and implementation of effective control measures. This report is intented to assist those persons or agencies interested in the protection of public health from naturally-occurring radionuclide contamination.

This document should be of special interest to State, local, and Federal radiation protection personnel in the United States and other countries.

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HISTORY AND PURPOSE OF THE TASK FORCE

Task Force No. 7 on Natural Radioactivity Contamination Problems was established by the Executive Committed of the National Conference of Radiation Control Program Directors in 1975 as an extension of Workshop No. 5 of the 1974 Annual Meeting of the NCRCPD.

The charge to this task Force is to:

(1) Provide assistance to the Conference, individual States, and Federal agencies in scoping the problem of contamination by natural radioactivity;

(2) Assist in developing appropriate radiation protection guidance and criteria;

(3) Assess the impact of naturally-occurring radioactivity contamination in the general environment and the Conference member States; and

(4) Serve as a focal point for State input to the programs of Federal agencies.

The charge given necessarily implies certain responsibilities in which the Task Force must assist the appropriate State and Federal agencies. These include the following:

(1) Defining the radiation level or concentration or stage of processing in which naturally-occurring radioactive material (NORM) becomes a contamination to the environment;

(2) Identifying who has authority to develop and implement guidelines and criteria for enforcement action;

(3) Identifying the impact that naturally-occurring radioactive contaimination has on the general population;

(4) Examining the need for control on the use of products and byproducts containing NORM and the associated economic impact of such control;

(5) Defining the sources currently known or suspected to contain possibly hazardous amounts of NORM and describing other potential problem areas; and

(6) Recommending priorities for control programs to address NORM problems.

INTRODUCTION

Natural radioactivity and its associated radiological impacts have generally been overlooked in the past. Although there have been strict controls on other sources of radiation such as byproduct material, X-ray machines, special nuclear material, etc., natural radioactivity control has been minimal, perhaps due to the fact that it is "natural" rather than "man made" radioactivity. This lack of strict controls has been due in part to the fact that the Federal Government has limited jurisdiction over naturally-occurring radioactive material, and control was left previously up to the States, who often times did not have adequate programs to deal with radiation. While there are over 100 naturally-occurring radionuclides, public health problems are usually limited to the 30 or more radionuclides in the uranium and thorium decay series because of their relative abundance and toxicity. The increased incidence of bone cancer in radium dial painters and lung cancer in fluorospar and uranium miners are examples of undesirable health impacts due to exposure to these radionuclides. Other examples of increased population exposure to NORM include the radon problems in several Western States due to the use of radioactive tailings and the use of reclaimed phosphate mining lands in Florida.

The majority of the work of this Task Force does not deal with what is usually referred to as truly natural radiation exposure, but more appropriately is concerned with exposure to radiation occurring as a result of alteration of the natural sources by technology. This new category for human radiation exposure, introduced by Gesell and Pritchard,[1] is termed "technologically enhanced natural radiation" (TENR), and is

defined as "exposures to truly natural sources of radiation (i.e., naturally-occurring isotopes and cosmic radiation) which would not occur without (or would be increased by) some technological activity not expressly designed to produce radiation." This exposure to increased radiation from living in a structure constructed over phosphate mining reclaimed land would constitute a TENR exposure; however, an exposure from a radium needle would not, since the latter is expressly designed to produce radiation. It is interesting to note the EPA in their May, 1976 report, "Radiological Quality of the Environment", has estimated from the sketchy data available, that for individuals, the largest radiation dose received from all sources is derived from TENR. This results in 140-14,000 millirems per year to the tracheobronchial surface tissue of the lung, mainly as a result of inhalation of radon daughter products from uranium mill tailings. Additionally, the third largest category of population dose is estimated to be from TENR which contributes approximately 2 million pers-Rem per year internal exposure. This internal dose results from exposure to ore mining and milling, radon in potable water, natural gas, LPG, mines, caves, and in geothermal energy production, and radioactivity in construction materials. This is comparable to the dose received from the use of radiopharmaceuticals, which is the second largest category of population dose contributing slightly greater than 3 million person-Rem per year.[2]

It is the objective of this Task Force to collect and study available information and data on naturally-occurring radioactive materials in the environment and to make recommendations to the Conference regarding the need for evaluating, monitoring, and controlling natural radioactivity.

The problem areas examined by the Task Force are as follows:

A. The phosphate industry

B. The radiological aspects of throium and daughter products

C. Radioactivity in fossil fuels

D. Radium and radon in ground water

E. Mineral extraction and processing activities

F. Radon in caves

G. Standards and guidelines for radioactive material concentrations in material and consumer products

H. Radioactivity in construction materials

I. Statutory authority of States to regulate naturally-occurring radioactive materials.

The Task Force has developed a series of recommendations based on findings and conclusion of data, and model State regulations have been developed for potential incorporation into the CSG model State regulations and adoption by those States where such action may be warranted.

In November 1975, the first formal meeting of the Task Force was held in Orlando, Florida. During this time, the members toured the base phosphate mining area of Central Florida and visited several housing projects built on reclaimed land in order to obtain a perspective on the magnitude of the problem. Radiation surveys were taken using a Micro-R meter over reclaimed land and housing projects, near slag piles, and over roads constructed with phosphate byproduct material. A report of this meeting was submitted to the Conference in January 1976 and can be made available to interested parties. Later in September, 1976, a follow-up meeting was held in Baton Rouge, Louisiana, at which time Task Force members toured a wet process phosphoric acid production plant,

prepared a working outline for preparation of the final report, assigned specific areas to be completed by the members, and established preliminary recommendations.

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1. Gesell, T. F. and Prichard, N. M., The Technologically Enhanced Natural Radiation Environment. Health Physics, V. 28, No. 4, 197

2. Radiological Quality of the Environment, U. S. EPA, ORP, EPA-52/1-76-010, 1976.

PHOSPHATE INDUSTRY

The radioactivity of phosphate rock was probably first observed in 1908 when the British physicist R. Strutt (1908) found that samples of phosphorite were several times more radioactive than the average rocks of the earth's crust. More recent studies of the concentrations of natural uranium and thorium in phosphate ores produced in the United States indicate that concentrations of these natural materials range from about 10 to 400 ppm (5.4-267 pCi/gm) and 2 to 20 ppm (0.4-4 pCi/gm), respectively.[1,2] Uranium daughters in the phosphate ores, at least through radium-226, are usually in secular equilibrium.

Industrial Operations

In 1974, the total U. S. production of marketable phosphate rock was about 46 million tons.[3] At present, the domestic marketable phosphate rock production accounts for about 40 percent of the total world production. The Florida phosphate industry produces about 80 percent of the total domestic phosphate rock output. The remaining output originates from several Western States. Consequently, the large scale operations of this industry in one regional area may lead to several types of impact on the environment including that from the radioactive impurities in the ores, wastes, and other materials.

The standard mining practice in Florida is to strip the overburden and mine the phosphate matrix. This overburden is stacked on unmined ground adjacent to the mining area. Approximately 5000 acres of land

are mined per year in Florida, removing about 163 million cubic yards of overburden, and extracting 112 million yards of matrix. In the Western U.S., dry mining technics are used extracting the ore with power shovels and shipping it to manufacturing facilities in trucks and rail cars.[7]

At the beneficiation plant, the matrix is processed to upgrade its P_2O_5 concentration. The output materials from this operation are marketable phosphate rock, sand tailings and slimes. These materials are produced in a ratio of about one to one to one. Table 1 lists the uranium, thorium, and radium-226 activities for these materials.

TABLE 1: Natural Radioactivity Concentrations in Florida Phosphate Mine Products and Wastes (pCi/gm)[4]

Material	Ra-226	U-238	Th-230	Th-232
Marketable Rock	42	44	42.3	0.44
Slimes	45	44	48	1.4
Sand Tailings	7.5	5.3	4.2	0.89

In beneficiation, water is used for processing in addition to being used as a transportation medium. Mined-out areas are used for the disposal of sand tailings and slimes, in addition to overburden. Several Florida slime ponds have discharges to the environment, and beneficiation wastes are present in the slimes. The concentration of dissolved radium-226 in slime discharges was less than 5 pCi/liter at all facilities. The undissolved radium-226 concentration ranged from 10 to 7000 pCi/liter and was highly dependent on the total suspended solids in the slimes. Although no chemical process is used to treat the discharge from the slime ponds, concentrations of radium-226 in effluents were all less

than 3 pCi/liter. The reduction of total radium-226 from the raw slime discharge to the effluent discharge ranged from 92 percent to greater than 99.9 percent. This was primarily due to the removal of suspended solids by settling. Consequently, effective solids removal technics are necessary to insure minimal release of radium-226 to receiving waters.

Marketable phosphate rock is processed into two major products, fertilizers and elemental phosphorus. Processing for these products takes place in "wet process" phosphoric acid plants and electric furnace plants, respectively.

In the "wet process" phosphoric acid plant, the raw materials are ground phosphate rock, 93 percent sulfuric acid, and water. Phosphate rock is mixed with the sulfuric acid. This reaction produces phosphoric acid and gypsum. Following the reaction in the attack vessel, the mixture is filtered to separate the gypsum from the phosphoric acid. The gypsum is pumped as a slurry to a large pile near the facility where it is allowed to dewater. Since approximately 4 metric tons of gypsum are produced per ton of phosphoric acid, a large phosphoric acid plant would produce about 2.5 million metric tons of gypsum per year.[5] Approximately one percent of the radium-226, 60 to 80 percent of the thorium-230 and 80 percent of the uranium in phosphate rock are dissolved by digestion with sulfuric acid.

Table [illegible] lists the average radioactivity concentrations for the fertilizer products and phosphogypsum byproduct of several wet-process phosphoric acid facilities in Florida. Phosphoric acid samples were found to contain from 50,000 to 100,000 pCi/liter uranium-238 and about 1000 pCi/liter radium-226. The concentration of uranium appears to vary directly with the concentration of P_2O_5.

TABLE 2: Natural Radioactivity Concentrations in Materials Produced from Florida Phosphates (pCi/gram)

Material	Ra-226	U-238	Th-230	Th-232
Normal Superphosphate	21.3	20.1	18.0	0.6
Diammonium Phosphates	5.6	63	65	0.4
Concentrated Superphosphate	21	58	48	1.3
Monoammonium Phosphates	5	55	50	1.7
Phosphoric Acid*	1	25.3	28.3	3.1
Gypsum	33	6	13	0.3

*29 percent acid.

Each "wet process" phosphoric acid plant incorporates a large cooling pond (~ 500 acres) of contaminated water for recycle in the facility. During periods of excess rainfall it becomes necessary to discharge water from these ponds to nearby streams. Field studies at several Florida facilities indicate raw process water contains approximately 50 to 90 pCi total radium-226 per liter and approximately 400 to 2000 pCi/liter of uranium-238.

To prepare process water for discharge to the environment, the pH must be increased from 1.5-2.0 to 6-9. To accomplish this, slaked lime is normally added to the discharge water in a step called "double liming." Studies have shown that this treatment is highly effective in removing radionuclides from the effluent. Radium-226 reductions of greater than 96 percent were observed in all situations studied. Similar reductions in uranium and thorium were also observed. As a result of the effectiveness of this treatment, EPA discharge permits usually stipulate an acceptable pH range of 6-9 for treated effluent to ensure minimization of radioactivity in phosphoric acid plant discharges.

In the thermal processing of phosphate rock, silica and coke are added; this mixture is electrically reduced to form elemental phosphorus. Ferrophosphorus and calcium silicate slag byproducts are also formed in the process. Data from analyses of these samples indicated that most of the uranium and radium-226 present in the input phosphate rock is transferred to the slag during the process.

Reclaimed Land Use

Approximately 100,000 acres of land have been mined for phosphate rock in Florida. To date, about 25,000 acres of the mined lands have been reclaimed for residential and commercial development, farming, and grazing. It is estimated that about 1000 structures have been built on these lands. Since reclaimed lands are composed of overburden, leach zone material, matrix, sand tailings, and/or slimes, they frequently contain radium-226 concentration substantially higher than the 0.1 to 3 pCi/gram typical of U.S. soils. Concentrations up to 99 pCi/gram have been measured in these reclaimed soils. However, radium-226 concentrations in the reclaimed land soils generally range between 10 to 30 pCi/gram. Such radium-226 concentrations often persist to soil depths greater than 20 feet. Due to the elevated soil radium-226 concentrations, a considerable quantity of radon-222 is produced. This radon-222 diffuses to the ground surface and through the foundations of structures where it can lead to the buildup of short-lived radon daughters in the indoor environment. Data on average gross indoor radon [illegible] over a one-year period were obtained for several structures potential for radon on reclaimed land and on land distant from the Florida phosphate region. The data from these structures are summarized in Table 3.

TABLE 3: Percentage Range of Radon Daughter Levels

Reclaimed Land (n=13)	Nonreclaimed Land (n=9)
0.05 to 0.1 WL : 38%	0.05 WL : 44%
0.01 to 0.05 WL : 31%	0.01 to 0.05 WL : 22%
0 to 0.01 WL : 31%	0 to 0.01 WL : 7%

From the data obtained to date, it is believed that the demonstrated excess lung cancer risk associated with the higher levels, warrant additional studies to delineate more fully the scope and magnitude of this problem. Based on the assumption that excess lung cancers will double per 60 CWLM exposure, we can associate the highest annual average working level observed of 0.1 WL for continuous occupancy and an average lifetime (70+years), with a 6 to 10 times increase in lung cancer. This estimate of the doubling dose is based on the excess cancer observed in uranium miners. If, as seems likely now, the doubling dose is lower for a general population, the estimated health risk would be proportionately greater, possibly by as much as a factor of 2. We recognize, of course, the need for further efforts to reduce the large uncertainties in these risk estimates.

Principle Exposure Pathways

There are numerous pathways which could cause exposure to the public due to operation of the phosphate industry. These include exposures resulting from effluents, emissions, ground waters, using the industry's products and byproducts, living and working on reclaimed land, and working in the industry.

As shown by the data presented, the effluents from phosphate mine slime ponds and phosphoric acid plants are readily controllable to limit

total radium-226 discharges to surface waters to less than 3 to 4 pCi/liter. The two principal rivers in Florida receiving such effluents are the Alafia and Peace Rivers. Estimation of the population dose resulting from discharges to these rivers depends on the total discharges, the river characteristics, the number of mines discharging, and the downstream population. All of these factors are quite variable from year to year and season to season in Florida. However, it is highly unlikely that normal discharges from mines and phosphoric acid plants to these Rivers would result in radium-226 concentrations greater than 0.5 pCi/liter above normal to downstream users. However, accidental failure of slime pond dikes could significantly increase the radium-226 concentrations in the rivers since slimes contain greater than 2000 pCi/liter total radium-226.

Many shallow well water supplies in the Central Florida area have been shown to contain radium-226 concentrations greater than the limit of 5 pCi/liter radium-226 and radium-228 established by the Environmental Protection Agency's Safe Drinking Water Regulations of 1975. However, since no assessment was made of these ground waters prior to extensive mining, it is presently uncertain to what extent the levels are due to the natural presence of uranium in phosphate ores or the operations of the industry. Additional work is underway to investigate this question further.

Data collection and evaluation of air emissions from elemental phosphorus and phosphoric acid plants are incomplete. However, there are some preliminary indications that significant quantities of Po-210

may be emitted from these facilities due to volatization during calcining or furnace operations.

Workers in the phosphate operations come in close contact with large amounts of phosphate ores, products, and wastes along with inhalation of dust generated by unloading, crushing, drying, and other activities. The worst exposure situations were observed in areas of high dust concentrations and in or around the phosphoric reactor vessel and its associated equipment. Based on normal worker occupancy and radiation levels measured in Florida facilities, it has been estimated that direct gamma dose equivalents for workers in phosphoric acid or elemental phosphorous plants range from 30 to 300 mrem per year.[6] The annual dose equivalent rate to the tracheobronchial region of the lung, due to inhalation of radon daughters, has been estimated to be a high as 5 rem/yr for these workers. Estimates of the average lung dose would of course be very much lower.

From the data collected and analyzed to date, population exposures to the indoor radon daughters in structures appear to be the most significant public health problem, and efforts are being made to develop radiation protection guidelines to evaluate and control exposures to this source. As an interim measure, the EPA has provided the State of Florida a screening level which allows continued land development without a significant health impact. This interim guideline is based on a gamma exposure of less than 10 uR/hr (including background) which can be associated with a estimate of a radon daughter level less than 0.01 WL.

Other aspects of the industry which require further study include the impact of using byproduct slag and gypsum for construction materials,

the uptake of radionuclides by crops due to fertilizer use or growing on reclaimed lands, evaluating control technologies to limit indoor radon daughter levels, assessing the impact of recovering uranium fuel from phosphate materials, and the use of defluorinated phosphate as a livestock feed supplement.

Phosphates as Livestock Feed Supplements

The presence of uranium (2-180 ppm) in livestock feed supplements has been reported in proportion to their phosphorus content. This is due apparently to the transfer of uranium with phosphorus from the original rock phosphate to the feed regardless of whether the mineral has been chemically processed or used more or less directly. The occurrence of radium-226 in the feed supplements falls into two groups depending upon the type of phosphate material used, and is either 70% of the equilibrium amount or 6% of the equilibrum amount. Beneficiated defluorinated phosphate rock has been shown to contain the higher amount of radium-226 while feed supplements utilizing fertilizer phosphate as a primary source for phosphorus exhibit the lower radium concentration.

Based on metabolic studies of uranium ingested by dairy cattle with their normal diet, and assuming the maximum transfer of uranium (0.2%) from the highest U:P ratio feedstock directly to the cow's milk, the following data is derived.

> Elemental phosphorus requirement for 600 Kg lactating dairy cow - 55 gm/day
>
> Daily ingestion of uranium if supplement provides 25% P - 0.016 gm
>
> Concentrations of uranium in milk assuming 20 liters per day - 1.6 μgm/l

Thus it appears that the uranium content of feed supplements should not be of great health significance to the cow or man based on current guidelines.

Of greater concern was the radium in the supplements. Using the 600 Kg cow and assuming (1) that 0.02% of the dose of ingested radium-226 is secreted into the cow's milk per liter and (2) that the highest Ra:P ratio feed was used to provide 25% of the cow's P requirement, then the contribution of the supplement to the radium content of milk is calculated to be 0.7 pCi/l. The cow's daily ingestion from the supplement would be approximately 3253 pCi. Published volumes for radium in milk range from 0.09-0.3 pCi/l. The FRC recommended maximum permissible radium-226 dose for a human being is 20 pCi/day. In order to achieve this ingestion rate, one would have to consume approximately 29 liters of milk per day.[7]

As a result of the foregoing, the radium content of feed supplements does not appear sufficient to result in a significant transfer to human beings; however, more data would be desirable to confirm this conclusion.

One additional item of consideration with respect to natural radioactivity in feed supplements concerns the storage of large quantities of material in warehouses and the possibility of radon buildup resulting in occupational exposure. Data is currently unavailable for evaluation of this potential exposure, however research is currently on-going.

This Task Force does not consider the radioactivity in livestock feed supplements to be high priority at this time and will not include specific recommendations; however, we will continue to evaluate related studies and will defer any conclusion until such time that additional research in this area can be completed.

Uranium Recovery

Uranium for use as a fuel in nuclear power plants has historically been extracted from Western ores containing high concentrations of the element. At present, Western ores mined for their uranium values average about 0.2 percent uranium. With the existence of high grade ores waning and the price of uranium drastically increasing from $8 per pound to over $40 per pound, U_3O_8 low grade deposits are becoming more important.[8] Two types of ore which contain uranium in economically recoverable concentrations based on today's technology and economic climate are phosphates and copper.

The production of phosphoric acid results in dissolving about 80 percent of the uranium in the ore.[9] Through technology developed at Oak Ridge National Laboratory (ORNL) and several industries based upon solvent extraction methods, it is now possible to recover 90% or more of the uranium from the phosphoric acid.[10] Present estimates indicate that uranium can be recovered from large wet-process phosphoric acid plants for about $8 - $15 per pound U_3O_8 using variations of the basic ORNL process.[11]

All present practicable uranium recovery techniques for phosphates apply only to phosphoric acid. Further, recovery from lower production volume plants may be more risky and costly than others. While recovery from facilities with the capacity of 200,000 tons P_2O_5 per year may become practicable, initial recovery efforts will probably be restricted to larger facilities. In 1976, the capacity of wet-process phosphoric acid plants (greater than 200,000 tons P_2O_5 per year) was about 7 million

tons P_2O_5. This amount of phosphoric acid would contain about 10 million pounds of U_3O_8 which is enough to fuel about 20 one thousand megawatt(e) nuclear power plants per year.

The U.S. Environmental Protection Agency is presently conducting studies to determine the environmental impact of uranium recovery from phosphoric acid.[12] In general, potential exposures are anticipated to result from emissions, effluents, dust in calcining and packaging operations, and transportation. At this time, it is difficult to estimate the potential impact although it is not expected to be of major significance.

Uranium recovery from copper leach solutions through ion exchange is being considered at several mine-mill operations. The potential radiological impact of this recovery is unknown at present although it should be similar to ion exchange operations at present uranium mines.

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RADIOLOGICAL ASPECTS OF THORIUM AND DAUGHTER PRODUCTS

Thorium-232 is the 35th most abundant element in the earth's crust, 0.001 to 0.002 percent being most generally accepted. It is about three times more plentiful than uranium-238.

Uses

In addition to the thorium fuel cycle which is currently under investigation, thorium has long been used in the following non-nuclear applications:

A. Before the advent of nuclear energy, thorium was used chiefly in the manufacture of gas mantles because of the brilliant light-emitting qualities of their oxides. Even to this day, the Coleman gasoline camp lanterns find continued use with the mantles.

B. Thorium coated tungsten wire has been used for a long time as cathodes in vacuum tubes. Because of the low thermionic work function, the thorium can produce high electron emission. Where size of the focal spot is not vitally important (for example, water or oil-cooled therapy tubes), and in uses demanding high X-ray emission efficiency, thorium may replace tungsten to increase X-ray production and strength.

C. Tungsten-thorium alloys increase the efficiency of filaments for incandescent electric lamps.

D. Thorium finds increasing use in heliarc welding of tungsten electrodes because it offers the advantage of instant arc starting and arc stability. Welds made with 10 percent Th-Mo filler are ductile even at room temperature.

E. Thorium is an important alloying element in magnesium, imparting high strength and creep resistance at elevated temperatures. This is because of its high melting point (3220°C). Many aircraft structural parts contain this alloy with magnesium.

F. Thorium oxide is the most stable of the refractory oxides and has been used to a limited extent in specialized melting operations as crucible construction material and in other ceramics.

G. Highly purified thorium in small amounts goes into special optical glass giving it a high refractive index and a low dispersion. Consequently, they find application in high quality lenses for cameras and scientific instruments.

H. Thorium oxide also has industrial use as a catalytic agent for oxidizing sulfur dioxide to sulfur trioxide in the production of sulfuric acid, in the conversion of ammonia to nitric acid, in the making of water gas from carbon monoxide and in petroleum cracking.[1]

Sources

As a consequence of its inability to form soluble higher valency compounds, only six minerals containing thorium as an essential constituent are known, compared with about seventy uranium minerals.

Unlike most other rocks and minerals, thorium-bearing minerals are not soluble in water, and so are not destroyed during erosion. Instead, they accumulate as placer deposits in riverbeds and in sand on ocean beaches. There are thorium veins known to exist in this country and other places in the world just recently discovered. Should there be a heavy need for thorium in the future, these veins could be exploited. The three most significant minerals from which thorium has been recovered are monazite, thorianite, and thorite. The world's supply of thorium has been obtained most entirely from monazite. Monazite, $ThPO_4$, a phosphate, is very brittle, fractures unevenly and is radioactive. It is sufficiently magnetic to concentrate electromagnetically in a strong field. It occurs characteristically as a very minor accessory constituent in granitic and syenitic igneous rocks. These rocks seldom contain enough monazite to warrant recovery, but the natural process of erosion concentrates the monazite sands in the beach and stream deposits. Monazite occurs typically as small but distinctive round, glassy grains, colored honey-yellow to yellowish-brown. Thorium occurs as a trace element in various mineral deposits including rare earths.

The principal monazite sources have been the beach sands of India and Brazil. Monazite concentrations in commercial or near-commercial quantities can be found in numerous geographic areas within the continental U.S. On the East coast are stream placers or old beach deposits in the Carolinas and Florida (Jacksonville Beach). The most important commercial deposits are found in Idaho, particularly along stream and river placers on the western edge of the Idaho batholith, a large granite area in the

central part of the State. Thorianite, ThO_2, is soluble in sulfuric acid and is slightly less radioactive than pitchblende. Thorianite deposits in Madagascar are now being commercially worked for thorium. It is also being processed in Siberia and in New Zealand. Deposits have also been reported in sand and gravel beds in Canada, California and Montana. Thorite, $ThSiO_4$, the third major mineral containing thorium, is principally found in the beach sands in South Island, New Zealand. Similar deposits are found in Central California gold placers, especially along two rivers, the Tuolumne and Consumnes. As mentioned above, recent discovery of thorium veins in the U.S. containing either monazite, thorionite, thorite or all three, could provide a great resource for this country easily exploitable in the near future.[1]

Preparation

The established thorium extraction process starts from monazite, the chief commercial ore. Monazite is chemically inert, and the dissolution or "opening" process must be drastic; highly concentrated solutions of sulfuric acid or sodium hydroxide at 140°-150° C are used. The thorium phosphates are rapidly converted to sulfates with the liberation of phosphoric acid. Dissolution of thorium sulfate in water and basic separation by addition of ammonia gives essentially thorium hydroxide. The thorium hydroxide is dissolved in nitric acid and the slurry is allowed to go through an evaporation process. Another breakdown with sodium hydroxide followed by water washing produces the oxide. Also use is made of the oxide's magnetic quality allowing further magnetic separation from impurities. Good ventilation is necessary to carry away

the radioactive inert gas daughter thoron, which is liberated from the mineral or breakdown. The oxide, like the mineral thorianite, can then be reduced with calcium to form the pure metal.[2,3]

Radiological Problems

The extraction of thorium from its ores leaves thorium-238 in the purified material which decays directly to the short-lived ($T_{\frac{1}{2}}$ = 3.64 days) parent of thoron. Even outside of the mining and extraction of thorium, commercial non-nuclear usage, i.e., in mantle making, ceramics, electronic tube filament making and other such industrial handling, there is the hazard from the inhalation of thoron (radon-220), a daughter product of thorium. There are not near as many epidemiological studies compared to radon-222 in the providing of evidence for an exposure-risk relationship. Although natural levels of thorium in soil and construction materials are comparable to and in many cases exceed those of radium and uranium in the source form of monazite sands or thorite ores, there is not the seepage problem associated with thoron ($T_{\frac{1}{2}}$ = 55 seconds) as with radon-222 ($T_{\frac{1}{2}}$ = 3.8 days). The short half-life of thoron means that the air concentrations are generally much lower than those for radon whose diffusion period through the ground is comparable to its half-life. Thus the prime concern for the hazards relating to the daughter products of thorium are narrowed around the extracting processes and the industrial machining and usage of the metal.[4]

The deposition of the thoron decay products depends mainly on (1) the fraction of inhaled uncombined radioactive atoms; (2) the particle size distribution of the carrier aerosol of the combined radioactive

atoms; (3) the concentration of thoron daughters in the local atmosphere; and (4) the degree of attachment of these daughter ions to the aerosol particles. The consequent lung dose further depends on the deposition distribution in the tracheobronchial tree and within the various mucous layers. The "working level" concept originally applied only to radon daughters (MPC's). This has, however, been extended to include thoron daughters: A thoron working level used to be defined as equivalent to the potential alpha energy released due to the complete decay of 100 pCi of each of the thoron daughters per liter of air as was done for radon. However, now the preferred definition is merely any combination of radon daughters including thoron which on ultimate decay will liberate 1.3×10^5 MeV of potential alpha energy per liter of air. In light of the original definition, the thoron WL appears to be 13.5 times greater than the radon WL.[5] This allusion does not, however, mean that the dose to the respiratory tract on exposure to one thoron WL is also 13.5 times greater than that due to an exposure to 1 WL of radon daughter. The radon daughter elements deposited in the lungs undergo radioactive decay at the site of deposition because of the short half-lives involved, whereas the thoron daughters, having comparatively longer half-lives, are partially eliminated by biological processes. In fact, calculation shows that approximately 43% of the thoron daughters only remain with the lungs and decay. This translates to a lung dose of only about 5.8 times that delivered by an equivalent concentration of radon daughters, all other conditions being identical.

The latest radiological activity surrounding thorium-thoron and the daughter products has been a decision on the most correct lung model to

be used in describing inhalation doses. There has been a preponderance of models from different authors all producing widely uncomparable results. In spite of the differences, the studies all agreed in conclusion, that the highest alpha dose should be expected in the upper and medium bronchial region. This result is of great importance in the interpretation of the enhanced lung cancer mortality.

The current problems, concerns and activities surrounding the radiological aspects of thorium, thoron and the daughter products seems to be centered around the determination of doses to the various respiratory-related organs as a result of inhalation of the daughter products. In summary thorium seems to present just as much of a problem as does radon and its daughters, but perhaps of a slightly less hazardous nature.

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RADIOACTIVITY IN FOSSIL FUELS

Description of the Problem

It is generally known that fossil fuels contain trace quantities of naturally-occurring radioactive materials in the uranium and thorium decay series. The U. S. consumes vast quantities of fossil fuels. For example, a single 1000 MW_e coal-fired power plant would consume about 560 tons of coal each hour, and would produce about one-half million tons per year of ash. Even with only trace quantities of radioactive materials in these fuels, the large volumes of fuels consumed would involve the re-distribution of significant amounts of radioactivity into the environment. The problem addressed in this section of the report is what the States, EPA and other members of the Conference should do in evaluating, monitoring, or controlling radioactivity from the utilization of fossil fuels.

Activities to Date

The radiological impact of natural gas as a source of radon has been evaluated by several investigators.[1,2,3] The Bureau of Radiological Health has conducted a radiological survey of an oil-burning power plant,[4] and the EPA has conducted a similar study around a coal-fired power plant utilizing Eastern coal.[5] The Oak Ridge National Laboratory has measured trace elements at the Allen coal-fired steam plant which also utilizes Eastern coal.[6] The Idaho Department of Health and Welfare, Radiation Control Section has performed an evaluation of a proposed 1000 MW_e coal-fired power plant which would utilize Western coal.[7]

The conclusions and recommendations in this report are based in part on the results of the above studies.

Areas of Concern

A. Natural Gas

Natural gas contains radon which emanates from radium-bearing geological strata. Doses to the bronchial epithelium from radon in natural gas consumed in homes have been estimated by the EPA.[8] The annual dose committment from radon in natural gas has been estimated to be 2.73×10^6 man-rem. This dose committment is based on radon concentrations in natural gas being 20 pCi/l.

The annual dose committment appears to be insignificant when compared to background and other sources of radiation. In view of the limited future use of natural gas due to depleting supplies, the total environmental dose committment becomes even less significant.

It appears there could be a localized problem with natural gas if concentrations of radon in gas approached 357 pCi/l. This may be possible for users close to the well head. Such levels could result in radon concentrations in houses on the order of 1.0 pCi/l which might cause indoor radon daughter levels to approach .01 WL.

In view of the small potential dose committment from radon in natural gas and the high radon concentrations in natural gas

necessary for a local radon problem, no recommendations are made here regarding the continuous evaluation or monitoring of radon gas exposures from the use of natural gas. However, evaluation of specific uses, especially those close to well heads may be warranted.

B. Oil

Oil contains trace quantities of uranium, thorium and their daughters. No environmental distribution of radioactive material around an oil-fired power plant was observed by Gordan. The radium-226 content of fly ash from oil-fired plants is 21 times less than the radium-226 content of fly ash from coal-fired plants.[9] It would appear then, that the radiological impact from oil-fired plants would be insignificant. Therefore, no recommendations are made here regarding the continuous evaluation or monitoring of oil-fired plants.

C. Coal

It is generally known that coals contain varying concentrations of uranium and thorium and the radioactive daughters of these elements.

According to a paper by J. D. Vine[10], large potential reserves of uranium are contained in coal and lignite, and the concentration of uranium in the ash of coal provides a possible means of recovering uranium as a byproduct. Uranium bearing lignite occurs in the Fort Union formation of Paleocene age in the Northern Great Plains, in the Salt Lake formation of

Pliocene age in Southern Idaho, and in Tertiary sediments in Nevada and Southern California. Uranium-bearing coal is present in the Wasatch formation of Eocene age in Wyoming, in the Laramie formation of Cretaceous age in Colorado, and in the Bear River formation of Cretaceous age in Idaho. Bituminous coal and anthracite in North Central and Eastern United States contain only very small quantities of uranium.

Vine reports that the uranium content of Western lignite varies from 0.001 to about 10 percent and averages 0.008 percent. Western coal samples from Wyoming and Idaho range from about 0.001 to 0.05 percent uranium with averages being about 0.003 percent for Wyoming and 0.05 percent for Idaho. The author points out however, that high rank, low ash coals of the type most desired for fuels are rarely uraniferous.

In a paper by Stocking and Page[11], the uranium content of coals, especially Western coals, is reported. The difference in the uranium content of different types of coals is not entirely a consequence of the availability of uranium. Laboratory experiments demonstrate that peat, lignite, and subbituminous coal extract 98% of the contained uranium from aqueous solutions, while bituminous and anthracite (the type of coal most suitable for fuel) capture less than a fifth and slightly more than a third, respectively.

In a report by Abernethy and Gibson[12], values of the uranium concentrations in Western coal are, on the average, 2 to 100 times higher than in Eastern coal.

The radiological implications of the extensive combustion of fossil fuels has been briefly dealt with in past literature. The radium-226 concentrations in European coals, in fly ash from power plants, and in contemporary fossil snows have been documented. The radium-226 releases into the atmosphere as a result of burning Appalachian coal have also been discussed.[14]

There has apparently been very limited radiological surveillance conducted around fossil fuel power plants. Two such studies were conducted by the Oak Ridge National Laboratory and the U.S. Environmental Protection Agency.[15] Both studies concluded that the utilization of fossil fuels for power generation does not present a significant radiological health concern; however, it has been pointed out by Rowe that these conclusions may require modification for the utilization of Western coal, since both studies addressed only low uranium content Eastern coal.[16]

The State of Idaho has evaluated the radiological impact from a proposed 1000 MW_e coal-fired plant to be constructed near Boise.[17] The proposed plant, called Pioneer, was to utilize Western coal from Wyoming.

Table 1 gives the estimated releases from such a plant and compares these releases to maximum permissible release concentrations to unrestricted areas. The assumptions used in calculating the releases are listed in footnote (1) of the

Table. For example, it was assumed that the concentrations of uranium and daughters was 0.23 pCi/g and the concentration of thorium and daughters was 0.18 pCi/g. The efficiency of the electrostatic precipitators was assumed to be 99.7%. It should be emphasized that if lower grade coals containing more radioactivity are used, and if the efficiency of effluent controls goes down, then radioactivity releases could be much higher for a specific power plant.

Table 2 gives the results of fly ash analysis taken from the Idaho report. The fly ash samples were from the Jim Bridger coal-fired plant in Wyoming, which utilizes Wyoming coal from a different deposit than the proposed Pioneer coal. It can be seen from Table 2 that radium concentrations in fly ash average about 3.1 pCi/g. It appears possible that radon levels in houses constructed in fly ash disposal areas could be elevated.

In view of the Idaho report and other papers cited here, the following recommendations are made regarding coal-fired plants:

(1) The radiological aspects of each proposed coal-fired plant should be evaluated by the appropriate agencies. Radioactive releases should be estimated from an analysis of the coal to be consumed and the operating characteristics of the proposed plant. This recommendation is especially pertinent to any proposed plant that intends to derive its fuel from low-grade, uraniferous Western coals.

(2) The States, the EPA, or other appropriate agencies should proceed to measure radon levels on fly ash piles and in structures built in fly ash disposal areas, if there are any.

(3) The EPA and appropriate States should study and evaluate uses of fly ash from coal-fired plants, particularly those uses which are likely to cause elevated radon levels in habitable structures.

TABLE 1

ESTIMATED AVERAGE AIRBORNE RADIOACTIVITY RELEASES FROM PIONEER - TWO UNITS, 500 Mw_e EACH[1]

ISOTOPE	RELEASE CONCENTRATION (μCi/ml x 10^{14})	MAX. PERMISSIBLE RELEASE[2] CONCENTRATIONS (μCi/ml x 10^{14})	% RELEASE STANDARD	RELEASE/YR (Ci)
Uranium Series				
^{238}U	4.63	200	2.31	0.0031
^{234}U	4.63	400	1.15	0.0031
^{230}Th	4.63	30	15.43	0.0031
^{226}Ra	4.63	200	2.31	0.0031
^{222}Rn	1545	300,000	0.52	1.0280
^{210}Po	1545	700	220	1.0280
^{210}Pb	4.63	800	0.57	0.0031
Thorium Series				
^{232}Th	3.63	100	3.63	0.0024
^{228}Ra	3.63	100	3.63	0.0024
^{228}Th	3.63	20	18.15	0.0024
^{224}Ra	3.63	2,000	0.18	0.0024
^{212}Pb	3.63	70,000	0.01	0.0024
TOTALS			267.89 %	2.083 Ci

(1) Assumes: (a) 0.3% release of all trace elements in coal except ^{210}Po and ^{222}Rn (100% release).
(b) Uranium and thorium are in equilibrium with daughters.
(c) Coal contains 0.23 pCi/g U and 0.18 pCi/g Th.
(d) Total effluent is 4.48 x 10^6 ft^3/min.
(e) 562 tons of coal is consumed per hour.

(2) Maximum permissable releases of airborne radioactive materials to unrestricted areas - Idaho Radiation Control Regulations, Part C, Appendix A, Table II. Assumes radioactive materials particulates are insoluble.

TABLE 2
Jim Bridger Fly Ash

ANALYZED BY	ISOTOPE	Fly Ash #1 (12/75) pCi/g(1) ± σ(2)	Fly Ash #2 (11/75) pCi/g(1) ± σ(2)
	Uranium Series		
LFE	^{238}U	8.58 ± 1	2.57 ± .3
LFE	^{230}Th	7.0 ± .4	3.3 ± .2
LFE	^{226}Ra	4.9 ± .2	2.4 ± .1
RCS	^{226}Ra	3.8 ± .3	—
LFE	^{210}Pb	18.0 ± 1	3.0 ± .2
	Thorium Series		
LFE	^{232}Th	4.7 ± .3	2.8 ± .2

NOTES:

(1) Based on dry weight.

(2) σ is one standard deviation due to counting statistics.

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RADIUM AND RADON IN GROUND WATER

Description of the Problem

It is common for ground water in areas throughout the United States to have widely varying concentrations of radium, radon and radioactive daughter products. Radium may range from trace quantities to over 50 pCi/l, while radon (both in the presence and absence of significant radium concentrations) may be found in concentrations greater than 50,000 pCi/l.[1,2,3] As a noble gas, radon in water is readily released with mild aeration. However, the radioactive daughters in various stages of equilibrium will remain in the water. Because of the potentially high levels of radon in water, it is possible in private and commercial uses to create significant working levels in air.

Radium in water and radon in air have been the subjects of much Federal interest and research, resulting in appropriate standards. However, inadequate attention has been devoted to the health significance of ingested and inhaled radon and radon daughters from potable water supplies. A large segment of our population is subjected to exposure to all of these naturally-occurring radioisotopes through the use of private, commercial, agricultural, public, and community ground water supplies. In addition, geothermal water may become another significant source of highly contaminated water.

Activities to Date

After extensive study and deliberation, Federal regulations have been established for radium in drinking water.[4] Additionally, several

States have initiated and/or completed studies of radium and radon concentrations in their ground water supplies. However, only a limited amount of research has been done on the health effects of ingesting radon and its daughters. Much of what has been done is through uncoordinated efforts and has been inconclusive. Researchers have evaluated the radium decontamination characteristics of conventional water treatment techniques and have also developed and evaluated special radium removal techniques.[5]

Area of Concern

If the EPA drinking water regulations are to be enforced, there will be a large number of ground water supplies which will be required to undergo radium removal procedures. As this occurs, consideration will have to be given to the disposition and disposal of radium contaminated water treatment residues.

While much work has been done on the health effects of radon in air, very little has been done to quantitatively evaluate the health significance of high radon levels in water as a source of airborne concentrations. Judging from the high levels which are seen in water and from the private and commercial uses of water in poorly ventilated areas, airborne radon due to aeration of water may have significance. Duncan, et. al. have calculated that water with 1000 pCi/l radon-222 could result in air concentrations of 1 pCi/l due to normal residential use.[6] Further, they have estimated that if a population uses potable water with a radon concentration of 500 pCi/l, 20 health effects per year might result from inhalation for every one million people exposed.

Because of the comparatively large concentrations associated with radon and its daughters in drinking water, it is desirable to better

understand the health significance of these radioisotopes from the viewpoint of ingestion. Little work has been done in this area, yet it is not uncommon for individuals over their entire lifetimes to daily ingest radon and its daughters, as well as radium, from highly contaminated supplies. Further, since contaminated ground and geothermal waters may be used for food crop irrigation, there are questions which must be answered regarding human exposure through the food chain. Before the public health impact of the sources can be determined, considerable laboratory study is needed in the area of radium and radon daughter transport and dosimetry in the environment and man.

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MINERAL EXTRACTION AND PROCESSING ACTIVITIES

Numerous industries such as copper, fluorospar, vanadium, bauxite, titanium, and rare earth's mining and processing, extract ores which often occur in strata containing above-average concentrations of uranium, thorium, and their daughter products.

Unfortunately, little investigation has been done to assess the radiological impacts from these industries. However, sufficient data is available to provide an overview of the copper mining and processing industry and the zirconium extraction process.

Major copper deposits occur generally in three regions of the United States. These are the Appalachian Province, Keweenaw Peninsula (upper Michigan), and the Cordilleran Province (southwest United States). The latter region, consisting of Arizona, Utah, New Mexico, and Nevada, encompasses the largest deposits.

Open pit mining accounted for 89 percent of the copper extracted in the United States in 1973 with underground mining method supplying the remainder. There are three broad methods of beneficiation utilized in the copper industry. They are hydrometallurgical processing, physical-chemical separation, and leach-precipitation-floatation.

The processing activities involve the production of solid wastes, effluent streams and emissions.

As Figure 1 details, four areas of potential occupational and/or public radiation exposure have been identified for the copper industry. For underground mining, hazardous occupational radon levels could result

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*This subject was excerpted from the following paper: Fitzgerald, Joseph, "Radioactivity in the Copper Ore Mining and Dressing Industry", Proceeding of the Tenth Midyear Topical Symposium of the Health Physics Society, pp. 58-80, October 11-13, 1976, Saratoga, New York.

from the presence of a uranium co-deposit. With open pit mining, the disposal, storage, or utilization of waste rock from a uraniferous deposit could have a public health impact if not controlled. Likewise, the discharge of large volumes of pumpout water and seepage from tailing ponds could have an impact on public water supplies in the area. Efforts have been made to collect radiological data, when available, for each of these effluent pathways.

Still provides a detailed analysis of the uranium and thorium occurrence in porphyry copper deposits.[1] His geological surveys at the Copper Cities and Castle Dome open pit mines in Arizona have shown a positive relationship between the deposition of uranium and the occurrence of porphyry copper. Assay data from the Copper Cities mine showed uranium concentrations in the ore body to exceed that of normal igneous rocks of the same composition in this region (about 4 ppm) by a factor of 11 to 38. An average U_3O_8 concentration of up to 100 ppm is estimated for the primary copper zone of this mine.

Similar surveys performed at Castle Dome also showed evidence of this geological co-occurrence, although at lower uranium concentrations of about 20 ppm. Frequency distribution graphs for the uranium assay data obtained from 45 feet bench composite samples, are provided for the two mines in Figures 2 and 3.

Still concludes that uranium exists in commercially extractable concentrations in a number of porphyry copper deposits. The author notes, however, that assays at other mines showing relatively low uranium content indicate that the occurrence of uranium with copper is a site-specific phenomenon associated with certain geological conditions.

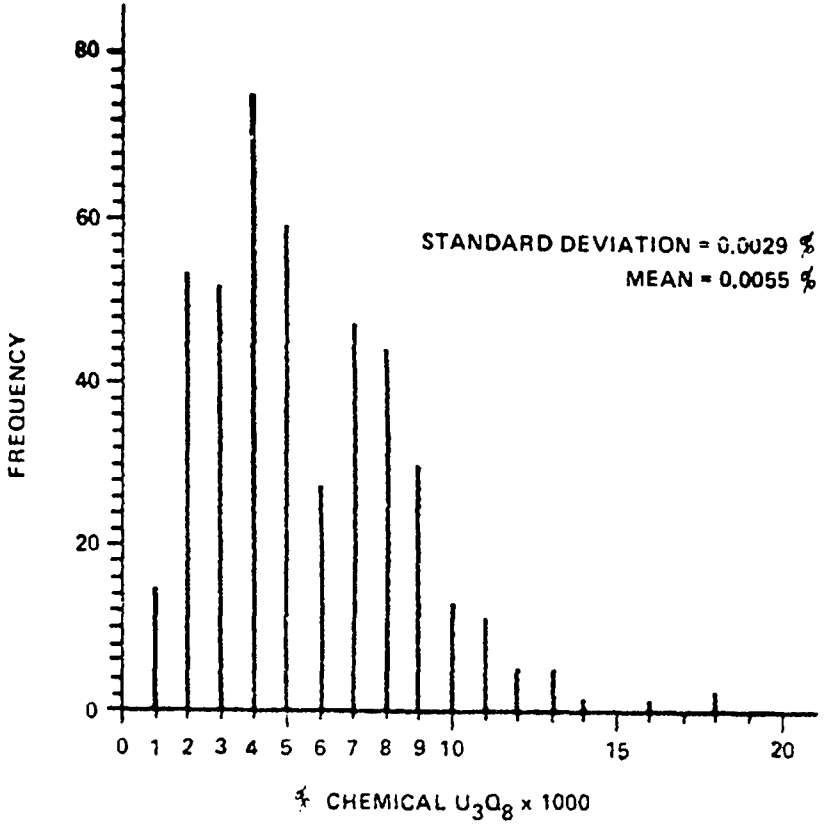

FIGURE 2

FREQUENCY DISTRIBUTION GRAPH

URANIUM CHEMICAL ASSAY DATA, COPPER CITIES

441 SAMPLES

FROM STILL, A.P., URANIUM AT COPPER CITIES AND OTHER PORPHYRY COPPER DEPOSITS, MIAMI DISTRICT, ARIZONA (UNPUB. THESIS), HARVARD UNIVERSITY, CAMBRIDGE, MASSACHUSETTS (1962). REPRINTED BY PERMISSION.

Moxham, et al. measured the radiation levels of hydrothermally altered rocks in the vicinity of several copper and copper-lead-zinc deposits in Arizona.[2] The study sites were the Bagdad porphyry copper deposit, the United Verde sulfide copper deposit, and the Old Dick and Iron King copper-lead-zinc sulfide deposits. The survey measurements obtained for the Bagdad deposit are graphed in Figure 4.

A recent geological survey of uranium concentration is reported by Davis and Guilbert for several open pit porphyry copper mines in Arizona and New Mexico.[3] Their results are tabulated in Table 2. These levels are not consistent with background levels, which are on the order of 4 ppm for igneous rocks of this region. One explanation for these observations is that the ore samples analyzed are not from the highly mineralized primary ore zone, but from associated zones. Assuming, though, average concentrations in the mineralized zones an order of magnitude higher than they observed, the low levels still resulting would also lend support to the premise of site-specificity for uranium occurrence.

Another source of radiological data is the pumpout waters of mines, both open pit and underground. The Office of Radiation Programs, EPA, undertook a survey of radioactivity levels in such effluent for copper mines in Michigan, Montana, and Arizona.[4] The mine operations surveyed are the White Pine (Michigan), Butte (Montana), the Old Reliable, and Bagdad (both Arizona). The radioactivity levels measured are tabulated in Table 3.

In several underground copper mines, radon daughter concentration levels have posed potential health hazards to mining personnel. As part

TABLE 2

Average Abundances of Uranium in Porphyry Copper Intrusions and Barren Intrusions of Similar Composition*

Location	Occurrence	Samples	Average ppm Uranium	Range ppm Uranium
New Cornelia Mine	mineralized stock	17	1.25	2.1-0.4
New Cornelia Pluton	barren intrusion	12	0.68	1.3-0.2
Mineral Park Mine	mineralized stock	25	0.79	3.5-0.1
Gross Peak-Martin Ridge	mineralized stock	13	0.46	0.9-0.2
Turquoise Mountain	mineralized stock	23	1.27	2.4-0.6
Morenci Pit	mineralized stock	23	0.71	2.5-0.2
Morenci Stock	barren stock	-	---	---
Santa Rita Deposit	mineralized stock	32	1.24	4.7-0.3
Fierro-Hanover	barren stock	6	0.52	0.3-0.8

*Uranium spectrometrically analyzed. All analyses are from whole-rock samples except for soil samples of uranium at New Cornelia Pluton, Gross Peak-Martin Ridge, and Turquoise Mountain.

TABLE 3

RADON-DAUGHTER SAMPLE CONCENTRATIONS
IN UNDERGROUND COPPER MINES BY STATE[1]

Mine	Location	No of Samples	Average Radon Daughter Conc. (WL)	Range of Radon Daughter Conc. (WL)	Approx. Avg. Existing Ventilation (CFM)
Indian Creek*	Missouri	8	0.023	.004 -0.116	Not Available
Virurnum*	Missouri	8	0.065	.002 -0.117	Not Available
Eagle Mine**	Colorado	10	Less than .1	Not Available	Not Available
85 Mine	New Mexico	10	0.180	0.060 -0.280	10,000
White Pine	Michigan	11	0.030	0.01 -0.04	330,000
Calloway	Tennessee	11	Negligible	Negligible	Not Available
Copper Queen	Arizona	42	0.330	0.02 - 1.7	5,000

*Lead, Zinc Copper
**Lead, Zinc, Copper, Gold, Silver

[1]Data provided by U. S. Dept. of the Interior, Mining Enforcement and Safety Administration, Denver, Colorado.

of its program to determine which mines require control technology, the Mining Enforcement and Safety Administration (MESA) of the Department of the Interior has surveyed a number of underground mines for radon daughter activity levels.[5] High activity is a strong indication that above background concentrations of uranium exist within the copper matrix. The radon daughter concentration data for underground mines in Arizona, New Mexico, Michigan, Colorado, Missouri, and Tennessee are given in Table 4.

As the data shows, the highest levels were recorded in the Arizona and New Mexico mines (average radon working levels of .33 and .18, respectively), which correlates with data showing uranium concentrations in soil ranging from 2 to 10 times background levels. The Michigan, Colorado, and Missouri mines all showed radon daughter levels less than .1 WL, while the copper mine sampled in Tennessee showed a negligible concentration of radon daughters. The amount of mine ventilation has a direct effect on the radon daughter concentration. Although ventilation information was only available for three mines, the average amount of air circulated in those mines varied by as much as a factor of 60.

The potential for radiation exposure from the products, by-products, and wastes of the copper industry may be addressed for two groups - the general population and those occupationally exposed. For the former group, the potential environmental interfaces would include exposure due to: 1) utilization of copper mill tailings; 2) construction on reclaimed uraniferous mining land; 3) seepage from tailing ponds; and 4) radon gas emanation and dust from waste piles.

TABLE 4

RADIOLOGICAL ANALYSIS OF COPPER MINE
PUMPOUT WATER*

MINE	SOURCE	Ra-226 (pCi/l)	TOTAL URANIUM (mg/l)
White Pine (Michigan)	Mine Water Discharge #3	13.6	< 0.1
	Mine Water Discharge #2	4.2	< 0.1
	Mine Water Discharge #3	27.3	< 0.1
Butte (Montana)	Kelley Operation Mine Water	4.8	< 0.2
	Berkley Pite Mine Water	1.5	< 0.1
	Continental East Mine Water	3.0	< 0.1
	Input to Emergency Pond	1.7	< 0.1
Old Reliable (Arizona)	300' Level Mine Drainage	2.6	< 0.1
Bagdad (Arizona)	East Pit Mine Water	1.6	< 0.1
	West Pit Mine Water	12.7	0.25

*Performed under contract to U.S. Environmental Protection Agency, Effluent Guidelines Division

Utilization of copper mill tailings has been of a negligible scope despite the vast amounts available. While other mineral tailing wastes find suitable applications, there are a number of reasons why copper mill tailings have not:

1. The projected growth for building construction in the Southwest and Mountain States is the lowest in the country.
2. Transportation costs would make its use uneconomical outside of the immediate vicinity of the mine.
3. Due to its high silicate content, impaction is difficult making its use as a construction or fill material unsuitable in many cases.

At several mines near population centers, however, tailings have been utilized in road construction and as general land-fill material (often being mixed with other soils to improve impaction). As no records are maintained concerning the amounts of tailings removed for these purposes, only estimates are possible. For a mine near Tucson, Arizona, for example, approximately 500 thousand tons of tailings were utilized for road construction. A small but indeterminable amount of tailings from this mine were also used for land-fills and construction material.[6]

Pigott, et al. demonstrated the suitability of tailings materials in the production of dry-pressed building bricks.[7] Pilot-plant production illustrated that bricks of superior quality were possible. A barrier to commercialization, however, was again, the distance between the source material and the market. If the price of construction materials continues to climb at the present rate, though, these bricks may become competitive.

The potential radiation problem associated with the use of these tailings which contain an, as yet, undetermined amount of uranium is at best conjecture. If uranium concentrations do prove significant (at least an order of magnitude higher than background), then the potential hazard would be greatest in homes either built with materials constructed from the tailings, or where tailing material has been used for fill or grading. The radiation exposure from construction materials and landfills would be a result of direct gamma radiation and lung exposure due to radon daughter alpha radiation.

In the University of Arizona's publication "A Balanced Approach to Resource Extraction and Creative Land Development", the task force involved proposed the long-term development of copper waste heaps and ponds for residential and commercial use.[8] Their plans called for "satellite" communities to be built on the terraced piles with agriculture and commercial zones. Although use of reclaimed copper mining sites is very small at present, the proximity of cities such as Butte, Montana, Salt Lake City, Utah, and Tucson, Arzona, to large mining operations makes future development a possibility. This potential is further enhanced with the ongoing environmental drive to climinate such eyesores as mined-out open pit areas and abandoned waste piles.

The potential exposure from uranium and its daughter products is again, difficult to ascertain without measuring the levels of the waste material. There is a possibility that homes built on, or adjacent to, former settling ponds would experience even greater radiation exposure due to higher residual uranium concentrations from the dewatered slimes and tailings.

It is evident that elevated radioactivity levels have been found to be associated with some copper mine pumpout waters. As an active mine must be continuously pumped, this effluent presents a potential groundwater contamination, particularly at the volume typical of a mining operation (1-10 million gpd). Likewise, as a high sulfur content is often associated with copper tailings, sulfuric acid produced with rainwater can gradually leach out uranium and its daughter products from storage piles. These effluents should meet Federal and State discharge standards where applicable before such releases are permitted.

Radon gas emanation and blowing dust are potential problems associated chiefly with tailing piles and dry evaporation ponds. While radon gas would be given off as a daughter product of the radium in the waste material, any potential hazard would be that associated with emanation and diffusion into closed structures.

Occupational exposure to natural radiation in the copper mining and dressing industry can be divided into two sectors, the mine and the mill. In an underground mine, exposure would be primarily due to radon daughter products and the critical organ would be the lung. However, as most mining is open pit (42 of 56 active mining operations), exposures would only be of concern on an individual mine basis. In the mill, radiation exposure would be dependent upon the type of beneficiation being performed. For enclosed areas, radioactive particulates may pose a problem. However, as no data has been collected concerning occupational radiation levels in copper mills, no conclusions can be made at this

time. Additionally, uranium recovery operations utilizing copper leachate solutions could result in some occupational exposure. Such operations would involve the handling and processing of concentrated leachate, and the handling, transportation, and storage of the yellow cake.

The obvious conclusion from this preliminary evaluation is that there is a paucity of available radiological data concerning copper mining and milling wastes. Although it is apparent that the generally low concentrations of uranium found in primary ore has discouraged such investigation, significantly elevated radionuclide concentrations in underground mine atmospheres, mine runoff effluent, and leaching solutions tends to refute the conclusion that a potential source of radiation exposure cannot exist. There are numerous chemical and physical processes by which the uranium concentration of waste materials can be increased, including dewatering, leaching, and precipitation. From a review of the beneficiation processes, experience from EPA's current phosphate industry study, and taking into consideration the chemical properties of uranium, it is likely that a large fraction of the uranium is ultimately discharged with the tailings. A need exists for a radiological impact assessment of this and other mining and milling effluents. This analysis should include primary ore, waste rock, beneficiation solutions, tailings, and all effluent streams.

There is a growing consideration given to the utilization of these waste materials for construction and landfill purposes which could lead to an increased public exposure. With the immense amount of waste materials being generated, the likelihood of reclamation and utilization is becoming greater. As the most significant potential exposure problem is the construction of homes on uraniferous reclaimed land, the pattern

of population growth is critical. At the present time, the lack of development in the immediate copper mining areas of the Southwest make such reclamation less of a concern. However, at present growth rates in these "sunshine" States, this situation could change dramatically.

Occupationally hazardous radon working levels have been measured in a number of underground copper mines. Although measures are being taken to rectify this problem, there are other facets of the mining and milling process which have a potential for occupational exposure which deserve scrutiny. Enclosed structures in which beneficiation solutions are exposed to open air, for example, should be monitored to determine the working level exposure to radon daughters.

Similar preliminary assessments are needed for other mining industries to determine their potential radiological impact and the need for field studies to document impacts.

ZIRCONIUM EXTRACTION PROCESS

In the zirconium extraction process, zircon ore (sand) is dressed with coke in a ball mill to a very fine consistency. This step may be preceded by a magnetic separation to remove some thorium contamination from the sand.

The coke-zircon mixture is introduced into a chlorination reaction chamber, the temperature elevated to 1200°C, and chlorine gas is reacted with the mixture. The primary reaction is:

$$ZrO_2.SiO_2 + 2C + 4Cl_2 \xrightarrow[1200°C]{H} Zr\,Cl_4 + SiCl_4 + 2CO_2$$

A two-stage condenser (H_2O:$SiCl_4$) separates crude $Zr(Hf)Cl_4$ from $SiCl_4$ which is processed for sale.

The process now proceeds to the Zr-Hf separation. This is a solvent extraction process using methylisobutylketone and an aqueous solution of

ammonium thiocyanate. The Hf is carried into the MIBK fraction and the completeness of this separation is measured by activiating the natural Hf-180 to radioactive Hf-181 using Cf-252 sources. The Zr is carried into the aqueous phase as zirconium oxychloride ($ZrOCl_2$) and is precipitated as the sulfate with the NH_4Cl being further processed to recover NH_4OH. The $Zr(SO_4)_2$ is repulped with aqua ammonia (NH_4OH) to form $Zr(OH)_4$ and ammonium sulfate which is boiled down and can be used as fertilizer. The zirconium hydroxide ($Zr(OH)_4$) is filtered and sent to the calciner where it is fired to the oxide (ZrO_2).

The zirconium oxide is then remixed with the coke and sent through a pure chlorinator to yield $ZrCl_4$ free of Hf. The zirconium chloride is condensed and goes to the magnesium reduction process (known as the Kroll process). Here, magnesium metal is reacted with $ZrCl_4$ to produce magnesium chloride ($MgCl_2$) and zirconium sponge (metal). A flow chart illustration is presented in Figure 6.

The first residue generated by this process is the sand chlorinator residue (tailings). This residue is primarily coke (90%), unreacted silicates, and non-volatile chlorides. There is a significant amount of Ra-226 and daughters in soluble form in this residue (ranging from 150-1300 pCi/g(dry)). The chlorinator residue ends up in a pile as indicated in Figure 6. There are also drains and general waste from this process that are sent to pond 1A, which is a holding pond for the clarifier. There is a turnover in the caustic alkali scrubbers used here that is partially recovered and partially sent to the clarifier.

The sludge coming from the clarifier comprises the largest volume of waste generated in the zirconium extraction process. Radium-226 concentrations in sludge have been measured between 87 and 154 pCi/g(dry).

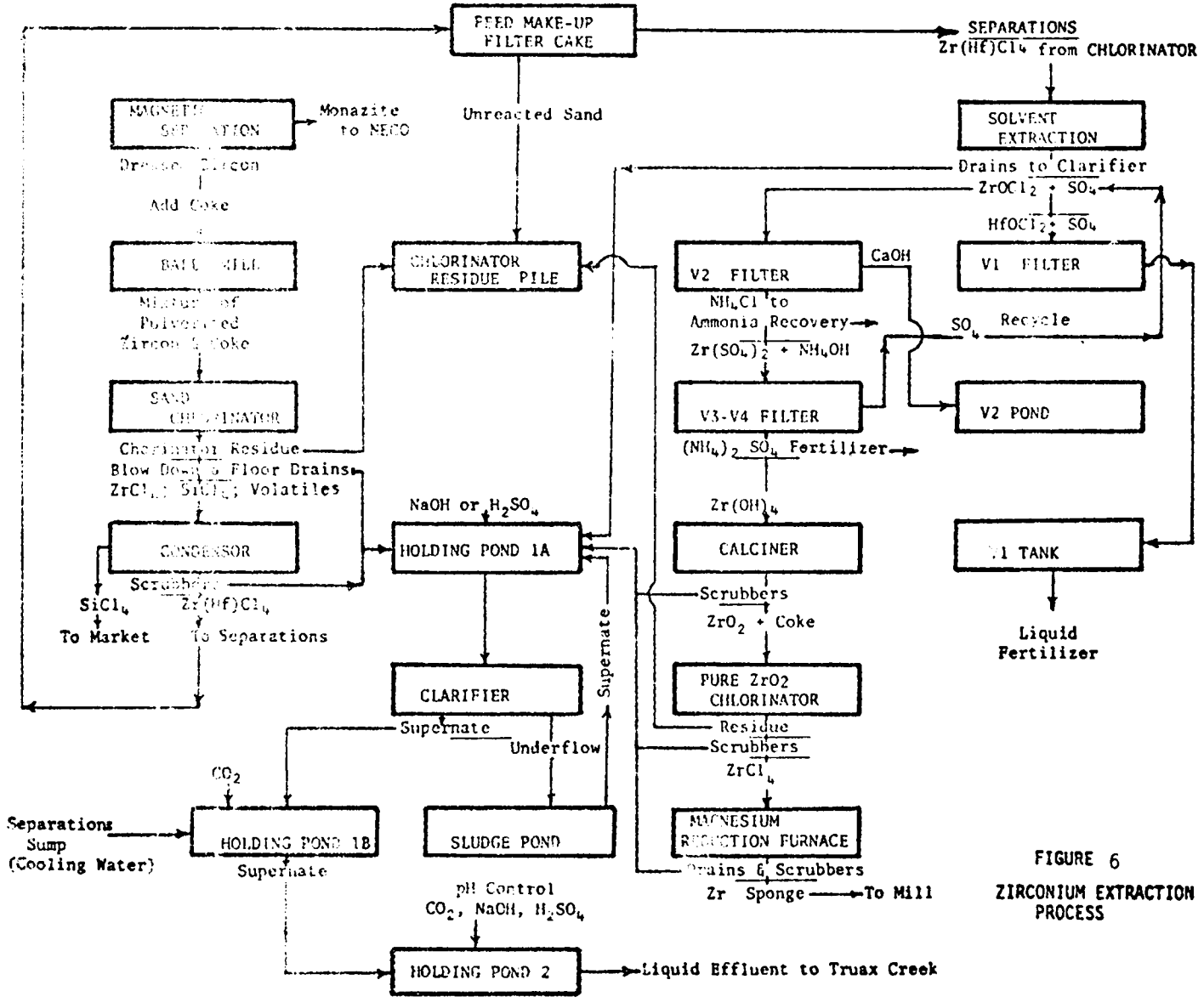

FIGURE 6

ZIRCONIUM EXTRACTION PROCESS

In the sulfate precipitation (V_2) ammonium chloride is generated and is sent to a recovery site to recover ammonia. The ammonium chloride is operated by the neutralization of HCl with NRH OH (ammonia).

The $Zr(SO_4)_2$ is repulped with ammonium hydroxide to form $Zr(OH)_4$. This process yields ammonium sulfate ($(NH_4)_2SO_4$) which is further processed to liquid fertilizer.

There are scrubbers associated with the final three (3) steps: the calciner, the pure chlorinator, and the Kroll process magnesium reduction furnaces. These scrubbers are vented to the clarifier. In addition to this, there is a pure chlorinator residue that is also dumped on the residue pile. The pure chlorinator residue contains about 200 pCi/g R-226.

The most significant radiological problem presented by the zirconium extraction process appears to be the potential contamination of surface or ground water from the chlorinator residues. It has been demonstrated that the radium in these residues is extremely soluble. Radium-226 concentration in water under a chlorinator residue pile has been measured to be as high as 45,000 pCi/l.

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RADON IN CAVES

It has recently come to the attention of State radiological health officials that potential radiation hazards may exist in a number of National Park Service caves and "cave air" conditioned buildings as indicated through field measurements of radon and radon daughter levels performed by the Mining Enforcement and Safety Administration. A particular concern is during the summer months when visitor use is the greatest and the interchange of interior air with exterior air is at a minimum. This situation would result in the working level exposure being maximized. Since there are a number of State and privately owned caves and "cave air" conditioned buildings, it will be strongly recommended that further investigation in this area be undertaken.

During a three or four hour underground visit, the individual exposure probably would not be large; however, a significant population person-Rem dose per year, may result from the approximately one million persons visiting caves operated by the U. S. National Park Service each year. At the request of the National Park Service EPA made interim recommendations on exposure limits for persons employed in the Carlsbad Caverns. On June 3, 1976, EPA recommended a 4 WLM annual limit for workers in these caverns as an interim recommendation and requested public comment on the general applicability of these recommendations to other caves and caverns open to the public (41 F.R. 22409).

EPA further recommended that measures be implemented to keep exposures below the 4 WLM annual limit where feasible. No limit was set for visitors to the caverns. EPA also stated that the individual exposure limit

of 4 WLM per year cannot be characterized as safe since the risk of lung cancer would expect to double after 10 to 20 years of employment; therefore, it might be advisable to rotate long term employees working in elevated radon areas.[1]

This Task Force recommends that a more thorough evaluation be performed on this source of possible radiation exposure in State and privately owned caves. If control methods are found to be necessary, ventilation has been suggested as a corrective measure, but in such a way as not to eventually destroy the cave features and ecology that persons came to view. It is recommended that representatives of this Task Force, EPA, NIOSH, and MESA meet to discuss the preliminary investigations and measurements and determine what other studies should be undertaken to properly evaluate this situation. Additionally, the interim recommendations made on the basis of existing Federal guidance for the protection of underground uranium miners should be evaluated as additional information and data are obtained.

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STANDARDS AND GUIDELINES FOR RADIOACTIVE MATERIAL IN CONSUMER AND CONSTRUCTION PRODUCTS

The NRC and some of the States currently have regulations controlling the use of uranium mill tailings for construction and other purposes. The question immediately arises as to the need for similar controls on other industrial products or byproducts that contain radioactivity, such as slag, gypsum, and other materials from phosphate plants, or tailings from other types of mining and milling operations. Idaho has proposed a regulation prohibiting the use of slag under or within habitable structures, but this same regulation authorizes the use of slag outdoors, i.e., for road construction, railroad ballast, etc. (This regulation appears as Attachment A in this report.) Controls on the use of other radioactive materials will be difficult in the future without standards and guidelines for radioactivity concentrations. For example, what concentration of radium-226 in slag is acceptable for the use of slag under or within houses, and what is the maximum permissible concentrations of radium-226 in gypsum that could be tolerated for use in wallboard? Also, what level of radioactivity is permissible in materials used for road construction, general fill, and other uses?

There is an obvious need for answers or at least guidelines pertaining to questions like those above. It is therefore recommended that the EPA develop standards and guidelines for the use, distribution, and disposal of industrial products and byproducts containing naturally-occurring radioactive materials.

RADIOACTIVITY IN CONSTRUCTION MATERIALS

It is estimated that the average person in the United States spends 95% of his time indoors. Over 78% of this time is spent in the home.[1] Selected members of the population, such as the very young, the elderly, and the chronically ill, spend an even higher percentage of their time indoors. Yet, knowledge of the radiation exposure which occupants within buildings receive as a result of naturally-occurring radionuclides present in construction materials is far from complete. This situation exists in spite of the fact that estimates show that the radiation exposure to the population from natural background (including the dose equivalent from radionuclides in building materials) is greater than that from any other source.

Radionuclide Content of Building Materials

The literature review by Eadie notes the paucity of significant published data relating to radionuclide concentrations in specific building materials.[2] No comprehensive surveys of the radionuclide content of construction materials in the United States are available at the present time. However, analyses of radionuclide concentrations of commonly used building materials have been conducted in the United Kingdom, the Soviet Union, the Federal Republic of Germany, the German Democratic Republic, Taiwan, Sweden, and Hungary. These studies indicate

*This Section contains excerpts from the following report sponsored by the EPA: Moeller, D.W. and Underhill, D.W., "Final Report on the Study of the Effects of Building Materials on Population Dose Equivalents", Harvard School of Public Health, December, 1976.

that large variation in the radionuclide content exists among different types of building materials. Generally, wood products, natural plaster, metals, and cement display the lowest concentrations. Highest concentrations are observed in materials such as granite, pumice stone, and clay brick and in some byproducts of industrial processes such as artifical gypsum and concrete composed of coal-fired power station fly ash.

The most comprehensive studies published to date dealing with the natural radioactive content of building materials are those of Krisiuk, et al., using gamma spectrometic analysis, determined the radium-226, thorium-232 and potassium-30 concentrations of over 300 samples of building materials from many regions of the U.S.S.R.[3] Materials of volcanic origin including granite, tuff, and facing materials composed of tinguaite and endialite, as well as materials manufactured from industrial wastes such as boiler slag, blast furnace slag, and coal fly ash exhibited high radionuclide content. In contrast, materials comprised of natural gypsum, chalk, lime, and cement showed relatively low radionuclide content. Radionuclide concentrations in some of these materials are presented in the Table, along with estimates of the air-absorbed dose rates the materials would produce if they served as the wall, ceiling, and floor of a room of standardized dimensions. The data on radionuclide concentrations in many construction materials showed large variability from region to region.

Situations which result in the use of high radioactivity materials in structures in the U.S. include uranium mill tailings used as backfill and construction material made from byproducts of the phosphate mining and milling industry in Florida.[4,5] The radionuclides of primary concern in both of these cases are uranium and its daughter products. Another

radionuclide of probable importance in the U.S. is potassium-40. Lowder, et al., have stated that concrete blocks manufactured from Conasauga shale deposits in Eastern Tennessee, for example, may contribute to elevated exposures due to the potassium-40 content of the micaceous clay commonly associated with this type of shale.[6]

Radiation Exposure

Building materials constitute both a source of and shield from external radiation exposure. The source characteristics result from naturally-occurring radionuclides in the building materials themselves, and the shielding characteristics are determined by the degree to which natural terrestrial and cosmic radiation sources are attenuated by these materials. In general, wood frame buildings have low source qualities and are relatively poor shields for terrestrial and cosmic radiation. Masonry buildings are often significant sources but provide good shielding for terrestrial background radiation.

Radon and radon daughter concentrations inside buildings have been measured by several investigations. Yeates, et al., measured radon daughter concentrations in several frame dwellings and multi-story masonry buildings in the Boston, Massachusetts, area.[7] In single family frame dwellings, radon daughter concentrations were of the same order for outside and first floor measurements. Basement concentrations were from 4 to 23 times higher than first floor concentrations. Daughter concentrations in masonry office buildings tended to be slightly higher than first-floor concentrations in residences, but the concentrations in

offices showed little variation with building height. Home ventilation rates varied between 1 to 3 air changes per hour, while measured office rates ranged from 5 to 14 per hour.

According to Auxier the overall dose to the bronchi of people occupying homes built of uranium bearing materials (e.g. some granites, low density concretes, and gypsum boards) over a fifty year period at an average of 15 hr/day would approach that at which the incidence of lung cancer in miners is doubled. In a poorly ventilated basement laboratory, Parthaserathy found the background concentration of polonium-218 to range from 955 to 1915 pCi/l.[8]

It appears that the average contribution from radionuclides in building materials to the external dose equivalent rate to occupants in brick and masonry houses is about 10 to 20 mRem/yr. For some population groups, values range up to 100 mRem/year or more. Dose equivalent rates to the lungs may be even higher,and it would appear that control measures should be considered. Such measures include (1) material substitution, (2) improved manufacturing standards, (3) changes in basic building designs, (4) application of surface sealants, and (5) increased ventilation accompanied by processes for the adsorption and/or filtration of airborne radionuclides. Economic analyses show that several of these measures appear to be justified if one applies the cost-effectiveness guidelines proposed for nuclear power plants by the U.S. Nuclear Regulatory Commission of $1000 per person-Rem.

RADIOACTIVE CONTENT OF BUILDING MATERIALS*

Type of building material	Country	Number of samples	Average activity concentration (pCi g^{-1})			Air dose rate** (μrad h^{-1})
			^{40}K	^{226}Ra	^{232}Th	
Bricks, clinker	Fed. Rep. of Germany	-	17	2.2	2.6	25
Clay bricks	United Kingdom	23	17	1.4	1.2	16
Red bricks	Soviet Union	55	20	1.5	1.0	16
Bricks	Sweden	109	-	-	-	25
Heavy concrete	Soviet Union	87	15	0.9	0.8	12
Light concrete	Soviet Union	16	14	2.0	0.9	15
Concrete	United Kingdom	5	11	2.0	0.8	15
Concrete without alum shale	Sweden	23	-	-	-	14
Concrete containing alum shale	Sweden	29	-	-	-	130
Cement	Fed. Rep. of Germany	-	4	1.4	1.4	12
Cement	Sweden	10	-	-	-	9
Cement	Soviet Union	7	6	1.2	1.2	8
Slagstone and sand	Fed. Rep. of Germany	-	9	2.2	2.8	24
Natural sand and sand rejects	Soviet Union	32	7.1	0.65	0.5	7
Natural plaster	United Kingdom	69	3	0.6	0.2	4
Natural plaster	Fed. Rep. of Germany	-	2.4	0.5	0.5	5
Plaster	Soviet Union	1	10	6.25	0.17	5
Chemical plaster	Fed. Rep. of Germany	-	2	14	0.5	50
Chemical plaster	United Kingdom	6	2	21	0.5	70
Granite	Soviet Union	2	40	3	4.5	46
Granite bricks	United Kingdom	7	28	2.4	2.3	28
Tuff	Soviet Union	13	18	2.6	2.0	24
Pumice stone	Fed. Rep. of Germany	-	24	2.2	2.3	26
Slag pumice	Soviet Union	6	1.7	5.5	1.5	21
Rock aggregate	United Kingdom	3	22	1.1	0.1	12
Lime	Sweden	4	-	-	-	2
Phosphorus slags	Soviet Union	15	4	6.1	0.6	24
Facing materials	Soviet Union	35	39	1.9	2.3	30

*Beninson, et al., 1975

**The absorbed dose rates in air have been calculated assuming a 1 m geometry and an infinite thickness

Radioactive Phosphate Slag in Outdoor Construction

Phosphate slag, a calcium fluorosilicate byproduct of the thermal phosphate industry, has been commonly used throughout Southeastern Idaho for many years. Its rock-like qualities make it an ideal substitute for common gravel in many outdoor construction projects, i.e., highway sidecasting, asphalt and concrete aggregate, road fill, stabilization material in feedlots, and as railroad ballast. Since 1971, all asphalt-coated surfaces in one Idaho city have included the use of phosphate slag, i.e., all city streets, all parking lots, elementary school playgrounds, etc.

Radioassay of phosphate slag shows a radium-226 content of 35 pCi/gram. This may be contrasted to the radium-226 assay of common gravel at 0.40 pCi/gram. The direct gamma exposure rate measured at 3 feet over the surfaces of city streets, parking lots, playgrounds, etc. built with phosphate slag was 15 to 40 μR/hour. Measurements made over similar surfaces constructed with common gravel showed gamma levels of between 6 and 10 μR/hour.

It is anticipated that the continued use of phosphate slag within a city's boundaries will eventually result in a human population living within a network of streets measuring two to seven times above the natural gamma radiation background. Although not defined, the increased radon-222 concentrations in the city's environment will pose an additional radiation exposure to the population. Additionally, upon removal of worn-out asphalt surfaces from the city's streets, there is no perpetual assurance that the radioactive slag will not be used as a fill material under habitable structures.

The problems indicate a thorough evaluation of the environmental consequences of continuing to use phosphate slag in outdoor construction is warranted.

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RELATED EPA—TASK FORCE ACTIVITIES

The United States Environmental Protection Agency is presently conducting several programs to determine the need for radiation protection standards, guidelines, and criteria with respect to exposures from naturally-occurring radionuclides. The following problem areas have been identified for initial efforts because of their public health importance:

1) The development of recommendations to the State of Florida for the control of radiation exposures associated with phosphate materials. These recommendations will include acceptable indoor radiation level guidelines and criteria for evaluating undeveloped land to preclude radiation problems in homes that might be constructed on the land.

2) Land contamination problems exist in several States in addition to Florida, because of uranium mine and milling and other mineral processing activities. Therefore, the Agency is evaluating the need to develop national guidelines for acceptable land use of areas containing elevated concentrations of naturally-occurring radioactive materials.

3) Construction materials such as phosphate slag, pumice, fly ash, and phosphogypsum can contribute significant radiation exposure to occupants of structures made from these materials. To provide adequate public health protection from these

materials, the Agency is assessing the need to develop national guidelines for acceptable radiation concentrations in construction materials used in structures.

4) These same construction materials are frequently used for road pavement, railroad ballast, backfill, and other applications. While the public health significance of these uses are probably not as great as when used in structures, it is not clear that such practices are prudent and in the overall public interest. The State of Idaho has requested the Agency to provide guidance for appropriate radiation protection criteria for these applications. In conjunction with the State of Idaho's request, and because such problems are of national interest, the Agency is considering national radiation protection recommendations for such applications of naturally-occurring radionuclides.

All of the problems noted above are of significant importance to one or more States. Further, existing legislation provides many States with some authority to control such exposures. Therefore, it is recommended that this Task Force, on behalf of the Conference, work closely with the Environmental Protection Agency in their efforts to develop the criteria and guidelines described above. The Task Force would interact with EPA in the following manner:

1) Review and evaluate any technical support documents developed by the Agency.

2) Evaluate the practicality and overall quality of standards, criteria, and guidelines developed by the Agency including assessing their compatibility with existing State regulations.

3) Provide EPA with input in the development of criteria and guidelines including any data and information available from the States.

4) Any other coordination that its determined to be appropriate.

STATUTORY AUTHORITY OF STATES TO REGULATE NATURALLY—OCCURRING RADIOACTIVIE MATERIALS

The Task Force recommends that all States adopt uniform regulations pertaining to the control of radioactive mineral tailings and industrial byproduct piles. Louisiana, for example, has adopted such a regulation (See Attachment D). These regulations should not be simply limited to "uranium mill tailings", because it is clear that other types of radioactive piles must also be controlled.

The States, in some instances, should investigate their statutory authority to control naturally-occurring radioactive material. There is the belief in some States that Agreement State radiation control agencies have authority only over byproduct or agreement material, and not over naturally-occurring radioactive materials. Consequently, there may be a need for additional statutory authority to control naturally-occurring radioactive materials.

The obvious method of achieving the needed control program would be through specific radioactive materials licensing.

Specific licenses could be issued for the naturally-occurring radioactive materials in the uranium decay series. The mining, beneficiating, processing, and crushing operations could be licensed to possess, use, and store this radioactive material. Specific licensing could provide for:

(1) Restricted use of industrial byproducts such as slag.

(2) Long-term control of slag and gypsum piles.

(3) Periodic radiation monitoring in order to continuously define and assess radiological problems.

(4) Imposition of radiation regulations and standards.

Safe uses of slag and other radioactive industrial byproducts could be provided for by a license. A license could:

(1) Restrict the use of these materials to road construction, other outdoor uses, and other uses authorized by the regulatory agency.

(2) Prohibit the use of these materials for any purpose that would result in or would likely result in slag being under, incorporated into, or within dwellings.

Examples of specific licenses that could be issued to phosphate industries are given in Attachment B of this report. Attachment C is a licensing guide for a phosphate industry application for a radioactive material license.

Other industries with significant radiological aspects such as zirconium recovery plants and rare earth mills and processing plants should also be licensed in order to insure adequate controls over the naturally-occurring radioactive material. The State of Oregon presently licenses a zirconium plant and requires the plant to transfer all radioactive tailings containing thorium to the waste disposal site near Hanford, Washington.

RECOMMENDATIONS

A. Pending the establishment of appropriate standards, the use of phosphate mining and milling byproducts in occupied structures should be discouraged if they contain elevated levels of radium-226. EPA should place high priority on evaluating this application of these byproducts and balancing this against the environmental problems associated with waste storage of the materials.

B. Phosphate byproducts, particular gypsum, are presently used to a limited extent as an agricultural soil conditioner. Although little is specifically known regarding the environmental impact of this use, and considering the small addition of radioactivity to the environment, it is unlikely that this application would result in significant radioactivity uptake in crops. It is not recommended that this use of the byproduct materials be discouraged at this time. Additional controlled studies should be performed to document the anticipated minimal environmental impact of this use

C. Slag from thermal process phosphorus acid plants should not be used for any purpose that results in its being under or within an enclosed structure. Any other use of slag should be thoroughly evaluated.

D. States, where applicable, should adopt regulations pertaining to control of radioactive mineral tailments and industrial byproduct piles. States should investigate their statutory authority over naturally-occuring radioactive material and if necessary, enact appropriate legislation.

E. The States should perform additional radiological air monitoring inside and outside phosphate plant areas.

F. States should inventory mineral mining and processing operations referred to in Part VI of this report, and examine potential radiological aspects of their effluent, emissions, products and wastes. A report of the inventory and other findings should be submitted to Task Force #7 with any pertinent recommendations.

G. This Task Force as a representative of the Conference, should meet with EPA, NIOSH, OSHA and MESA and discuss their respective jurisdictions and activities regarding occupational radiation exposure in the phosphate industry.

H. EPA should study the uptake of naturally-occurring radionuclides in food crops from irrigation water, fertilizer, and radiation-bearing soil.

I. EPA should study the whole body and organ doses resulting from ingestion of radon daughters in drinking water.

J. EPA should evaluate the inhalation hazard which may result from the release of radon from water containing high concentrations of naturally-occurring radioactive material during certain home and commercial uses.

K. EPA should develop national criteria and guidelines concerning the use and distribution of industrial byproducts containing naturally-occurring radioactive material. The EPA should consider seeking appropriate authority to promulgate such guidelines as national standards.

L. EPA and appropriate States should study and evaluate all uses of fly-ash from coal-fired plants, particularly related to construction materials.

M. The radiological aspects of each proposed coal-fired plant should be evaluated by the appropriate agencies. Radioactive releases should be estimated from an analysis of the coal to be consumed and the operating characteristics of the proposed plant. This recommendation is especially pertinent to any proposed plant that intends to derive its fuel from low-grade, uraniferous Western coals.

N. The EPA should develop standard procedures and guidelines for sample collection and analysis of naturally-occurring radioactive material to ensure the acquisition of uniform and comparable data by State and Federal programs. The EPA should expand their cross-check quality assurance program in conjunction with the standard analytical procedures and to include NORM.

O. The States, the EPA, or other appropriate agencies should proceed to measure radon levels on fly ash piles and in structures built in fly ash disposal areas, if such case exists.

P. It is recommended that representatives of this Task Force, EPA, NIOSH, and MESA meet to discuss the preliminary investigations and measurements of radon-222 in caves and determine what other studies should be undertaken to properly evaluate this situation. Additionally, the interim recommendations made on the basis of existing Federal guidance for the protection of underground uranium miners should be evaluated as additional information and data are obtained.

Q. This Task Force recommends that a thorough evaluation be undertaken concerning the occurrence of radon-222 as a source of possible radiation exposure in State and privately-owned caves.

R. In view of the measured high concentration of radium-226 in oil brines, the impact of uncontrolled discharge of these brines on the biosphere should be evaluated by appropriate State and Federal agencies. The proposed study in Attachment E to investigate this impact should be considered for funding by EPA or other appropriate agencies.

S. Phosphate industries and all other industries having significant radiological impacts should be licensed in order to assure adequate controls over naturally-occurring radioactive materials.

ATTACHMENT A

Explanation of the Proposed Revision
of Part I
Idaho Radiation Control Regulations

It is generally known that when radium-bearing material is placed under houses or other structures, radioactive radon gas emanates from the radium and may diffuse into the structures causing internal radiation exposures to occupants.

The Radiation Control Section has established that slag from thermal phosphate plants is radioactive and contains radium as well as other radioactive materials.

The Radiation Control Section has determined that radon concentrations in houses constructed with slag as aggregate in concrete can exceed the Surgeon General's recommended radon concentration guidelines.

Since slag is radioactive, the provisions of Part I, "Radiation Safety Requirements for Radioactive Mineral Mill Tailings", Idaho Radiation Control Regulations, can be applied. Section I.2(f) prohibits the removal of tailings material from the slag pile without specific written approval of the Agency.

Slag has been used for a number of years for various purposes, including as aggregate in asphalt, as railroad ballast, as aggregate in drain fields, as stabilization material in cattle yards, and as aggregate in concrete. It is the purpose of these proposed revisions to Part I to (a) provide for the limited use of slag where no hazard has been established, e.g. road construction and (b) to prohibit the use of slag where a hazard has been established, i.e., under or within habitable structures.

Sec. I.2 Maintenance of Piles and Ponds at All Mills.

(f) Except as provided in Sec. I.5 of this Part and for reprocessing at the site, prior written approval of the Radiation Control Agency must be obtained before any tailings material is removed from any active or inactive mill site or tailings pile.

Sec. I.5 General License.

(a) A general license is hereby issued to transfer, receive, acquire, own, possess, and use licensable concentrations of naturally-occurring radioactive material in slag from thermal process phosphate plants provided that:

(1) A written notice containing the following statement or substantially similar statement shall accompany each transfer of slag:

NOTICE - SLAG FROM THERMAL PROCESS PHOSPHATE PLANTS CONTAINS TRACE QUANTITIES OF RADIUM, A RADIOACTIVE MATERIAL. SLAG IS NOT SUITABLE FOR USE UNDER OR WITHIN HABITABLE STRUCTURES. THE IDAHO RADIATION CONTROL REGULATIONS PROHIBIT THE USE OF SLAG AS FILL OR AS AGGREGATE IN CONCRETE THAT WILL BE UNDER OR WITHIN HABITABLE STRUCTURES.

(2) Slag containing licensable concentrations of naturally-occurring radioactive material shall be used for only the following purposes:

(i) For road construction, either as aggregate in asphalt or concrete, as fill, or as sidecasting.

(ii) For railroad grades as ballast.

(iii) For parking lots, driveways, sidewalks, bridges, or other outdoor structures, as aggregate in asphalt or concrete.

(iv) For stock yards as fill or as stabilization material.

(v) For other purposes specifically authorized in writing by the Radiation Control Agency.

(b) It shall be specifically prohibited to use slag containing licensable concentrations of naturally-occurring radioactive material for the following purposes:

(1) As aggregate in concrete or other material that will be under or within habitable structures.

(2) For any purpose that will result or is likely to result in slag being under, incorporated into, or within habitable structures.

(3) For any other purpose except as authorized in Sec. I.5(a)(2).

(c) Persons who transfer, receive or acquire slag from thermal process phosphate plants pursuant to the general license contained in Section I.5(a) shall be exempt from the requirements of Part C of these regulations.

Sec. I.20 Definitions. As used in this part:

(a) "Habitable structure" means any dwelling, house, garage, building or other enclosed structure that is likely to be occupied by an individual.

(b) "Licensable concentrations of naturally-occurring radioactive material" means concentrations of the uranium or thorium series radioisotopes greater than those concentrations listed in Part B, Schedule A, Exempt Concentrations, of these regulations.

(c) "Mill" means any ore processing plant, a thermal or wet phosphate processing plant, or any other processing or manufacturing plant.

(d) "Slag" means that tailings material of the thermal process phosphate plants.

(e) "Tailings material" means any residue separated in the preparation of various products.

RADIOACTIVE MATERIAL LICENSE

ATTACHMENT B

LICENSEE	LICENSE NUMBER	EXPIRATION DATE
Elemental Phosphorus Plant	AMENDMENT NUMBER ☐ INITIAL LICENSE	Previous Amendments Are Void
	THIS LICENSE ISSUED PURSUANT TO AND IN ACCORDANCE WITH ☐ Application ☐ Letter ☐ Telegram ☐	SIGNED BY: DATE:

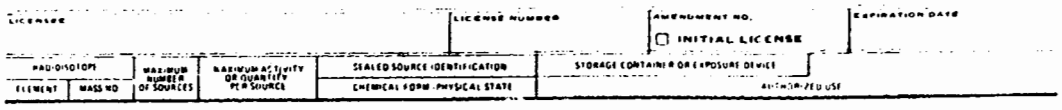
RADIOISOTOPE ELEMENT	RADIOISOTOPE MASS NO	MAXIMUM NUMBER OF SOURCES	MAXIMUM ACTIVITY OR QUANTITY PER SOURCE	SEALED SOURCE IDENTIFICATION CHEMICAL FORM—PHYSICAL STATE	STORAGE CONTAINER OR EXPOSURE DEVICE	AUTHORIZED USE
Naturally occurring radioactive materials in the uranium and thorium series.			As necessary for mining, processing, and recovery of elemental phosphorus.	Phosphorus bearing shale or ore containing licensable concentrations of radioactive materials.		Phosphorus bearing shale or ore containing licensable concentrations of radioactive material may be mined, beneficiated, and processed for the purpose of extracting elemental phosphorus.
Naturally occurring radioactive materials in the uranium and thorium series.			As necessary for mining, processing, and recovery of elemental phosphorus.	Industrial by-products containing licensable concentrations of radioactive materials.		Unless otherwise provided by the Conditions of this license, industrial by-products containing licensable concentrations of radioactive materials shall be for storage only.

1. All industrial by-products containing licensable concentrations of radioactive materials shall be stored at the licensee's phosphate plant near _____.

2. Radioactive furnace slag may be transferred to slag crushing operations which hold a specific license issued by the State of _____ or another State which authorizes the receipt and use of such material. Crushed slag shall be used only for (specify uses).

3. The licensee shall maintain records of all furnace slag transfers which indicate the amount of slag transferred to each slag crushing operation. Records shall be made available for inspection by the Radiation Control Agency.

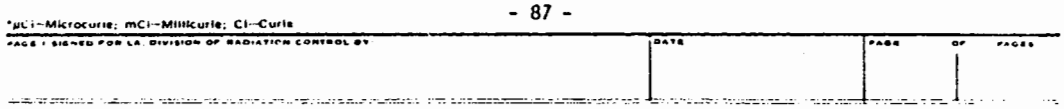
4. FEP may be transferred or used by the licensee and is exempt from the provisions of this license and the _____ Radiation Control Regulations.



5. The licensee shall comply with the provisions of Part _____ of the _____ Radiation Control Regulations, as they apply to furnace slag piles and other industrial by-product piles containing licensable concentrations of radioactive materials.

6. The license shall comply with statements, representations and procedures contained in his application dated _____ and signed by _____.

*μCi-Microcurie; mCi-Millicurie; Ci-Curie



RADIOACTIVE MATERIAL LICENSE

LICENSEE

Wet Process Plant

LICENSE NUMBER | EXPIRATION DATE

AMENDMENT NUMBER | Previous Amendments Are Void

[ ] INITIAL LICENSE

THIS LICENSE ISSUED PURSUANT TO AND IN ACCORDANCE WITH

[ ] Application [ ] Letter [ ] Telegram [ ] .....

SIGNED BY: | DATE:

RADIOISOTOPE ELEMENT	RADIOISOTOPE MASS NO	MAXIMUM NUMBER OF SOURCES	MAXIMUM ACTIVITY OR QUANTITY PER SOURCE	SEALED SOURCE IDENTIFICATION CHEMICAL FORM - PHYSICAL STATE	STORAGE CONTAINER OR EXPOSURE DEVICE	AUTHORIZED USE
Naturally occurring radioactive materials in the uranium and thorium series.				As necessary for mining, processing, and production of phosphoric acid and fertilizer.	Phosphorus bearing shale or ore containing licensable concentrations of radioactive materials.	Phosphorus bearing shale or ore containing licensable concentrations of radioactive materials may be mined, beneficiated, and processed for the purpose of producing phosphoric acid and fertilizer.
Naturally occurring radioactive materials in the uranium and thorium series.				As necessary for mining, processing, and production of phosphoric acid and fertilizer.	Industrial by-products containing licensable concentrations of radioactive materials.	Unless otherwise provided by the Conditions of this license, industrial by-products containing licensable concentrations of radioactive materials shall be for storage only.

1. All industrial by-products containing licensable concentrations of radioactive materials shall be stored at the licensee's chemical plant near _____.

2. The licensee shall comply with the provisions of Part ____ of the _____ Radiation Control Regulations, as they apply to the gypsum piles and other industrial by-product piles containing licensable concentrations of radioactive materials.

3. Phosphoric acid and fertilizers produced by the licensee are exempt from the provisions of this license and the _____ Radiation Control Regulations.

4. The licensee shall comply with statements, representations and procedures contained in his application dated _____ and signed by _____.

LOUISIANA DIVISION OF RADIATION CONTROL | DATE | PAGE ____ OF ____ PAGES

RADIOACTIVE MATERIAL LICENSE

LICENSEE	LICENSE NUMBER	EXPIRATION DATE
Slag Crushing Company	AMENDMENT NUMBER ☐ INITIAL LICENSE	Previous Amendments Are Void
	THIS LICENSE ISSUED PURSUANT TO AND IN ACCORDANCE WITH ☐ Application ☐ Letter ☐ Telegram ☐ ... SIGNED BY: DATE:	

RADIOISOTOPE ELEMENT	RADIOISOTOPE MASS NO	MAXIMUM NUMBER OF SOURCES	MAXIMUM ACTIVITY* OR QUANTITY PER SOURCE	SEALED SOURCE IDENTIFICATION CHEMICAL FORM--PHYSICAL STATE	STORAGE CONTAINER OR EXPOSURE DEVICE AUTHORIZED USE
Naturally occurring radioactive materials in the uranium and thorium series.		As necessary for the commercial uses of slag authorized by this license.		Furnace slag containing licensable concentrations of radioactive materials from phosphate industries utilizing a thermal process.	Unless as otherwise provided by the conditions of this license, slag containing licensable concentrations of radioactive material shall be for hauling, crushing and storing only.

1. Slag containing licensable concentrations of radioactive material shall be crushed and stored at the licensee's facility _____ near _____.

2. Slag containing licensable concentrations of radioactive material may be used by the licensee _____ for purposes of _____ and in any other State where such use is not prohibited.

3. The licensee shall maintain records of all uses and all transfers of slag containing licensable concentrations of radioactive materials which indicate the specific use, location, and amount of slag used or transferred. Records shall be made available for inspection by the Radiation Control Agency.

4. The licensee shall comply with statements, representations and procedures contained in his application dated _____ and signed by _____.

LOUISIANA DIVISION OF RADIATION CONTROL	DATE	PAGE	OF

ATTACHMENT C

INSTRUCTIONS
Application for Radioactive Materials License

1. (a) Name and Street Address of Applicant. Give name and address of company.

   (b) Street Address(es) at Which Radioactive Material will be Used. Must include all locations where ore, products, or industrial byproducts containing licensable concentration of radioactive material are mined, processed, stored, or distributed from. (Also, list names and locations of all underground mines.

2. Department to Use Radioactive Material. Self explanatory if applicable.

3. Self explanatory.

4. Individual User(s). Not applicable.

5. Radiation Protection Officer. Name of person designated as the individual responsible for insuring compliance with all license conditions and all applicable radiation regulations.

6. (a) Radioactive Material. Specify naturally-occurring radioactive material in the uranium and thorium decay series.

   (b) Chemical and/or Physical Form and Maximum Quantity of Each Chemical and/or Physical Form That You Will Posess at Any One Time. For the form of material specify: (1) phosphorus bearing shale or ore containing licensable concentrations of radioactive material and/or (2) industrial byproducts containing licensable concentrations of radioactive material including slag, gypsum, fluid dust material, etc. (make list complete) and/or (3) industrial products containing licensable concentration of radioactive material including phosphoric acid, fertilizer, elemental phosphorus, FEP, etc. (make list complete). Radioactive wastes (airborne or liquid effluents) should not be included in ...

   For maximum quantity simply specify: (1) As necessary for mining, processing, and manufacture of elemental phosphorus, phosphoric acid, fertilizer, etc. and/or (2) As necessary for the commercial ... as authorized by this license.

7. Describe Purpose For Which Radioactive Material Will Be Used. Specify all uses of ore, products, and industrial byproducts containing licensable concentrations of radioactive material that you want to be authorized for, including but not limited to such things as: (1) mining, (2) beneficiating,

(3) processing, (4) transfer to slag crushing operations, (5) transfer to vanadium extraction companies, (6) transfer to asphalt companies for road construction, (7) transfer to railroads for railroad grades, (8) all other transfers (specify ultimate uses), (9) storage, (10) all other uses.

8. Type of Training. Applies only to the Radiation Protection Officer. Individual should have training that will simply enable him to understand license requirements and applicable radiation regulations.

9. Experience with Radiation. Applies only to the Radiation Protection Officer.

10. Radiation Detection Instruments. List of instruments should include all survey meters, air samplers, and laboratory equipment necessary to measure radiation levels (including airborne radioactivity) in order to determine and insure compliance with license conditions and applicable radiation regulations. (Do not include personnel monitors). Or alternatively, describe facilities or consultant firms to be used or employed for radiation measurement purposes.

11. Method, Frequency, and Standards Used In Calibrating Instruments Listed Above. Self explanatory.

12. Film Badges, Dosimeters, and Bio-assay Procedures Used. The requirements for personnel monitoring are specified in Section ____ of the _____ Radiation Control Regulations. (See also, Section _____)

13. Facilities and Equipment. Self explanatory.

14. Radiation Protection Program. See Sections C.1, C.101, C.102, C.103, C.104, C.105, C.106, C.201, C.205 of the _____ Radiation Control Regulations.

15. Waste Disposal. Must include a description of all airborne and liquid radioactive effluents, if any. See Section _____ of the _____ Radiation Control Regulations.

ATTACHMENT D

PART __

RADIATION SAFETY REQUIREMENTS FOR RADIOACTIVE MINERAL TAILINGS AND INDUSTRIAL BY-PRODUCT PILES

Sec. __.1 SCOPE. The regulations in this part establish requirements for radioactive mineral tailings, piles and ponds and by-product piles containing radioactive material from industrial processes in concentrations in excess of $1x10^{-7}$ microcurie per gram[1]. As used in this part "by-product" means any material produced, other than the primary product, in an industrial process. The provisions of this part are in addition to, and not in substitution for, other applicable provisions of: (a) these regulations and (b) any specific license issued pursuant to Sec. C.30 of Part C of these regulations.

Sec. __.2 SPECIFIC REQUIREMENTS FOR TAILINGS, PILES AND PONDS. Unless specifically provided otherwise by the Division, the following requirements for tailing, pile and pond areas shall be fulfilled:

(a) Access to such areas shall be controlled and posted as specified by the Division.

(b) These areas shall be maintained in such a manner that excessive erosion of, or environmental hazards from, radioactive materials [illegible].

(1) Pile edges adjacent to a river, bayou, creek or other water course shall be stabilized to prevent erosion.

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[1] uCi/ml for liquids

(2) Drainage ditches sufficient to prevent erosion from surface runoff water shall be provided.

(c) Prior written approval of the Division shall be obtained before the surface area of the land shall be put to use.

(d) With the exception of reprocessing at the site, approval by the Division must be obtained prior to removal of any material from these areas.

Sec. __.3 SALE OR TRANSFER OF THE SITE. The Division shall be given written notice thirty (30) days in advance of any contemplated transfer of right, title or interest in the site by deed, lease or other conveyance. The written notice shall contain the name and address of the proposed purchaser or transferee.

Sec. __.4 ABANDONMENT OF THE SITE. Prior to abandonment of the site, the requirements of this section shall be fulfilled.

(a) Piles shall be stabilized against wind and water erosion and contoured in a manner which will prevent collection of water.

(b) In addition to the above requirements, any material which has been removed from the pile by natural forces shall be returned to the pile.

(c) Ponds shall be drained and covered with materials that prevent blowing of dust. Water drained from the ponds shall be disposed of in a manner approved by the Division.

(d) Detailed plans for compliance with paragraphs __.4(a), (b) and (c) shall be submitted to the Division, for review and approval.

Sec. __.5 WAIVER. Upon application to the Division, certain requirements of this part may be waived or modified if it can be shown that the requirements are unnecessary or impractical in specific cases.

ATTACHMENT E

INTRODUCTION

The occurrence of environmentally high concentrations of radium isotopes in oil field production waters (also called oil field brines, produced water, produced waste-water, or formation water) is well documented (Kuroda, 1953; Gott and Hill, 1953; Armbrust and Kuroda, 1956). Armbrust and Kuroda (1956) reported Ra-224, Ra-226, and Ra-228 in production waters from oil fields in Oklahoma and Arkansas, with activities ranging from 100-1100 pCi/l Ra-224, 1-1600 pCi/l Ra-226, and up to 400 pCi/l Ra-228. They also found 8 pCi/l Th-228 and 0.5 pCi/l Th-227 in one well sample. Gott and Hill (1953) reported environmentally high concentrations of radium in precipitates collected from the bottom of oil-water separators, and from ditches and ponds used for disposal of the production water.

The Mississippi, Louisiana, and Texas Gulf Coasts either are, or have the potential to become major oil-producing areas of the United States. Texas and Louisiana have numerous producing wells both on- and off-shore. During the period November 1972 to October 1973, approximately 1.7×10^{10} liters of production water were discharged to the Gulf of Mexico from operations on Federal Outer Continental Shelf (OCS) leases outside the three-mile limit (data furnished by U. S. Geological Survey, 1974). Records of discharges inside the three-mile limit and on-shore are maintained by the states in which the discharges occurred. Data provided by the Department of Conservation, State of Louisiana, show that for the year 1975 a state total of 8.1×10^{10} liters of formation waters were discharged into the surface environment, 17.5% off-shore (presumably inside the three-mile limit), and 82.5% or 6.7×10^{10} liters on-shore. Of the latter, 6.0×10^{10} liters were discharged into non-potable water bodies, 4.3×10^{9} liters were discharged into streams and rivers, and 2.4×10^{9} liters were disposed of in open holding pits from which gradual loss occurs via evaporation and seepage into the underlying ground. Similar data should be available from the State of Texas upon request. In 1974, several samples of formation waters from the Gulf Coast production region were obtained and analyzed for Ra-226. The results are presented in Table 1.

The data in Table 1 show that environmentally high levels of Ra-226 are common in production waters from the Gulf Coast oil fields. For comparative purposes, it is noted that average open ocean surface waters contain about 0.05 pCi/liter; coastal waters probably do not generally get much higher than about 1 pCi/liter, except in very restricted environments; drinking water standards restrict the permissible Ra-226 content to less than 5 pCi/liter; and agreement-state and NRC regulations governing the operations of licensees permit no more than 30 pCi/liter in liquid discharges to unrestricted access areas. According to Louisiana state officials, production waters do not come under these regulations at the present time, but it is notable that they contain up to 400 pCi/liter, or 10 times the permissible regulated inputs.

To our knowledge, there have been no scientific studies of the input levels, speciation (in terms of dissolved vs. particulate forms) or ultimate disposition (fate) of such naturally occurring radium being discharged into

local estuarine environments (marshes, streams, rivers). The occurrence of environmentally high levels of radium in production waters does not appear to be a widely known fact, as evidenced by conversations with both state officials and oil company officials, although such discussions have not been exhaustive.

THE LEEVILLE OIL FIELD

The Leeville Oil Field is a producing field covering about 62 km^2 (24 mi^2) in Lafourche Parish, Louisiana, centered near 29°14'N, 90°12'W (see Fig. 1). This field was discovered in 1928, and is operated by Texaco, Inc.

By Texaco's estimates (in a letter to Dr. T. Whelan dated 10/13/76), production is expected beyond the year 2000. The Leeville Oil Field is in and surrounded by a salt marsh complex typical of southern Louisiana. The annual average salinity of the marsh water is 13.3 °/₀₀ (Whelan et al., 1976), and the hydrology is seasonally controlled. There are three primary hydrologic conditions: in the winter, the winds are from the north and push the marsh water toward the south, resulting in seasonally low water levels in the marsh, and biologically, the marsh is least active; in the spring, a "spring tide" or "flood tide" reverses the prevailing condition, and seasonally deep water levels occur; and during the summer, the hydrologic conditions are most stable, with only small diurnal tides affecting the medium water level by a few tens of centimeters, and the marsh is biologically quite active. The predominant vegetation and primary producer is *Spartina alterniflora*, a marsh grass. There are also abundant mussel and clam banks, and commercially harvested oyster beds; benthic organisms, primarily amphipods and crabs, abound; pelagic biota include grass shrimp, the Gulf Killifish (*F. grandis*), and the Sheepshead Minnow (*C. variegatus*).

Figure 2 is a portion of a topographic map of the area. On it are plotted the Leeville Oil Field tank batteries which are reported to be discharging production waters into the surrounding surface environment. These production waters may properly be classified as brines since their average annual "salinity" does not vary much from 150 °/₀₀ (T. Whelan, personal communication). Although this field has been producing for several decades, records of volumes of production water discharges have only been maintained in recent years. Table 2 summarizes the five-year (1971-1975) production water discharge histories for the 11 tank batteries reported by Texaco to the Department of Conservation, State of Louisiana.

The total five-year discharge was 6.3×10^9 liters; the 1975 volume discharge of 1.03×10^9 liters was ~1.6% of the total on-shore discharge in the State of Louisiana. In October 1976, brine samples from the discharges at TB #4, in mid-field, and TB #8, in the northern, less built-up area of the field, were sampled and analyzed for Ra-226 in our laboratory. The results are presented in Table 3, and indicate that both tank batteries are discharging water with environmentally high concentrations of Ra-226.

Based on the data in Tables 2 and 3, in 1975, TB #4 discharged approximately 0.2 Curies Ra-226, and TB #8 discharged approximately 0.016 Curies Ra-226

into the local salt marsh complex. If it is assumed that the average production water from the Leeville Oil Field contains about 280 pCi/l Ra-226, then over the five-year period of record, up to 1.76 Curies Ra-226 were added to the marsh around the field. At this time, we have no data by which to predict the ultimate fate of that radium. We also do not know if any other isotopes from the uranium-thorium series are present in environmentally significant concentrations in the production waters.

We are prepared to design and implement a program which will answer some of these questions. We anticipate that such a program would involve collection and analyses of production water (brine) samples, marsh-water samples, and various biological samples from the study site. The number of each type of sample will have to be determined by the environmental conditions at the study site, but may be estimated as up to 75 each. The resulting data will be compiled, interpreted, and discussed in a final report, to be delivered with up to six (6) copies to the sponsor approximately 16-20 months after project initiation. It is also expected that one or more professional papers to be published in reputable scientific journals will result, and these will acknowledge the sponsoring agency.

REFERENCES

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TABLE 1

Ra-226 IN SELECTED SAMPLES OF FORMATION WATERS FROM

THE GULF COAST PRODUCTION REGION

IDENTIFICATION	TREATMENT	REFRACTIVE SALINITY (ppt)	Ra-226 (pCi/l)
ARCO, High Island Platform B, ~ 12 miles offshore Galveston, TX	Rough filtered, filtrate acidified	121	313 ± 4
EXXON, Grand Isle Terminal, Grand Isle, LA	No treatment	99	143 ± 3
EXXON, No location data provided, shipped from Lafayette, LA	No treatment Acidified, but not filtered	98	291 ± 3 298 ± 2
EXXON, Pelican Island Term., Pelican Island, TX	No treatment Filtered, Acid. Unfiltered, Acid.	100	22 ± 1 16 ± 5 46 ± 2
TEXACO, Bay de Chene, Jefferson & Lafourche Parishes, LA	Unfiltered, Acid. Filtered, Acid.	128	335 ± 10 327 ± 5
TEXACO, Garden Island Bay, Plaquemines Parish, LA	Unfiltered, Acid. Filtered, Acid.	110	397 ± 8 393 ± 6
TEXACO, No location, originate Houma, LA	Unfiltered, Acid. Filtered, Acid.	-	276 ± 3 131 ± 2

NOTE: Errors indicate precision (1σ) of replicate analyses; overall technique error is ±7%.

TABLE 2

LEEVILLE SALT WATER DISPOSED OF

IN NON-POTABLE WATER

	TB #1	TB #4	TB #5	TB #6	TB #7	TB #8	TB #9	TB #10	TB #11	TB #12	TB Bge	TOTAL
1971	4.64×10^7	7.71×10^8	1.91×10^8	5.80×10^6	3.19×10^8	1.51×10^8	-	5.80×10^7	3.48×10^7	4.64×10^6	5.80×10^6	1.59×10^9
1972	4.64×10^7	7.71×10^8	1.91×10^8	5.80×10^6	3.19×10^8	1.51×10^8	5.80×10^6	5.80×10^7	1.76×10^7	4.64×10^6	5.80×10^6	1.59×10^9
1973	3.55×10^7	6.93×10^8	1.63×10^8	2.58×10^7	3.19×10^8	8.46×10^7	1.47×10^6	6.29×10^7	-	4.98×10^6	4.64×10^6	1.39×10^9
1974	4.38×10^7	4.59×10^8	4.59×10^7	4.88×10^7	1.11×10^8	8.03×10^7	2.86×10^6	5.74×10^7	-	8.61×10^6	1.15×10^7	8.74×10^8
1975	5.47×10^7	5.89×10^8	8.36×10^7	2.04×10^7	1.44×10^8	6.46×10^7	-	5.90×10^7	-	1.08×10^7	7.73×10^6	1.03×10^9

TABLE 3

Ra-226 IN BRINE FROM TB #4 AND TB #8, LEEVILLE OIL FIELD

TB #4	No Treatment	327 ± 20 pCi/l
	Acidified	319 ± 9 pCi/l
	Filtered and Acidified	318 ± 5 pCi/l
TB #8	Acidified	260 ± 8 pCi/l
	Filtered and Acidified	248 ± 5 pCi/l

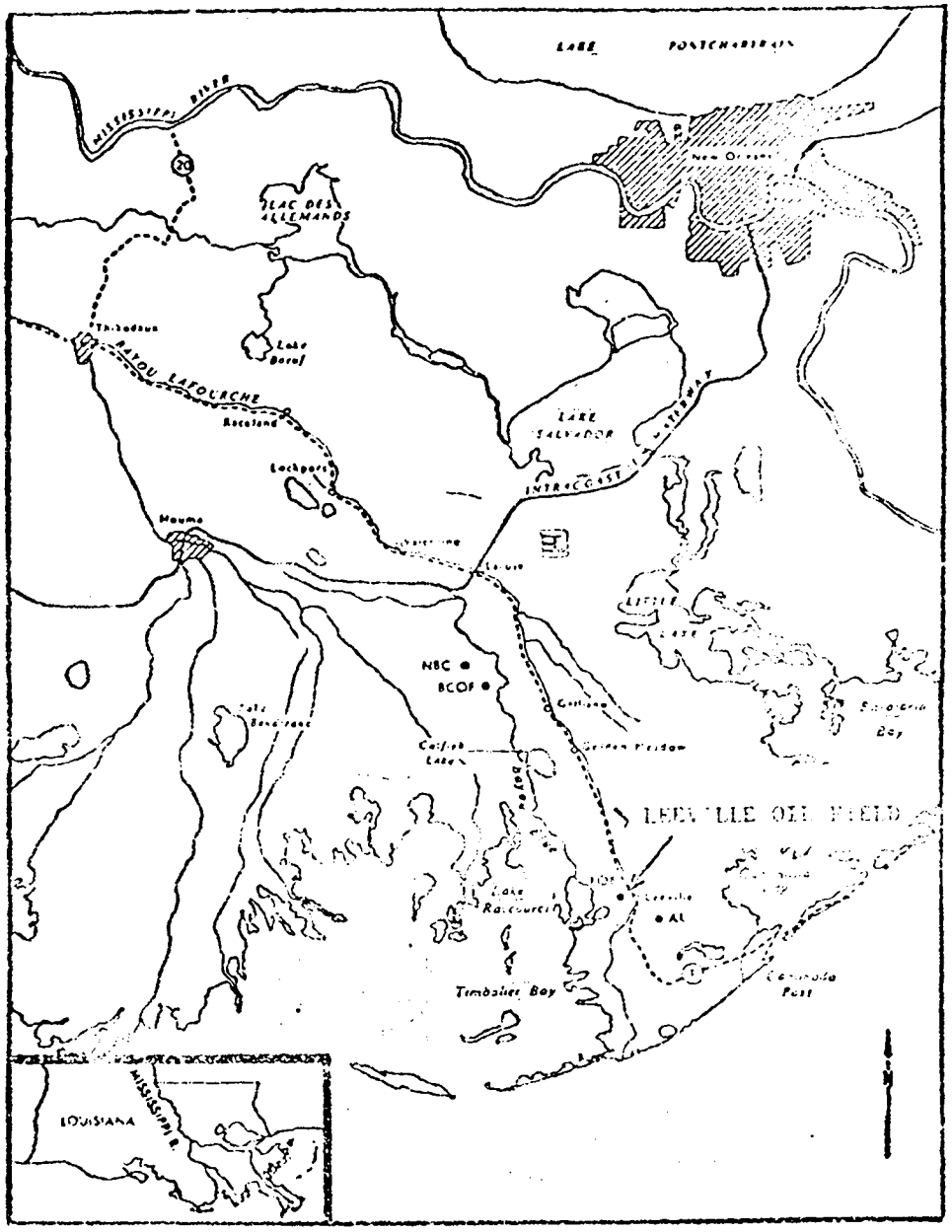

FIGURE 1: Location of the proposed study site

*U.S. GOVERNMENT PRINTING OFFICE:

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