PLUTONIUM :

STATEMENT OF THE PROBLEM

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Office of Radiation Programs
U.S. Environmental Protection Agency

I. Introduction

The transuranium elements include plutonium and the higher atomic number elements. Many of the nuclides of these elements are characterized by long radioactive half-lives and high radiotoxicity. Unlike most chemical pollutants, (except perhaps for the heavy metals and certain non-biodegradable toxic compounds) the transuranium elements tend to build up in the environment by virtue of their long persistence. Therefore, any release of these radionuclides must be considered as an irreversible commitment to the environment and entails a cumulative potential health hazard for many future generations.

Plutonium is a metallic, radioactive element with atomic number 94. It was the first man-made element to be produced in relatively large quantities. In the Periodic Chart, a general classification of the elements by similarity of properties, the transuranium elements are included in the actinide series, which starts with actinium and extends through lawrencium (Fig. I-1).

Elements 93 through 103 are synthetically produced by nuclear reactions, usually starting with uranium. The addition of a neutron to uranium-238 (mass number 238) forms uranium-239 which by radioactive decay forms neptunium-239 (Np-239). It decays to plutonium-239 (Pu-239). Subsequent neutron captures followed by radioactive decay leads to the formation of other elements. Figure I-2 is a diagram of the production scheme for important transuranium elements, starting with uranium-235 and 238.

The nuclear properties of the important transuranium elements produced are listed in the following table. It is notable that where short half-lives or beta (β) emissions occur the daughter product is a long lived alpha emitting radionuclide.

Essentially all transuranium alpha emitting radionuclides are considered to be extremely radiotoxic if inhaled or ingested. Plutonium has been studied more carefully than the other transuranium elements because it is being handled in large quantities in the manufacture of nuclear weapons. However, other transuranium elements such as neptunium, americium, and curium will also be present in appreciable quantities in spent reactor fuel.

Present plutonium levels in the environment have come primarily from two sources - worldwide fallout from atmospheric tests of nuclear weapons, and releases from various facilities and sites where plutonium is, or has been, used. Fallout over the area of the United States represents a large total existing inventory of the order of 10-15,000 curies, and is relatively uniformly distributed with current deposition levels ranging from 1-4 millicuries per

PERIODIC CHART OF THE ELEMENTS

1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	13 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57-71 La* Series	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89-103 Act Series	(101)	(105)	(106)	(107)	(108)										

*Lanthanide Series	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
†Actinide Series	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102	(103) Lw

The transuranium elements (in shaded squares) are part of the actinide series of elements which as a group occupy a single square, at actinium (number 89) in the main figure. Plutonium, element 94, is in this series. The rare earth (lanthanide) series of elements, also shown in a horizontal row, also occupies a single square (at lanthanum, element 57) on the main chart.

Figure I-1

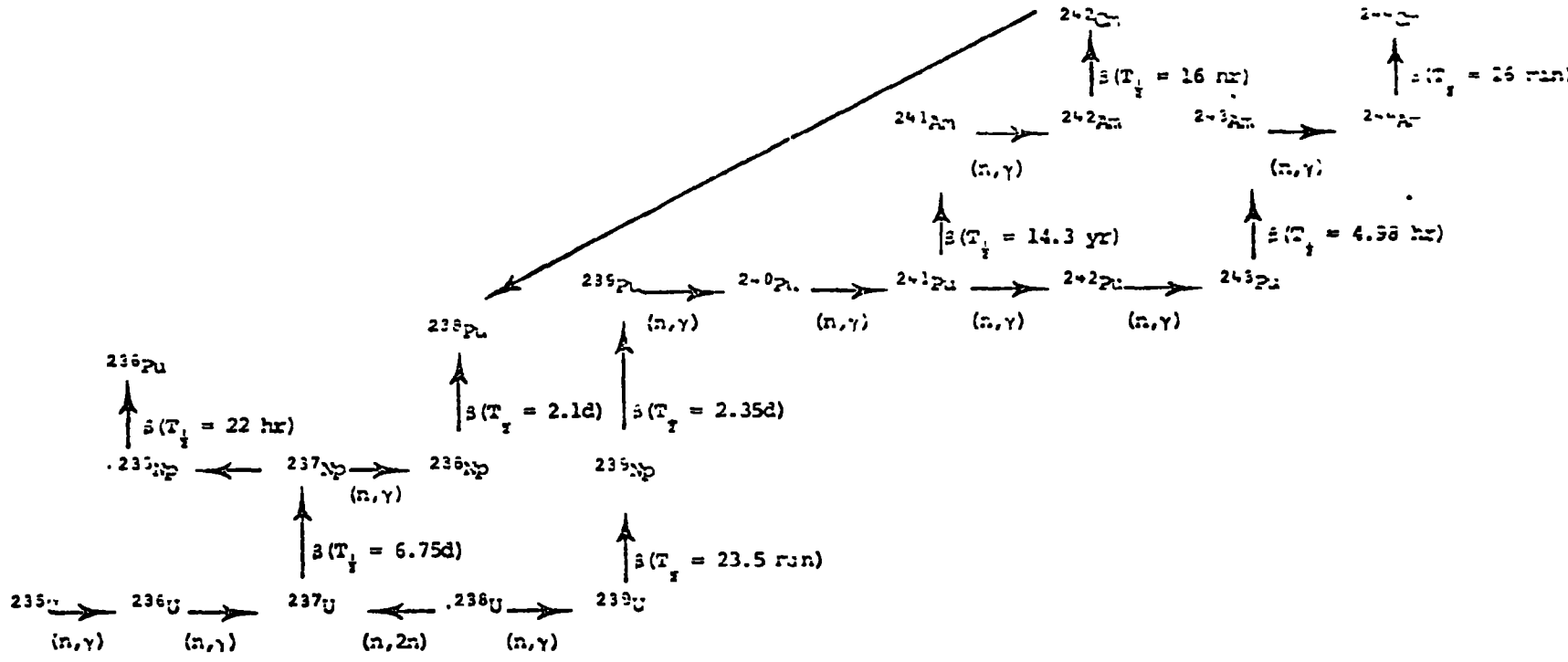

Figure I-2 Formation scheme for important transuranium elements

Table I-1

*Radionuclide	Particle Emitted on Decay	Half-life		Daughter
Np-236	β	22	hr	Pu-236
Np-237	α	2.14×10^{6}	y	Pa-233
Np-238	β	2.1	d	Pu-238
Np-239	β	2.35	d	Pu-239
Pu-236	α	2.85	y	U-232
Pu-238	α	87.4	y	U-234
Pu-239	α	2.44×10^{4}	y	U-235
Pu-240	α	6.6×10^{3}	y	U-236
Pu-241	β	14.3	y	Am-241
Pu-242	α	3.87×10^{5}	y	U-238
Pu-243	β	4.98	hr	Am-243
Am-241	α	433	y	Np-237
Am-243	α	7.37×10^{3}	y	Np-239
Am-244	β	26	m	Cm-244
Cm-242	α	163	d	Pu-238
Cm-244	α	18.1	y	Pu-240

*Np-neptunium Pu-plutonium Am-americium
Cm-curium U-uranium Pa-protactinium

square kilometer. Releases from weapons-related facilities and production sites represent a local addition to the general background level.

Indeterminate but large quantities of these elements have been produced for the nuclear weapons program and additional very large quantities (of the order of many millions of curies) will be produced by the nuclear power program. Releases from these activities (even at the most pessimistic levels) will not equal or exceed for several decades the quantities already in the environment from fallout, but rather could present substantial localized problems of levels tens or hundreds of times the existing background levels.

In addition, space nuclear power devices may contain from ten to over a hundred kilocuries of plutonium, and a total of more than a half million curies has been used in space missions. Commercial applications, such as the nuclear powered heart pacemaker, generally utilize relatively small quantities, but are rapidly proliferating. Because of the potential for long-term environmental contamination from all these sources and others, it would seem appropriate to analyze the problem from an overview perspective at this time. It must be recognized that control measures must be instituted at a time sufficiently far in advance of when the cumulative effect of all potential releases of these radionuclides to the general environment could become a significant public health problem.

The principal problem associated with standard setting activities for plutonium would appear to be that of achieving a proper perspective for all the material already committed to the environment and that which is likely to follow. As noted above, most of the current worldwide inventory of plutonium in the environment is due to atmospheric weapons tests. This is reasonably uniformly distributed, predominantly in the Northern Hemisphere, and less than 10% remains suspended in the atmosphere. Additional current releases, resulting from weapons fabrication operations, testing of various devices, and other operations contribute primarily excess <u>local</u> concentrations. Future additions, if confined to a very small fraction of the total inventory, would essentially continue this situation.

The statutory authority of the EPA for the development of radiation protection standards is derived from the authorities tranferred on its inception. Reorganization Plan No. 3 of 1970 tranferred the functions of the Atomic Energy Commission to the Environmental Protection Agency ". . . to the extent that such functions of the Commission consist of establishing generally applicable environmental standards for the protection of the general environment from radioactive material. As used herein, standards

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mean limits on radiation exposures or levels, or concentrations or quantities of radioactive material, in the general environment outside the boundaries of locations under the control of persons possessing or using radioactive material." As a result of this transfer, Section 161(b) of the Atomic Energy Act provides that the Administrator may, within the above framework, "establish by rule, regulation, or order, such standards to govern the use of special nuclear material, source material, and by-product material as (he) may deem necessary or desirable to . . . protect health or to minimize changes of life or property."

Section 274(h) provides that "The (Federal Radiation) Council shall advise the President with respect to radiation matters, directly or indirectly affecting health, including guidance for all Federal agencies in the formulation of radiation standards and in the establishment and execution of programs of cooperation with States."

II. Hazard Assessment

The physiological effects of alpha radiation are of short range and essentially limited to only the immediate vicinity of the radioactive emitter. Therefore, radioisotopes which emit primarily alpha radiation represent a hazard to health only if they are brought into intimate and prolonged contact with human tissue. Plutonium may enter the body by inhalation, ingestion, or through open wounds. Retention and/or transmigration tend to concentrate sites of potential damage to the lung and associated lymph nodes, the skeleton, liver, and soft tissue, as well as to the tissue at the site of accidental entry.

Evidence to date suggests that the onset of evident radiation induced damage directly attributable to exposure to plutonium apparently has a very long induction period. Animals exposed to plutonium aerosols or injected with soluble plutonium compounds have developed carcinomas at times ranging from months to years and approximately related to the total absorbed dose. No human fatalities due to plutonium exposure have yet been reported, although records of exposure date back to 1946. A comprehensive survey of such occupationally exposed individuals is being maintained by the National Transplutonium Registry, with continuing medical surveillance and autopsy after death.

For purposes of standard setting, where a large population-at-risk is involved, the numerical estimates of adverse health effects due to exposure can be developed on a statistical basis. The health hazard from a radiation dose has generally been assessed by using a linear, non-threshold theory to relate accumulated dose to effect, where the known values at high exposure levels are extrapolated linearly to zero. While there is no scientific information to definitely confirm this hypothesis, in light of current uncertainties this should be considered as a prudent and probably conservative assumption. Thus, it is assumed that every dose received, no matter how small, carries with it some risk of an adverse health effect.

A substantial body of information already exists on the long-term effects of exposure to various types of radiation. Considerable additional work is in progress on the specific bioeffects associated with continued exposure to the transuranium elements. Because of limited number of human exposures and apparent long induction time, definitive answers to all aspects of plutonium carcinogenic effects will not be available for a number or years. In the meantime, standards and guidelines will have to be based on the best available current information and revised as appropriate.

In order to determine the impact of releases of the transuranium elements, numerical estimates of potential health effects must be correlated with environmental concentrations of the elements. This involves the conversion of these concentrations to organ burdens, organ burdens to doses and doses to health effects. The models given for determining organ burdens are general in nature but specific comments refer to plutonium since it is the element for which the most information is available. Dose conversion factors are given for several elements. Health effects estimates are based on exposure to alpha radiation and are not necessarily applicable to all situations. Each model carries with it a set of assumptions which introduce uncertainties in its applications. As more information becomes available these models will undoubtedly be refined and more closely represent the actual situation and permit more accurate prediction.

The respiratory tract is the most common mode of entry into the body. The particles inhaled may be either soluble or insoluble in body fluids. Soluble compounds tend to move to other areas of the body and can concentrate in the liver or bone. Insoluble compounds tend to remain in the respiratory tract for long periods of time. For modeling purposes 1000 days is used for the clearance half-time from the lung.

The estimates of risk for inhaled radioactive particles are based on animal experiments and on human data from two sources. The first is from total organ or whole body exposure to x-rays or gamma rays, where every cell in the lung receives approximately an identical radiation exposure. This includes data on Japanese survivors from Hiroshima and Nagasaki, and on patients who have been exposed to extensive diagnostic or therapeutic x-irradiation of the chest and lungs. The second source of data is derived from persons exposed to the special conditions found in some mines. These persons inhaled not only radioactive particles but also rock dust, radon and daughter radioisotopes, diesel engine exhaust and other materials. The radon daughter isotopes are thought to be the causes of lung cancer in this case, and the radiation exposure of cells in various regions of the lung is different. Data is available for uranium miners, flurospar miners and others. From these data an estimate of the risk of lung cancer per rem of radiation has been derived.

In the case of plutonium-239, in solid tissue the dose rate at 40 μm from the surface of a particle is about 1.5 to 2.0% of the dose rate of the source point and by 45 μm distance the dose rate is virtually zero. In the lung, which is about 80% air, the corresponding distances are about 320 μm and 480 μm respectively. Depending on the number of particles inhaled, the fraction of the

lung actually irradiated may differ but usually only a small portion of the lung will be exposed to the radiation.

The use of animal data which may provide additional support to hypothesis concerning the relative efficacy of particulate vs uniform exposure is conflicting and has many limitations. Studies involving inhalation of radioactive materials have shown increases in lung cancers in experimental animals. However, these increases have not been in the type of cancer expected in humans exposed to a potent carcinogen. Observed species differences have further complicated interpretation. These experiments have involved plutonium as well as other elements.

Plutonium microspheres have also been injected into hamsters and single highly radioactive spheres have been implanted in other animals. The animals have shown little response in either case.

Experiments using uniform alpha radiation of pure radon have not been any more conclusive. From results of several studies it is suggested that "uniform" alpha irradiation of lung tissue may not be effective for inducing lung neoplasia.

The anatomy, respiratory physiology, histology, and pathology is different to a greater or lesser extent in each species of animal studied. Because of these differences and the fact that lung tumors developing in animals exposed to radiation are not the same as those seen in man, it is difficult to assess the consequences of inhaled radioactive particles in man. There is no data on human exposure from which a correlation with animal inhalation experiments can be made. Until an adequate animal model is developed there will be great uncertainty and disagreement about the relationship of animal experiments to possible human exposure and risk.

Considerable uncertainty persists in the evaluation of the biological hazard of inhaled radioactive particles. Although it is possible to make estimates for disposition of inhaled particles in the lung, and estimates of the risk of adverse effects following exposure of the lung to ionizing radiation, it has proven difficult to obtain agreement on the risk associated with the inhalation of radioactive particles.

The derivation of risk estimates has been discussed above. The question of how to apply these risk estimates is the area of most disagreement. Traditionally, dose estimates for internal emitters (radioisotopes contained within the body) have been based on the average organ dose, i.e., the total amount of radiation energy deposited in an organ divided by the mass of that organ. While this is a reasonable method for estimating the risk from x- or gamma radiation where all parts of the organ receive the same amount of

energy, it has been questioned whether it is adequate for estimating the risk from the short range alpha or low energy beta radiation in cases where the radioactive material is not uniformly distributed.

The basic question posed by these considerations is simply whether exposure of a few cells in an organ to high levels of radiation constitutes a greater, similar, or lesser risk than exposure of many or all cells in that organ to a lower level of radiation. This is often designated as the "Hot Particle Problem."

III. Technology Assessment

Commercial Nuclear Power

Immediate future uses of plutonium and other transuranium elements in the power industry rely on the recovery of plutonium from uranium fueled light water reactors (LWR). As the number of commercial light water reactors increase, the plutonium produced will require storage or use (recycle) in the light water reactors. The proposed Liquid Metal Fast Breeder Reactor (LMFBR) requires plutonium as fuel; substantial quantities of plutonium will be discharged from light water reactors before the fast breeder reactors are commercially available. The estimates of plutonium production used here are those reported by the USAEC. Figure III-1 is a plot of the estimated cumulative inventory of plutonium recovered from domestic nuclear power fuels without commercial plutonium recycle in light water reactor fuels. Long term projections of plutonium recovery and use including the future LMFBR is not included in the above figure. A projection which includes plutonium recovered from both LWRs and LMFBRs is shown in Figure III-2. In addition, the quantities of plutonium available for recycle are included in the projections. From these figures the projected quantitites of plutonium that have been handled would be 200 metric tons by 1985 and 4000 metric tons by 2000. It was announced in 1972 that the first commercial recycle of plutonium in light water reactors was anticipated in 1974, when Big Rock Point was expected to load plutonium fuel in one third of its core.

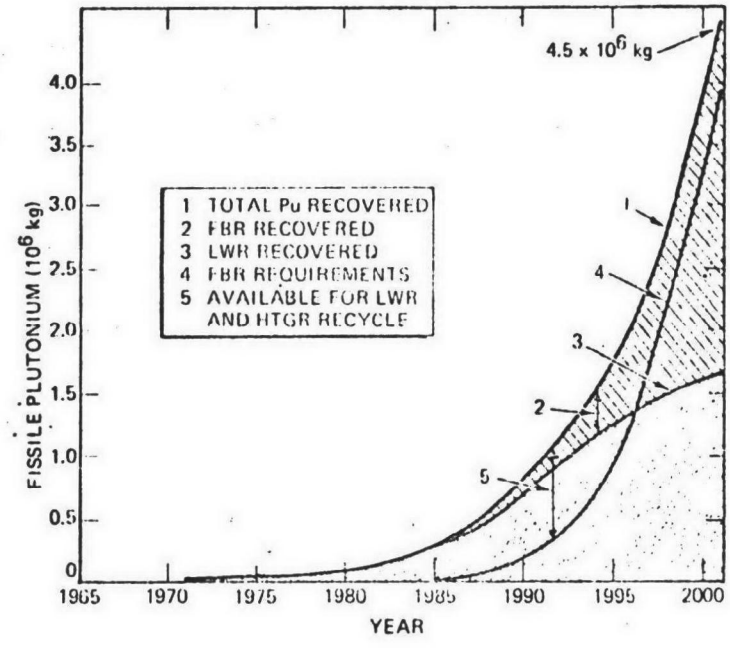
The major facilities involved in plutonium handling are the fuel reprocessing plants, the fuel fabrication plants and the reactors. Materials movement between these facilities involves transportation and solid waste generation.

A. Power Reactors

1. LWRs

Operations at a typical 640 MWe boiling water reactor (BWR) indicate that essentially all plutonium and other transuranics which escape into the primary coolant collect in the solid waste system.

Liquid waste treatment systems are very effective at removing the transuranics. Some of them will, of course, find their way past waste treatment systems and get to the environment. The amounts, however, are evidently extremely small as compared to transuranium elements released from fuel reprocessing and fuel fabrication. An exception is Np-239, produced by n, gamma reactions on U-238, which has been reported in several BWR operating reports.



Fissile plutonium—cumulative recovery and requirements

Figure III-1

Amounts to date have been small and generally less than 4×10^{-4}% of current AEC limits.

2. LMFBRs

Average plutonium inventories in a LMFBR will be less than an order of magnitude greater than those of a uranium-fueled LWR of the same size. Consequently, the quantity of transuranics, which are produced in the core by neutron activation reactions and ultimately discharged from the facility in the spent fuel, is also less than an order of magnitude greater. A LMFBR has only about a factor of two more plutonium and transuranics than a LWR fueled with recycled plutonium.

There will normally be no transuranics reaching the environment from an LMFBR, since the LMFBR is designed for much lower leakage of coolant than LWRs during normal operations. Also, sodium coolant systems have inherent and engineered removal mechanisms which effectively remove many impurities such as the transuranic elements, so that the amount of transuranics getting outside the system even in event of coolant leakage would be greatly reduced.

B. Fuel Reprocessing

1. Industry Description

The economics of the nuclear fuel cycle require the recovery of uranium and plutonium isotopes from spent reactor fuel for re-use in new fuel elements. This separation of the uranium and plutonium from irradiated fuel is carried out at fuel reprocessing plants.

Table III-1 presents the estimated concentrations of transuranic isotopes present in spent fuel. Estimates are shown for both uranium oxide and mixed uranium-plutonium oxide fuel. Calculations of the annual inventory of transuranics in reprocessed fuel for time periods up to the year 2000 are presented in Table III-2. These data were calculated using the concentrations in Table III-1 and the projections of amounts and types of fuel to be reprocessed. It was assumed that the average concentrations of transuranics in LMFBR fuel will be similar to the concentrations in plutonium recycle fuel. These data do not include fuel from high temperature gas cooled reactors HTGRs which for the Th-U fuel cycle will not add significantly to the transuranic inventory.

Figure III-3 presents estimates of the annual discharges of transuranics from fuel reprocessing plants up to the year 2000. Three curves are shown (1) the activity of all transuranic isotopes,

Table III-1

Estimated Concentrations of Transuranics in Reprocessed Fuel (a,b)

Radionuclide	Half-Life (Years)	Uranium Fuel Ci/MT	Plutonium Recycle Fuel Ci/MT
Pu-238	86	4,000	6,000
Pu-239	24,400	500	750
Pu-240	6,580	650	1,000
Pu-241	13	150,000	300,000
Pu-242	379,000	2	5
Am-241	458	750	2,000
Am-243	7,800	20	200
Cm-242	0.45	35,000	250,000
Cm-244	17.6	2,000	25,000

(a) Burnup - 33,000 MWD/MTU

(b) Cooling Time - 150 days

Table III-2

Estimated Annual Inventories of Transuranics
in Reprocessed Fuel[a,b]

Year	Fuel[c] Metric Tons	Transuranics Curies
1975	300	5.8×10^{7}
1980	2400	6.6×10^{8}
1985	5300	2.0×10^{9}
1990	8000	3.2×10^{9}
1995	10000	4.7×10^{9}
2000	19000	9.7×10^{9}

(a) Burnup - 33,000 MWD/MTU
(b) Cooling time - 150 days
(c) Does not include HGTR Fuel Reprocessing - Th-U Fuel Cycle will not contribute additional significant quantities of transuranics.

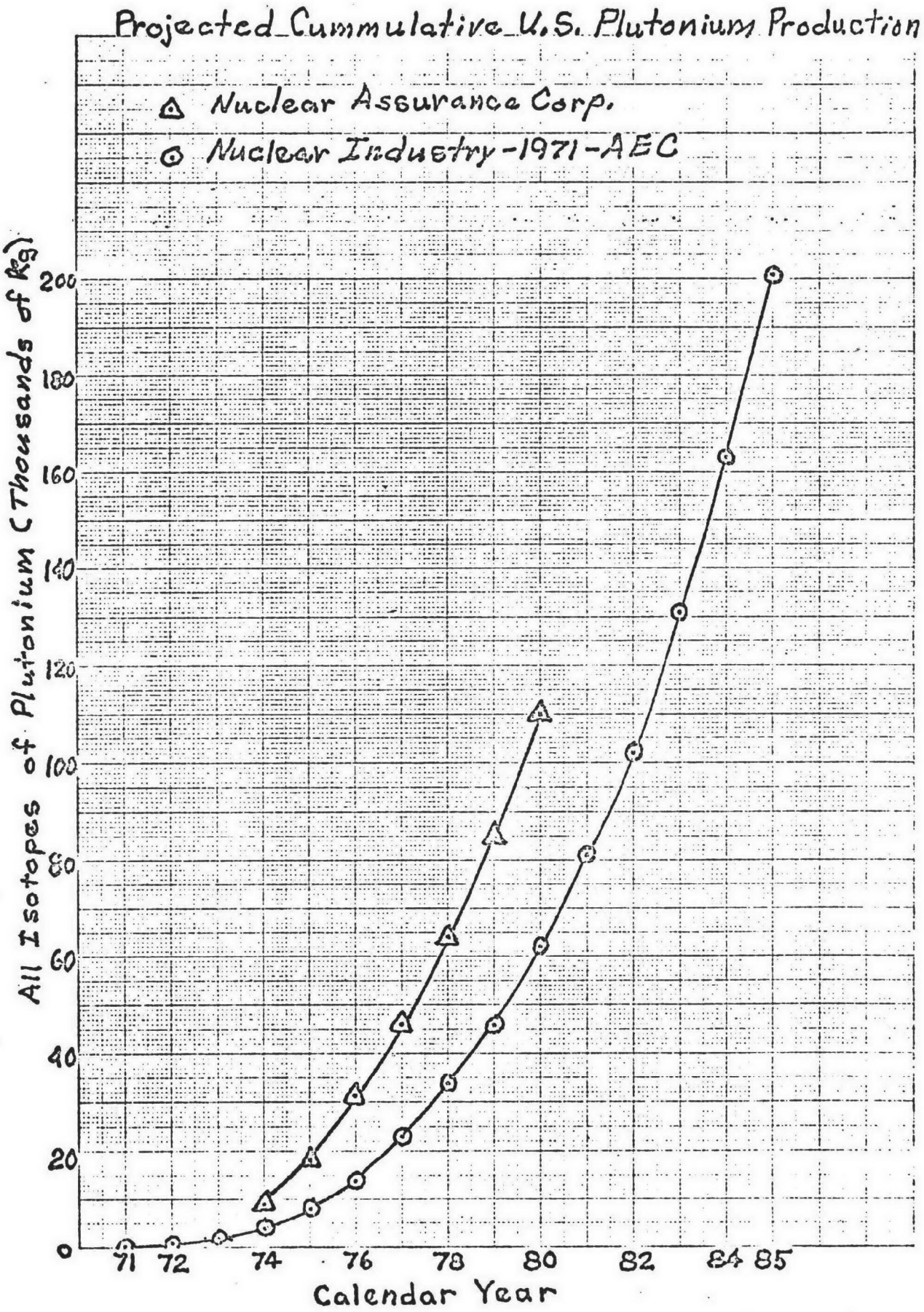

Projected Cummulative U.S. Plutonium Production
Nuclear Assurance Corp.
Nuclear Industry-1971-AEC
All Isotopes of Plutonium (Thousands of Kg)
0
20
40
60
80
100
120
140
160
180
200
71
72
74
76
78
80
82
84
85
Calendar Year

Figure III-2

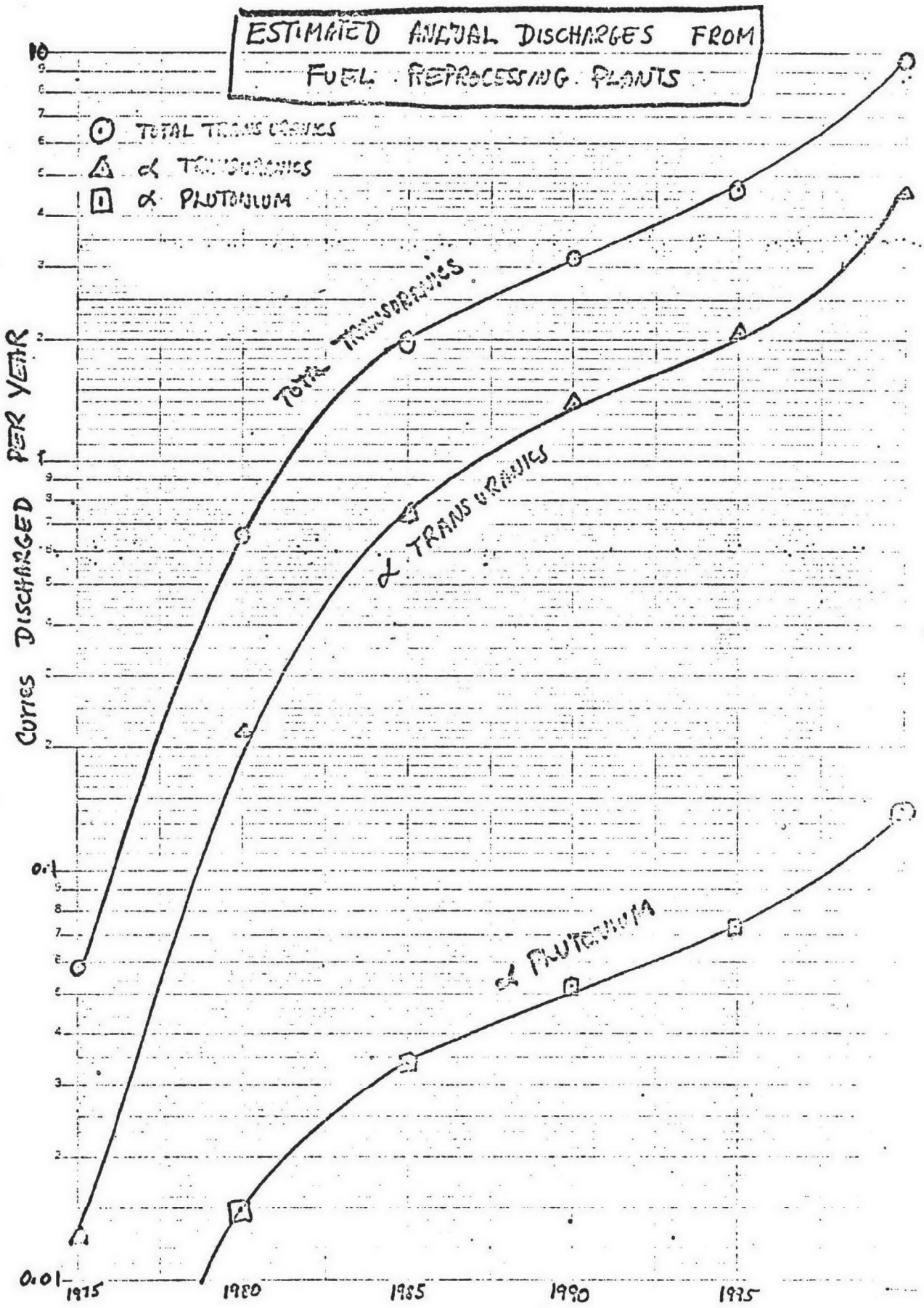

ESTIMATED ANNUAL DISCHARGES FROM
FUEL REPROCESSING PLANTS
TOTAL TRANSURANICS
α TRANSURANICS
α PLUTONIUM
CURIES DISCHARGED PER YEAR
10
1
0.1
0.01
1975
1980
1985
1990
1995

Figure III-3

(2) the activity of all alpha emitting transuranic isotopes, and (3) the activity of all alpha emitting transuranic isotopes. Figure III-4 presents similar curves for the cumulative environmental inventory of transuranics from fuel reprocessing operations.

Plutonium-241, a beta emitting isotope, makes up about 80% of the transuranic activity in the environment resulting from discharges from fuel reprocessing plants. The remaining 20% of the activity is comprised of alpha emitting isotopes. Although plutonium-241 is a beta emitter, it decays to americium-241 an alpha emitter and the resulting Am-241 will eventually represent a significant portion of the total transuranic alpha activity in the environment. Since the values for alpha emitting transuranics in Figure III-3 include the ingrowth of Am-241 from Pu-241, these values are probably the most significant in evaluating the environmental impact of discharges of transuranics.

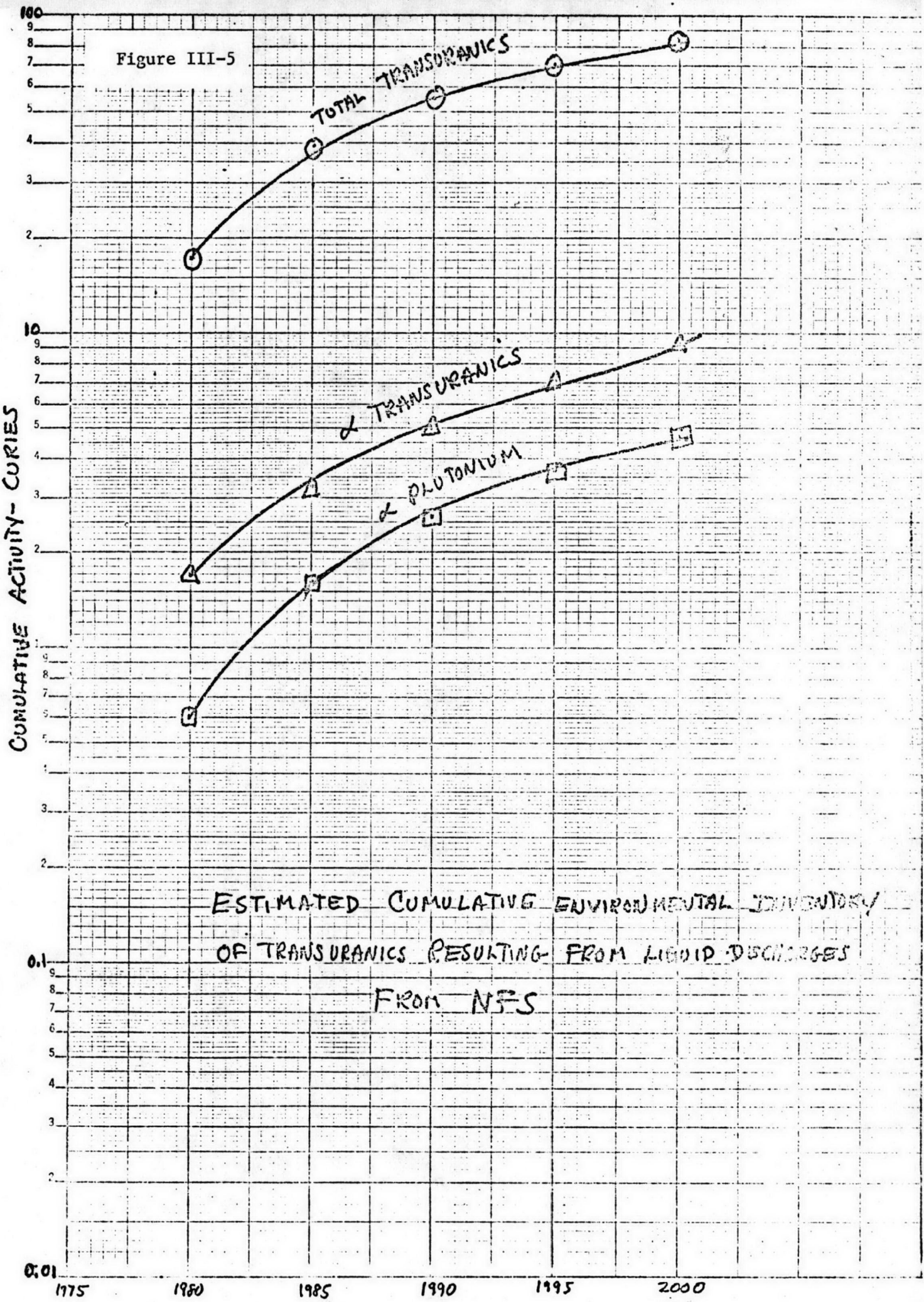
There are no commercial fuel reprocessing plants currently operating in the United States. One plant, Nuclear Fuel Services, Inc. operated for a 6 year period between 1966 and 1972 but is now shut down for plant expansion. Two other plants are presently in pre-operational stages. One is General Electric's Midwest 1 ton/day Fuel Recovery Plant (MFRP) which has postponed operations indefinitely and the other is Allied-Gulf's 5 ton/day Barnwell Nuclear Fuel Plant (BNFP) which will not be operational until 1976-1977. Future fuel reprocessing requirements indicate that about 18 plants processing about 26,000 MT/yr (metric ton per year) of fuel will be necessary by the year 2000 and that by the year 2020 about 50 plants processing 80,000 MT/yr of fuel will be required.

Figure III-5 presents estimates of the cumulative environmental inventory of transuranics which will result from the discharges of liquid waste from the Nuclear Fuel Services, Inc. plant if present waste treatment practices are continued when the plant resumes operation. These estimates are based on a release fraction of 5×10^{-8}.

2. Effluent Control

Almost all of the plutonium and other transuranics released to the environment from the operation of nuclear fuel reprocessing plants will result from the discharge of particulates which have passed through the off-gas treatment and filter systems. Only one of the three fuel reprocessing plants now under construction, NFS, will discharge radioactive liquid waste. It is not expected that any future commercial fuel reprocessing plant will discharge radioactive liquid waste.

Figure III-5



ESTIMATED CUMULATIVE ENVIRONMENTAL INVENTORY OF TRANSURANICS RESULTING FROM LIQUID DISCHARGES FROM NFS

The principal method for the control of plutonium and other transuranics at fuel processing plants is the installation of high efficiency particulate filters in the off-gas discharge lines. Usually two or more filters are used in series. It is estimated that the release fraction for particulate transuranics at fuel reprocessing plants will be less than 10^{-9}, and it is expected that future plants may attain even better particulate control than reflected by this estimate.

Recycling of condensates from low-level waste concentrators eliminates the discharge of radioactive liquid waste from MFRP and BNFP. The only process water discharged from these plants is that which is evaporated into the off gas system to dispose of tritium.

At NFS, the condensates from the low-level waste evaporators are discharged to the environment after passing through a series of holding ponds and a waste treatment facility. The release fraction for transuranics via the liquid waste is estimated to be 5×10^{-8} at NSF.

C. Fuel Fabrication

The estimated number of plutonium recycle fabrication plants of 150 metric ton (MT) capacity needed in 1980 is between 4 and 6 plants. After 1980, the amount of recovered plutonium would increase an added 3000 to 5000 kg per year. The increasing recovery would be sufficient to refuel about five additional light water reactors per year which would require one additional 150 metric ton (MT) capacity fuel fabrication plant per year. The growth in the number of plutonium recycle fuel fabrication plants would decrease or stop at the point in time when the plutonium discharged from light water reactors is required for the initial cores of the first commercial Fast Breeder Reactors. With introduction of the LMFBR in 1986 or later the fuel fabrication capacity required will begin to level off and decline. However, plutonium recycle fuel fabrication plants could change to fabrication of LMFBR fuels. Assuming the major uses of plutonium will be by light water reactors until 1985, the possible number of plants of 150 MT capacity could be 9 to 11 plants. For fabrication of LMFBR fuels the throughput of a mixed oxide plant designed for LWR fuels would be reduced by about a factor of 4 to 6 because of higher plutonium content. In an LMFBR economy the number and size of the fabrication plants is likely to increase beyond that expected for fabrication of recycle fuels of LWRs.

Aerospace Applications

Plutonium-238 oxide is used as a fuel in Radioisotope Thermoelectric Generators (RTG). Interaction of the radioactive decay particles within the fuel matrix produce heat which is converted to electricity. Plutonium fueled RTG's have been used by the National Aeronautics and Space Administration (NASA) and Department of Defense (DoD) to provide electrical power aboard satellites and on the surface of the moon. The plutonium-238 isotope for these devices is produced at the AEC Savannah River Plant in South Carolina. The isotopes are shipped to other AEC laboratories such as Mound Laboratory, Ohio or Los Alamos, New Mexico for processing into a stable fuel form and encapsulation into heat sources. After 1976, the processing into a stable fuel form is expected to be carried out at the Savannah River Plant.

Plutonium fueled RTG's have been used in space exploration in the past and present plans involve use in future space probes and satellites. Successful space missions using RTG's do not result in release of plutonium to the environment. However, launch aborts, failure to attain orbit or decay of orbit with reentry are potential events which may result in release of plutonium to the environment. In 1964, a transit satellite failed to attain orbit. Burn-up on reentry released 17,000 Ci of plutonium-238 to the atmosphere.

A launch accident resulted in two 17,000 Ci RTGs being dropped into the ocean off the coast of California. These were recovered. The failure of the Apollo 13 mission resulted in the reentry of the LEM and impact in the ocean of the experimental apparatus. Contained in the LEM was SNAP-27 generator containing 45,000 Ci of plutonium which is still in the ocean. Some future missions could result in earth orbit of a substantially greater quantity of plutonium than in previous missions. A major question about earth orbiting plutonium is the acceptability of the reentry of radioisotope powered satellites on a random basis.

Present containment design should prevent dissemination of the radionuclide under all forseeable conditions such as pre-orbital abort or uncontrolled atmospheric reentry and earth impact. The ceramic fuel matrix has been redesigned to minimize formation of particles in the respirable range in the unlikely event of burnup.

Research and development work is presently underway at Oak Ridge National Laboratory for the application of curium-244 to radioisotope thermoelectric generators.

Medical Uses

Some proposed medical uses of plutonium are research and development on the implantation of plutonium-238 powered heart pacemakers and radioisotope powered artifical heart devices. The radioisotope powered cardiac pacemaker has completed many of the tests necessary to qualify the unit for implantation in humans. Early in 1973, ARCO Nuclear Company started production of units for human implantation. Each unit contains about 5 Ci of plutonium.

The plutonium-238 powered artificial heart devices are under study at two AEC laboratories. One such study device contain 900 Ci of plutonium-238. It has been calculated that this device will subject the internal organs of the user to about 50 millirem/hour. The major potential for impact on the environment is the possible uncontrolled ultimate disposal of such pacemakers and other devices.

Military Applications

The present inventories of plutonium in the environment are primarily from military activities. Plutonium for this purpose is produced in reactors at the AEC Hanford and Savannah River reservations. Fuel slugs are reprocessed to recover the plutonium which is shipped to a weapons parts manufacturing facility. The Rocky Flats Plant in Colorado is the major weapons parts manufacturer; however, Los Alamos Scientific Laboratory and Lawrence Livermore Laboratory are involved in manufacture of special nuclear devices such as underground explosives. Assembly of the weapons parts take place at military contractor sites such as Pantex in Amarillo, Texas. From this point weapons are distributed to military facilities.

Plutonium has been released to the environment on a local level around plutonium testing and production sites. There are kCi quantities of plutonium dispersed at the Nevada Test Site and curies to tens of curies at the Trinity Test Site, Alamagordo, New Mexico. Also 3 to 5 curies have been released at the Rocky Flats plutonium processing plant near Denver, Colorado. An estimated 4 curies were leaked in 1973 from a waste storage tank at Hanford.

A continuing aspect of military applications is the possibility of plutonium dispersion in the environment due to accidents involving nuclear weapons on strategic missions. Two such accidents have occurred in Spain in 1966 and Greenland 1968.

Other military activities which involve movement of plutonium in the environment and oceans are nuclear powered submarines and ships. Each nuclear propulsion plant contains some quantity of plutonium and transuranium elements. In addition missile-launching submarines carry plutonium in the weapons. By the end of 1971, the AEC estimated there would be 95 nuclear powered submarines and 4

nuclear powered surface ships. Unknown quantities of plutonium and transuranium elements may be released during reprocessing of the nuclear fuels from these ships at the National Reactor Testing Station (NRTS) or a commercial fuel reprocessing plant. The releases during normal operation of naval reactors are not available from unclassified sources.

Consumer Products

Application of plutonium and transuranium elements in consumer products at present is limited. However, future availability could increase the number of items which could be produced for general population purchase. Manufacture of such items would require licensing action, but once in the hands of the consumer, control could be lost. Provisions for waste disposal are not now generally included with purchase of the product. Examples of such products include a smoke detector containing 40 mCi of americium-241, static eliminators at plastics plants, and snow gauges for use at remote sites.

IV. Existing Guidelines

Guidance for exposure to the transuranium elements began during World War II with the first recommendation for maximum permissible occupational levels for plutonium. In 1953 the National Committee on Radiation Protection (NCRP) recommended occupational limits for americium and curium. In 1959 and again in 1962 the International Commission on Radiological Protection (ICRP) recommended standards for an expanded list of elements which currently includes isotopes of americium, curium, berkelium, californium, einsteinium and fermium.

The first guidance for plutonium contamination came into existence almost simultaneously with the existence of the first reactor-produced plutonium. Two months after start-up of the Clinton, Tennessee reactor, in November 1943, a recommendation was made that 5 μg of fixed plutonium should be the maximum allowable body burden. This recommendation led to major changes in the plutonium handling areas to increase the protection of workers.

By the end of World War II the maximum permissible body burden was lowered to 1 μg (.06 Ci) and adopted as the standard for the nuclear weapons program generally. Tolerance values for air (3×10^{-11} Ci/cm^3) and water (3×10^{-6} Ci/cm^3) were also adopted. These standards were set before specific effects of plutonium were known, by relating plutonium to substance for which information was available. Body burden limits and maximum concentrations in water were derived by using .1 μg as a maximum permissible body burden for radium and making allowances for assumed differences between radium and plutonium. The critical organ was assumed to be bone. The tolerance limit for air was calculated by equating concentrations of plutonium with doses of gamma or x-rays to the lung.

In 1949 the Chalk River Permissible Dose Conference was held in Ontario, Canada, with the United Kingdom, United States and Canada participating. From this conference and subsequent discussions a maximum permissible body burden of .04 μCi of plutonium was adopted. The change was prompted by results of animal experiments which indicated a different plutonium toxicity, relative to radium, than had previously been assumed. New maximum permissible concentrations (MPC's) for plutonium in air (1.5×10^{-12} Ci/cm^3) and water (1.2×10^{-6} Ci/cm^3) were also adopted. A crude lung model was proposed to show the movement of plutonium from the lung to the bone and the MPC for air was based on it.

In 1950 the ICRP gave no firm recommendations for plutonium citing the lack of sufficient data. The commission did however publish essentially the same values as the final results of the Chalk River Permissible Dose Conference. This gave a maximum

permissible body burden of .04 Ci of fixed plutonium, an MPC for water of 1.5×10^{-6} Ci/cm^3 and an MPC for soluble plutonium in air of 2×10^{-12} Ci/cm^3. A value for insoluble plutonium in air was also given based on the lung as critical organ, but it was recommended that the MPC for soluble plutonium be used in all cases due to the possible transference of insoluble plutonium from the lungs to the skeleton.

In 1953 the NCRP gave new values for some of the metabolic constants used in calculating MPC's including a more detailed lung model. No changes in the recommended MPC values resulted.

In 1955 the ICRP revised its recommended MPC for water by employing the changes in metabolic constants proposed by the NCRP in 1953. The new MPC for water was 6×10^{-6} Ci/cm^3. There was no change recommended for the air MPC. Recommendations were also made at this time for MPC's based on dose to the gastro-intestinal tract.

Current MPC values were recommended by both ICRP and NCRP in 1959. In these reports the metabolic constants given in 1953, with some modifications, were used in calculating the MPC's for both air and water. The MPC for soluble plutonium in air was 6×10^{-13} Ci/cm and for water 5×10^{-5} Ci/cm^3. An MPC of 10^{-11} Ci/cm^3 was recommended for insoluble plutonium in air. MPC values were also computed based on dose to several other organs and a revision was made in the recommended MPC's based on dose to the gastro-intestinal tract in the light of new animal data.

The current recommended MPC's using bone as the critical organ for soluble plutonium and the lung for insoluble plutonium have been adopted by the AEC for use as occupational limits at nuclear facilities. To obtain suitable MPC's for the general population, the AEC has followed the recommendation of the ICRP and reduced the occupational limits by a factor of 30. Current MPC's for air are given in Table IV-1.

At this time there are no values recommended by either the ICRP or the NCRP for limits on plutonium surface contamination. This lack of guidance has caused several Federal agencies and installations to adopt individual guidelines which are geared to specific purposes. A summary of current guidelines in use is given in Table IV-2.

TABLE IV-1

MAXIMUM PERMISSIBLE CONCENTRATIONS IN AIR

FOR ^{238}Pu AND ^{239}Pu

ISOTOPE	CRITICAL ORGAN	$(MPC)_a$ RADIATION WORKERS $\mu Ci/cm^3$	$\mu Ci/cm^3$	$(MPC)_a$ GENERAL PUBLIC $\mu Ci/cm^3$	$\mu Ci/cm^3$
^{238}Pu					
Soluble	Bone	7×10^{-13}	7×10^{-7}	2.3×10^{-14}	2.3×10^{-8}
Insoluble	Lung	1×10^{-11}	1×10^{-5}	3.3×10^{-13}	3.3×10^{-7}
^{239}Pu					
Soluble	Bone	6×10^{-13}	6×10^{-7}	2×10^{-14}	2×10^{-8}
Insoluble	Lung	1×10^{-11}	1×10^{-5}	3.3×10^{-13}	3.3×10^{-7}

Table IV-2

PLUTONIUM-(GR / DPM) SURFACE CONTAMINATION LIMITS
FOR U. S. ORGANIZATIONS

ORGANIZATION	REF	[illegible]	[illegible]		
1. FRC		None			
2. AEC	1	100 dpm/100 cm^2	4.5×10^{-3}	removable activity - suitable for release to public	Source & Special Nuclear Material
3. NCRP		None			
4. DOD				no overall directive	
a. Army	2	1000 $\mu g/m^2$ 10 $\mu g/m^2$ (239 Pu)	6.17×10^{1} 6.17×10^{-1}	significant hazard level possible resuspension hazard	Nuclear Accident Contamination Control
b. Air Force	3	1000 $\mu g/m^2$ (239 Pu)	6.17×10^{1}	safe for continuous occupancy	Peacetime Nuclear Weapons Accident
c. Navy	4	<2 dpm/cm^2	9.0×10^{-3}	suitable for release to general public	Contamination Control Procedures for Special Weapon Accidents
d. DASA	5	>3500 $\mu g/m^2$ 3500-1000 <1000 (239 Pu)	$>2.15 \times 10^{2}$ --- $<6.17 \times 10^{1}$	major hazard normal hazard decontamination not necessarily required	Nuclear Emergencies
5. PHS		None			
6. Other a. NTS	6	>3500 $\mu g/m^2$ 3500-1000 1000-10	2.5×10^{2} 6.17×10^{1} 6.17×10^{-1}	extreme hazard some hazard little hazard	Test Site Operations
b. ORNL	7	30 dpm/100 cm^2 3 dpm/100 cm^2	1.3×10^{-3} 1.3×10^{-4}	direct survey - average for transferable large areas	Noncontamination zone laboratory [illegible]
c. LASL	8	<500 cpm	7.6×10^{-12}		Laboratory Operations

V. Plutonium in the Environment

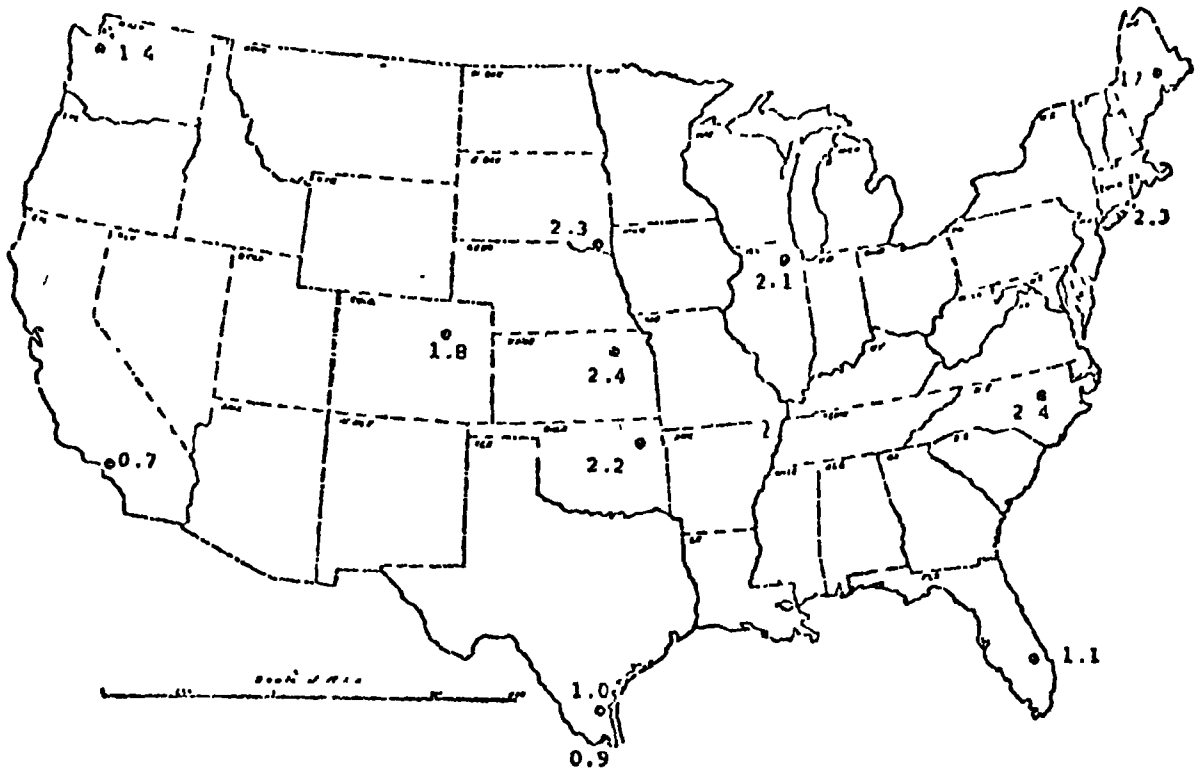
Plutonium has been released to the environment from nuclear weapons accidents and testing, spacecraft accidents, and processing and reprocessing plants. The testing of fission and fusion nuclear weapons has resulted in worldwide plutonium-239 deposition of around 300 to 500 kCi. The Health and Safety Laboratory of the AEC has measured deposition concentrations in the United States of around 1 to 2 mCi/km^2 (Fig. V-1) which results in 10 to 20 kCi being deposited in the United States.

In April 1964 a satellite, which contained a SNAP-9A (Systems for Nuclear Auxiliary Power) radioisotope thermoelectric generator, failed to orbit and re-entered the atmosphere over the Indian Ocean. The SNAP-9A contained about 17 kCi of plutonium-238 and particles from this burnup were detected in the Northern Hemisphere in late 1965 and have been measurable since that time. Another radioisotope thermoelectric generator (SNAP-27), which contained about 45 kCi of plutonium-238, was aboard the aborted Apollo 13 mission and re-entered the atmosphere and is believed to be intact on the bottom of the ocean.

Plutonium has also been released to the environment on a local level around plutonium testing and production sites. The greatest quantity of plutonium is dispersed at the Nevada Test Site, and appreciable amounts remain at the Trinity Test Site, Alamagordo, New Mexico, where the first atomic bomb test occurred. About 3 to 5 curies have been released beyond the site limits of the Rocky Flats plutonium processing plant near Denver, Colorado. An estimated 4 curies were leaked in 1973 from a waste storage tank at Hanford, and similar quantities have been released at the Savannah River Laboratory site in South Carolina and at the Mound Laboratory in Miamisburg, Ohio.

There have been two major accidents involving nuclear weapons. The first occurred in January 1966, near the Spanish village of Polomares in an aerial refueling explosion involving a B-52 bomber carrying four plutonium-bearing nuclear weapons. One of the weapons was recovered intact from the ocean floor and another recovered intact from a dry river bed near Palomares. The high explosive charge on the other two exploded on impact, and the plutonium was dispersed into the air and subsequently, deposited on agricultural areas. Extensive decontamination procedures, including the removal of vegetation and soil, were used to lower the concentrations to safe levels.

The other major accident occurred in January 1968, when a B-52, with four plutonium-bearing nuclear weapons, crashed while making an emergency landing at Thule Air Force Base in Greenland. The high



Accumulated deposit of ^{239}Pu at sites sampled in the United States during 1970

(mCi/km^2)

Figure V-1

explosive charge on all four weapons exploded and the plutonium was dispersed into the burning fuel. Decontamination included removal of the top layer of ice over about 15 acres.

Surveillance programs are routinely carried out at plutonium handling and processing facilities. A summary of these programs along with available data on plutonium concentrations at each facility is given in Appendix I. Data is also provided for the Trinity and Nevada Test Sites.

EPA and its predecessors began monitoring the plutonium-238 and -239 levels in the atmosphere in 1965 following the burnup of the SNAP-9A. The current air concentrations in the United States as measured by this network are 0.2 to 4 aCi/m^3 (aCi = attocurie = 10^{-18} curie) for plutonium-238 and 2 to 40 aCi/m^3 for plutonium-239.

VI. Environmental Transport

Plutonium released to the environment is transported to man via major ecosystem components (air, soil, water, microorganisms, plants, and animals) by a variety of environmental (physical) and ecological (food chain) processes. Analysis of the environmental pathways provides estimates of plutonium inhalation and ingestion rates by a real or hypothetical person living in the contaminated ecosystem. These estimates of plutonium input rates to man provide a basis for calculating potential radiation doses and dose commitments to critical organs. The resulting dose estimates then serve as a basis for evaluating the hazard to man due to releases of plutonium to the environment.

Details of the environmental pathways analysis will vary with respect to the characteristics of: (a) plutonium releases, (b) the environment contaminated by the releases, and (c) the dietary habits of the population living in the contaminated environment. The major environmental transport pathways for a typical terrestrial ecosystem include (1) exchange between air and water or vegetation by deposition and resuspension, (2) exchange between soil and water by erosion, leaching, adsorption and precipitation, (3) uptake from soil by plants, animals and man, (4) inhalation by animals and man, (5) ingestion of food and water by animals and man, and (6) redistribution within plants, animals, and man.

Enough is presently known about the pathways to conceive preliminary mathematical models for the environmental transport of plutonium. These usually take the form of systems of ordinary differential equations which describe the transport between major ecosystem compartments. Values and functional forms of the coefficients and parameters in these equations depend on the details of the pathways. In order to compensate for the current lack of knowledge for certain processes, the mathematics of the models is greatly simplified and conservative coefficient values which will tend to overestimate the potential plutonium hazard are used.

The environmental pathways of the transuranium elements are outlined in Figures VI-1 and VI-2. Air and water are the primary transport pathways in the environment, while the soil and the oceans are considered primarily as storage reservoirs from which small amounts of these elements become incorporated into food. The primary pathway of human exposure has generally been assumed to be by way of inhalation of air containing the transuranium elements. The air concentration results from air releases from the source or from resuspension of these elements from the soil back into the air. In areas with much plant cover or as the elements migrate downward in soil, the inhalation pathway may become less important than the food pathway.

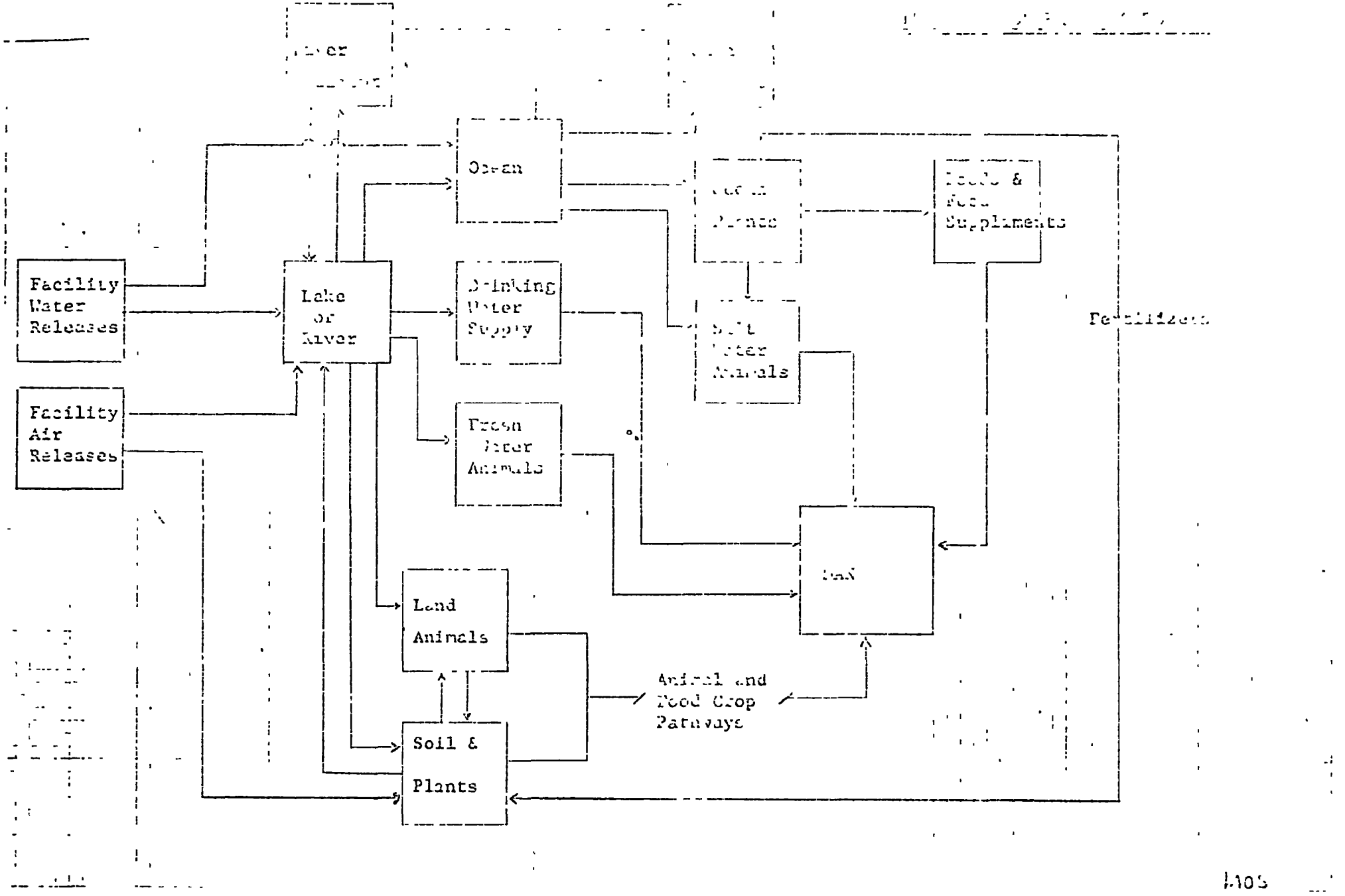

Ocean
Facility Water Releases
Lake or River
Drinking Water Supply
Facility Air Releases
Fresh Water Animals
Land Animals
Soil & Plants
Animal and Food Crop Pathways
Fertilizers

Figure VI-1

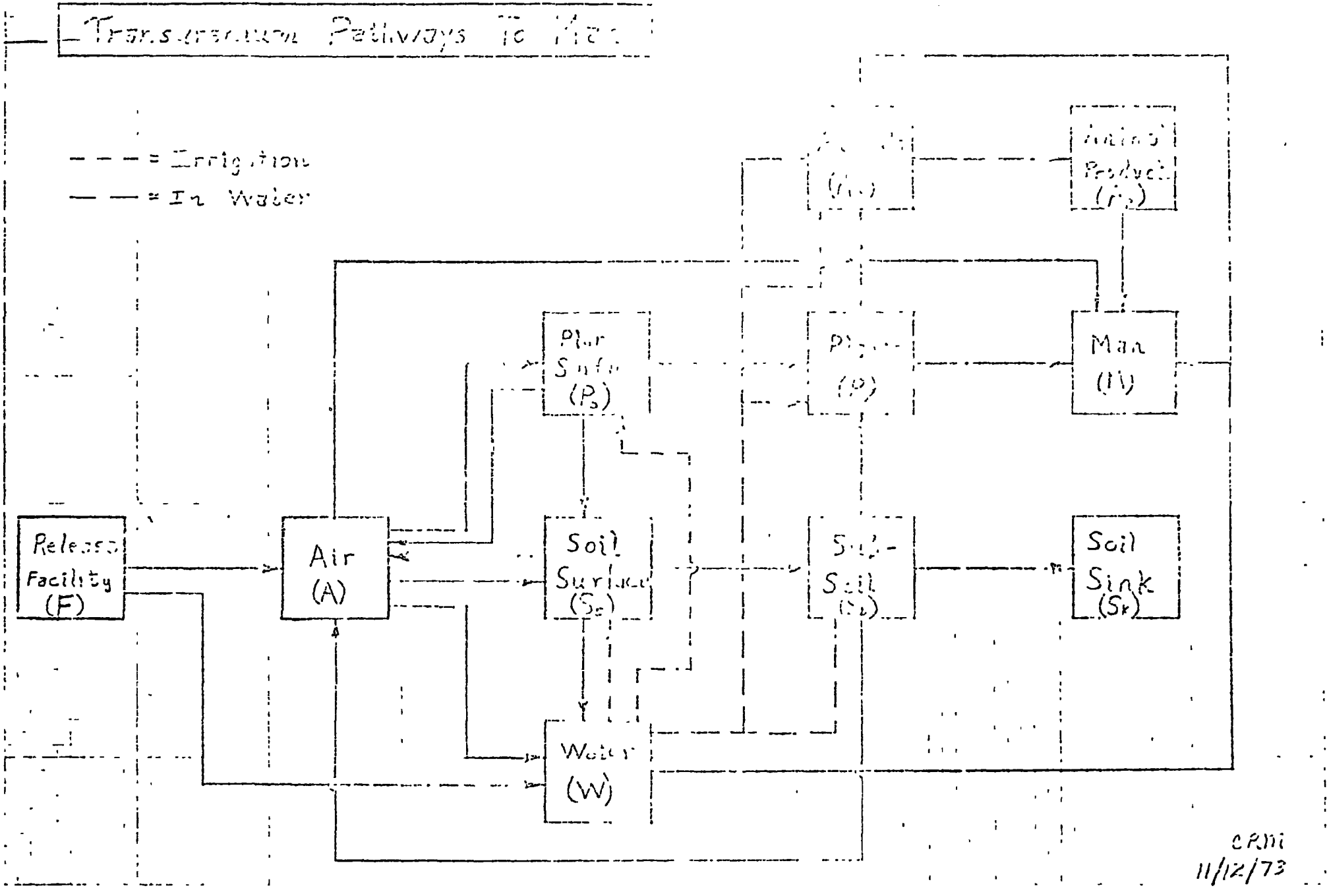

Transportation Pathways to Man
= Irrigation
= In Water
Animal Product (Pa)
Plant Surface (Ps)
Plant (P)
Man (M)
Release Facility (F)
Air (A)
Soil Surface (Ss)
Soil (Sl)
Soil Sink (Sk)
Water (W)
CPM
11/12/73

Figure VI-2

Air Transport

When transuranium elements enter the environment via the air mode, they are rapidly and widely transported by air currents to a more-or-less permanent distribution in the soils and oceans. Deposition occurs by both wet and dry methods. Many computer programs are available to describe the spreading out within the first 50 to 100 km of radioactive material released from a point source. These take into account wind speed, direction, and other meteorological parameters, and some account for cloud depletion by surface deposition.

Surface deposition occurs both by washout from the air by rain, and by dry deposition as the contaminated air touches the ground. The deposition rates for each of these methods are not very well known. For dry deposition the concept of deposition velocity is used. The deposition velocity is the ratio of the surface deposition over a given period of time and an average air concentration during that time. Values that have been measured for various radionuclides generally fall between 0.1 and 2.0 cm/sec.

For wet deposition the ratio of the concentration in rainwater to that in ground level air is used to determine total deposition. Most observed values of this ratio fall in the range of 100 to 1000.

A considerable fraction of the released radioactive material still remains in the air at distances of 50 to 100 km from the source. Very little has been done to study the fate of this material. However, several mathematical models have been developed to follow this material. The most simplistic model assumes that 50% of all air releases of transuranium elements in the U.S. are spread uniformly over the Eastern half of the U.S. The rest is deposited over the Atlantic Ocean. A more detailed model utilizes weather patterns and the wet and dry deposition techniques mentioned previously to calculate, with the aid of a computer, deposition patterns around a specific source. Using a deposition velocity of 0.1 cm/sec and a rain-to-air ratio of 500, it was found that for a source in Morris, Illinois, the amount deposited on the Eastern U.S. and Canada amounted to between 50 and 70 percent of that released with the remainder being deposited in the Atlantic Ocean. Thus the predicted patterns of deposition for the two independent models are consistent.

Ground Pathways

Transuranium elements deposited on the surface of the ground may expose the population through several pathways: resuspension of the deposited material back into the air, plant uptake through leaves and roots, animal uptake and subsequent incorporation into

the animal products, or incorporation into drinking water. The principal uncertainties associated with a determination of pathways to man are those of long-term transfer mechanisms. The extremely long persistence of these nuclides makes it necessary to predict ecological processes for decades and centuries. Very little is yet known to permit accurate predictions of such parameters and more research will be required to refine current estimates.

Resuspension is generally considered to be the major pathway of exposure. The resuspension factor, the ratio of air concentration due to resuspension to surface contamination levels, for freshly deposited material is estimated to range from 10^{-3} m^{-1} to about 10^{-8} m^{-1}, and decrease to 10^{-7} m^{-1} to 10^{-11} m^{-1} or less some time after deposition. Estimates fo this ratio are extremely variable with respect to time and numerous environmental factors (such as wind speed and direction, rainfall, disturbances affecting aerodynamic properties of soil surfaces) as well as the aerodynamic properties of plutonium-bearing particulates and their susceptibility to saltation and resuspension.

Plant and animal uptakes are very small, with the concentration in plants being generally less than 10^{-4} of that in soils and the concentrations in animals being about 10^{-5} of that in the plants they eat. There is some evidence, however, that the longer plutonium remains in soil the greater the plant uptake becomes.

Eventually much of the transuranium elements may diffuse far enough into soil so that resuspension and plant uptake will not occur. The soil would then be considered a "sink", or environmental removal mechanism, for these elements. Mathematical models to describe this diffusion mechanism are under development.

Water Pathways

One possible water pathway model for the movement of transuranics through the environment to man is given in Figure VI-2. While the model is not meant to be definitive, it does indicate the major pathways currently recognized and the complex interactions which can occur. The figure also shows the interrelationship of a water pathway model with an air pathway model.

Contamination of the drinking water supply represents the critical pathway for population exposure immediately after a liquid release, and movement of the transuranics through the complex ocean ecological systems may well represent the critical pathway for long term exposures. A study of the California coast indicated that fish, invertebrates, and plants will concentrate plutonium. Some very preliminary, unpublished data of plutonium concentration profiles in marine animals, plants and waters surrounding the

British Isles indicate that plutonium may remain in high concentrations near the shore. It is not rapidly lost or dispersed to the open oceans as was once thought. Additional preliminary data would seem to indicate that plutonium deposited in bottom sediments may become resuspended rather than remain fixed with the sediments. These early reports and the reconcentration effect of marine organisms reported above would suggest that the ocean may not be an infinite sink for liquid releases of transuranium, but it may indeed be a critical pathway for long-term population exposures.

VII. Health Effects

Plutonium and many of the transuranium elements emit alpha radiation, which has very low penetrating powers and is effectively damaging to tissue at only very short range. Therefore, these elements must be in immediate contact with a receptor to be harmful. Transport of these elements into the human body is generally via inhalation, ingestion, or contamination of an open wound. Once in the body, the elements may be retained by the lung or deposited at various sites including the liver, skeleton and soft tissues. The long half-lives of the elements involved makes them especially dangerous because of the possibility of cumulative damage over a long period of time.

To determine the effects of exposure to the transuranium elements, models have been developed to estimate the retention of these elements by various organs and to convert the resultant tissue exposures to doses. A more detailed discussion of these models is given in Appendix II. Most of the modeling and risk estimates developed have been based on experience with plutonium and this information extrapolated to the other transuranium elements.

Inhalation

The respiratory tract is the most common mode of entry into the body. The particles inhaled may be either soluble or insoluble in body fluids. Soluble compounds tend to move to other areas of the body and can concentrate in the liver or bone. Insoluble compounds tend to remain in the respiratory tract for long periods of time. For modeling purposes 1000 days is used for the clearance half-time from the lung.

Considerable uncertainty persists in evaluation of the biological hazard of inhaled radioactive particles. Although it is possible to make estimates for desposition of inhaled particles in the lung, and estimates of the risk of adverse effects following exposure of the lung to ionizing radiation, it has proven difficult to obtain agreement on the risk associated with the inhalation of radioactive particles.

The estimates of risk for inhaled plutonium are based on animal data and on human data from two sources. The first is from total organ or whole body exposure to x-rays or gamma rays, where every cell in the lung receives approximately an identical radiation exposure. This includes data on Japanese survivors from Hiroshima and Nagasaki, and on patients who have been exposed to extensive diagnostic or therapeutic x-irradiation of the chest and lungs. The second source of data is derived from persons exposed to the special conditions found in some mines. These persons inhaled not only

radioactive particles but also rock dust, radon and daughter radioisotopes, diesel engine exhaust and other materials. The radon daughter isotopes are thought to be the causes of lung cancer in this case, and the radiation exposure of cells in various regions of the lung is different. From these data an estimate of the risk of lung cancer per rem of radiation has been derived.

Hot Particle Problem

The question of whether the dose, and the associated cancer risk estimate, resulting from inhalation of a particular quantity of an alpha emitter, such as plutonium, should be derived from the average exposure of all cells in the organ or for only those cells within the small volume intensely irradiated was first raised in 1949 and has been the subject of debate ever since. Traditionally, dose estimates for internal emitters (radioisotopes contained within the body) have been based on the average organ dose, i.e. the total amount of radiation energy deposited in an organ divided by the mass of that organ. While this is a reasonable method for estimating the risk from x- or gamma radiation where all parts of the organ receive the same amount of energy, it has been questioned whether it is adequate for estimating the risk from the short range alpha or low energy beta radiation in cases where the radioactive material is not uniformly distributed. This is often designated as the "Hot Particle" problem.

In the case of plutonium-239, in solid tissue the dose rate at 40 μm from the surface of a particle is about 1.5 to 2.0% of the dose rate of the source point and by 45 μm distance the dose rate is virtually zero. In the lung, which is about 80% air, the corresponding distances are about 320 μm and 480 μm respectively. Depending on the number of particles inhaled, the fraction of the lung actually irradiated may be quite large or small but usually only a small portion of the lung will be exposed to the radiation.

Animal data which may provide additional support to the hypothesis concerning the relative efficacy of particulate vs uniform exposure is conflicting and has many limitations.

The anatomy, respiratory physiology, histology, and pathology is different to a greater or lesser extent in each species of animal studied. Because of these differences and the fact that lung tumors developing in animals exposed to radiation are not the same as those seen in man, it is difficult to assess the consequences of inhaled radioactive particles in man. There is no data on human exposure from which a correlation with animal inhalation experiments can be made. Until an adequate animal model is developed there will be

great uncertainty and disagreement about the relationship of animal experiments to possible human exposure and risk.

Ingestion

Although inhalation is considered to be the most important route of entry into the body, recent experiments with cattle indicate that the fraction going from the GI tract to blood may be as high as 10^{-3}. Although absorption from the intestinal tract into the bloodstream is poor, it should not be ruled out as a skeletal hazard, especially when those exposed are extremely young. Plutonium entering the bloodstream will distribute itself between bone and the liver in varying proportions depending upon the chemical state of the contaminent and whether the individual exposed is an adult or child. The potential ingestion hazard to man, especially over the long periods of time these elements may remain in the environment, is indeterminate. Short term exposures are reduced by the fact that the amount of plutonium deposited on external parts of plants is usually much greater than the amount of uptake, and this external contamination is removed by washing. However, over longer periods of time, reconcentration may occur in certain plants and the availability to man through the food chain may increase. Similarly, reconcentration in animals may occur over a number of years of exposure.

Wound Contamination

Plutonium deposited in a wound will react with the constituents of the tissue and the body fluids and be absorbed into the blood stream where experiments have shown it to eventually deposit in the skeleton.

APPENDIX I

MODELS FOR PLUTONIUM DOSIMETRY

Models for Plutonium Dosimetry

Inhalation

The model used by EPA currently for estimating deposition and retention of inhaled plutonium is based on the ICRP Task Group on Lung Dynamics (TGLD) Model [1,2] as modified for ICRP publication #19. (Table 11).

In this model three chemical classes of compounds are postulated. Class Y Compounds include carbides, oxides, hydroxides and tetravalent fluorides (rapid translocation of cerium oxide from the deep lung suggests the possibility of exceptions to this class Y retention for some actinide compounds). Class W Compounds include nitrates, carbonates and lanthanide halides and phosphates. Class D Compounds include those not listed above. Information is not available on most plutonium compounds, but the readily hydrolyzed, or incompletely soluble, suggests that no actinides should be considered a class D compound.

The parameters used for estimating fractional deposition within regions of the lung for particles of different activity median aerodynamic diameter (AMAD) are based on the [illegible] male, who is breathing through the nose at a rate of 15 [illegible] per minute and [illegible] deposition [illegible] or penetration [illegible] is based on the [illegible] adjustments for continuous intake of [illegible]

The organ burden is calculated on the basis of:

$$\text{Organ Burden} = I \int_0^t R_j(\tau)\, e^{-\lambda\tau}\, d\tau$$

where I = daily intake by inhalation

$R_j(\tau)$ = retention function for the j th organ during time τ

τ = period of exposure

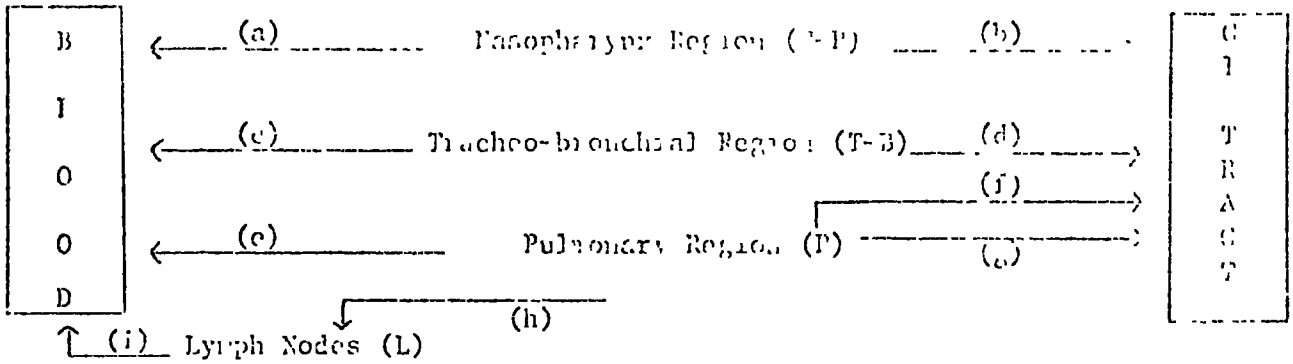
t = period over which the dose is computed

λ = physical half-life of the nuclide

In principle, the organ burdens can then be used with appropriate dose-conversion factors to calculate the exposure dose to the organ in rem per year. The organ dose in turn can be used to estimate possible health effects, e.g. malignancy in either terms of individual risk or population risk.

TABLE I

TGLM Model



Clearance Constants for TGLM Model

Region	Pathway	Compound Class X	Compound Class Y	Compound Class W
N-P	(a)	1%/0.01 day	10%/0.01 day	50%/0.01 day
	(b)	99%/0.4 day	90%/0.4 day	50%/0.01 day
T-B	(c)	1%/0.01 day	50%/0.01 day	95%/0.01 day
	(d)	99%/0.2 day	50%/0.2 day	5%/0.01 day
P	(e)	5%/500 days	15%/50 days	80%/0.5 day
	(f)	40%/1 day	40%/1 day	---
	(g)	40%/500 days	40%/ 50 days	---
	(h)	15%/500 days	5%/50 days	20%/0.5 day
L	(i)	90%/1000 days	100%/50 days	100%/0.5 day

In actual practice the uncertainty in such estimates may make some calculations meaningless, as outlined below.

Ingestion of Inhaled Plutonium

Some of the inhaled Plutonium will be transported to the GI tract and part will be absorbed there just as if it were ingested. For the nominal 1.0 µ AMAD aerosol used by ORP, the relative percent distribution by region is listed in Table 2. The calculated daily transport to the GI Tract indicates that about 35% of the daily inhaled plutonium will be transported to the GI Tract.

Ingestion of Plutonium

Absorption of plutonium from the GI Tract is estimated to be 10^{-4}% for insoluble forms such as PuO_2 and 3×10^{-3} for more soluble forms (due to hydrolysis)[1]. However the absorption may be much greater for [illegible] solution (up to [illegible]), for certain forms of plutonium (up to [illegible]) and in younger animals (possibly a factor of 100 increase in absorption).

Table 2

Region	Percent Deposition (1.0 AMAD)	Amount Transported to GI Tract: Class D Compounds	Class Y
N-P	30%	25.0%	22.[illegible]
T-B	3%	2.9%	1.[illegible]
P	34%	9.6%	9.6%
		37.5%	33.4%

There is no current ORP model for ingestion, however a model similar to that used for inhalation is applicable:

$$\text{Organ Burden} = I \int_0^t R_j(\tau) e^{-\lambda \tau} \, dt$$

if I = daily intake by ingestion

$R_j(\tau)$ = retention function during time τ for organ j

τ = period of exposure

t = period over which the dose is computed

λ physical half-life of the isotope

As before, the organ burden can be used with appropriate dose-conversion factors to calculate the organ dose in rem per year. From this value the number of adverse health effects can be estimated with various degrees of certainty, as outlined below.

SKIN CONTAMINATION

The case of skin contamination as a route of exposure for plutonium can be neglected unless the skin is not intact. [illegible] skin is damaged, there is not enough data to [illegible] estimate of the rate or amount of absorption [illegible]

DEPOSITION [illegible] [1]

Lung: More than half of the [illegible] deposited in the lung is retained in the lung with a [illegible] for the case of Class Y Compounds. For Class Y Compounds the value is [illegible] with a 50 day half-life.

Liver and Bone: About 3.2% of the Class Y material deposited in the lung is translocated to the liver and bone surfaces with a half-life of 1000 days. In Class W compounds the translocated compounds is retained with a half-life of 50 days.

Skeleton: 45% of the plutonium in the circulatory system is expected to deposit in the skeleton and to retain with a half-life of about 100 years. Only 10% of very stable biologically inactive complexes of plutonium (e.g. plutonium-DTPA) would deposit in the skeleton.

Liver 45% of the plutonium in the circulatory system may deposit in the liver. It would be retained with a half-life of 40 years. As in the case of the skeleton only 10% of stable biologically inactive complexes may deposit in the liver.

Soft Tissues About 2% of the plutonium in the circulatory system may be deposited in soft tissue in the spleen, ovaries, uterus, testis, adrenal plutonium deposits are retained for extended periods with/500 days or longer half times. However, there are no ICRP estimates of percent deposition in these tissues.

There is evidence that higher specific activity isotopes, e.g. ^{237}Pu and ^{233}Pu, are translocated more rapidly than ^{239}Pu. This difference probably reflects the reduced amount of colloidal material seen with high specific activity radionuclides.

Dose Conversion Factors

Dose conversion factors are based on the dose-equivalent rate for unit intake.

$$DE \quad \frac{51.2 \cdot E \int_0^t R_j(\tau)\,d\tau}{H}$$

where D_E = dose equivalent for the organ (rem/day)

51.2 = gram-rads per MeV

E = effective energy deposited in the organ (MeV/disintegration) and includes the quality factor for the type of radiation emitted

H = mass of organ (grams)

$R_j(\tau)$ = retention fractions during time τ for organ j

t = period over which the dose is computed

τ = period of exposure

λ = physical half-life of the isotope

Risk Estimates for Biological Effects of Plutonium in Man

At the present time, BEIR is making risk estimates based on data in "The Effects on Populations of Exposure to Low Levels of Ionizing Radiation" (BEIR Report, 1972).

Risk estimates are made both for populations after single exposure to radiation above background levels and for populations exposed continually to radiation above background levels (Table 4). None of these risk estimates are based on effects observed with plutonium nor are they directly relatable to the dose delivered by some of the exposure simulations or environmental situations. Only for risk due to exposure of bone to alpha radiation from deposited plutonium can the risk estimates be applied.

Table 4

The Dose-Risk Conversion factors in current use are:

(a) Risk following continuous lifetime exposure:
200 excess cases/yr/10^6 person-rem
200 excess fatalities/yr/10^6 person-rem
300 excess cases/yr/10^6 person-rem

(b) Risk for specific organs exposed:

Leukemia - 50 excess cases/yr/10^6 person-rem annual exposure

Lung - 50 excess deaths/yr./10^6 person-rem annual exposure

Bone - 12 cases/yr/10^6 person-rem annual exposure, 50% of the cases are fatalities

GI System - 60 cases/yr/10^6 man-rem annual exposure, 50% of the cases are fatalities

Other - 224 cases/year/10^6 man-rem annual exposure, about 25% of the cases are fatalities

(c) Thyroid cancers are considered separately since they are extremely age dependant.

Thyroid cancers in various age groups are:

0 - 1 yr of age - 93 cases/yr/10^6 man-rem annual exposure

1 - 9 yrs of age - 50 cases/yr/10^6 man-rem annual exposure

10 - 19 yrs of age - 66 cases/yr/10^6 man-rem annual exposure

20 years of age and older - 60 cases/yr/10^6 man-rem annual exposure

About 20% of the thyroid cancer cases would be fatalities. The risk rates in cancers/yr/10^6 man-rem annual exposure are numerically equal to the total number of cancers/10^6 man-rem from a single exposure.

Problems in [illegible] Modeling [illegible] Plutonium [illegible] Inhalation

There are a [illegible] of problems [illegible] plutonium [illegible] physiological and biological [illegible] factors [illegible].

To some extent the ICRP Task Group on Lung [illegible] Model [illegible] conservative in assumptions for inhalation [illegible].

The model uses a tidal volume of 750 cc or 1450 cc. [illegible] values are adequate for an adult [illegible] adult [illegible] be 333 cc or 633 cc, [illegible] sleeping [illegible] born.[5] In this aspect the model provides [illegible] exposure which are higher than would be expected in a general population.

The ICRP Model has other assumptions which are not conservative.

(1) The model uses a respiratory rate of 15 respirations per minute. For a heterogenous aerosol the percentage of deposition varies with breathing rate. The minimum level of deposition occurs at 15 to 20 respirations per minute and increases on either side of this value.[4] Deposition rates in hard workers, or in sleeping or sedentary individuals would be higher than the model predicts.

(2) No provision is made in the model for the known regional distribution of inhaled gases within the lung. [illegible], [illegible] and Bryan have shown that in the upright individual, the distribution and rates of wash-in and wash-out of gases is different by about 40% between the upper and lower zones of the lung.[5] This has profound implications in that it affects both the distribution of inhaled aerosol and settling or diffusion within alveolar.

(3) The model is based on laminar flow in tubes at a constant rate per the calculations of Findeisen and Landahl[1]. A [illegible] of the assumptions in the model that need refining include:

(a) The pattern of air flow during respiration is not constant, but goes from zero to [illegible] and then returns to zero. [illegible] on the effect of this pattern on deposition.

(b) Pulsating air flow is caused within the lung by the filling and emptying of the heart. The effect of this action on local deposition is not known.

(c) Air flow in the [illegible] be investigated.

(d) The bulk of new air [illegible] with lung air. [illegible] will penetrate only [illegible] while inner part [illegible] penetrate the depth [illegible] similar to [illegible] implications [illegible] particularly as it relates to [illegible].

(e) The respiratory tree is not composed of circular tubes but irregular cross section tubes often corregated or folded over. The effects of these irregularities on turbulance and deposition are not known.

(f) The effects of respiratory [illegible] (con[illegible]) on respiratory clearance are not considered.

Even if the lung model were considered to be accurate enough for practical purposes, difficult problems peculiar to plutonium distribution and retention are not settled. There is evidence that the high specific activity isotopes of plutonium ([illegible], ^{237}Pu) are translocated from the lung more readily than [illegible] and have a different distribution pattern in the body after translocation.

In general the ^{238}Pu has a distribution pattern after translocation from the lung which resembles the pattern of injected monomeric [illegible]Pu.[a] [illegible]Pu is translocated from the lung the pattern of distribution which has been [illegible] case of injected polymeric [illegible]Pu.[b] This suggest that the [illegible]Pu is [illegible] being transported as a transferrin [illegible] the [illegible]Pu [illegible] and transported as particulate [illegible] (P. Durbin, IRPA Congress, 1973).

It has been suggested that the more rapid translocation of ^{238}Pu [illegible] to [illegible] effect [illegible]. The [illegible] and the [illegible] 197[illegible]). [illegible] developed for each [illegible].

The available [illegible] of [illegible] for [illegible] of plutonium [illegible] subject to [illegible]. The data on retention particularly [illegible] relatively high levels of exposure.

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a/ Monomeric plutonium [illegible] the form of [illegible] plutonium compound, not a large number of particles aggregated.

b/ Polymeric plutonium [illegible] a form where a number of molecules of the plutonium compound aggregate together as a colloid.

c/ Transferrin is the serum protein which binds iron as an iron-transferrin complex to transport the iron throughout the body.

Dogs at Hanford, exposed to $^{239}PuO_2$ had retained lung burdens of 1.5 to 900 nCi/g of lung (100 to 27,000 times the maximum permissible lung burden for man)[8]. One of the most common responses to this high level of exposure is local cell death followed by the possibility of scarring in the area[24]. The radiation induced changes in the tissue at the high levels of exposure used change the functions of the tissues and interfere with normal metabolism. [illegible] retentions seen at these dose levels to lower [illegible] [illegible] suspect until confirmatory evidence is available.

Much environmentally distributed plutonium has the unusual property of being in the form of very small submicron particles weakly attached to larger dust particles. Unfortunately [illegible] model does not address the question of extremely small particles bound to 1.0 μ (or larger) particles. This question must be addressed since any [illegible] must consider [illegible] for plutonium that escapes through HEPA filters.

Problems in Dosimetric Modeling for Ingested Plutonium

Problems related to [illegible] of plutonium [illegible] plutonium [illegible] of 3 × 10⁻ [illegible] the [illegible] human [illegible] of [illegible] in dairy cattle [illegible] absorption values for [illegible] and being different [illegible]

Problems in Dose Commitment Models for Plutonium [illegible]

Distribution of inhaled or ingested plutonium in the body

Although quite a lot is known about distribution [illegible] of injected plutonium compounds, less information is available on what happens after inhalation or ingestion.

Current evidence suggests the ingested plutonium enters the circulatory system in ionic form and is distributed through the body as a plutonium-transferrin complex much as monomeric injected plutonium. In this case the skeleton; liver; tissue and excreta distribution would be about 60%; 20%; 20%.[7]

lymph nodes, liver, skeleton, kidney and occasionally the spleen. A real animal contains many more tissues.

The calculation of dose from inhaled plutonium particles is complicated by the fact that the tissue in which the [illegible] really is deposited is not known. Since plutonium [illegible] "hot particles" or is a diffuse aerosol, [illegible] not deposited [illegible] lung parenchyma, the dose is unevenly distributed in the lung. Also, even though the range of plutonium alpha particles may be only 50μ, the mass of the whole lung is used in calculating the dose. These inequalities are certain to lead to under estimation of dose to the tissues being irradiated, [illegible] to a very [illegible] under estimate of dose to critical tissues. More data is needed for better dose calculation.

Problems in Using Health Effects Data With Plutonium Models

Even if the inherent uncertainty in health effect estimates calculated by the BEIR Committee is disregarded, care must be taken in applying these estimates to [illegible]. BEIR risk estimates are based on observations [illegible] of [illegible] well defined tissues. [illegible] [illegible] [illegible] risk of bone cancer from [illegible] dose and risk of leukemia based on [illegible] [illegible] (BEIR [illegible]). [illegible] dose [illegible] basal cells of the bronchi [illegible] dose [illegible] lung [illegible] BEIR can be [illegible] the specific [illegible] to BEIR risk estimates can be computed. [illegible] the thyroid dose from plutonium can be calculated, BEIR risk estimates for thyroid related health effects can be used to determine the expected health effects.

There are also unresolved questions in interpreting the observed effects of plutonium on animal health and correlating them with man for whom we have no observations.

(a) Liver and bile duct tumors have been observed in animals, but there is no corresponding data for man.

(b) Lung tumors have been observed in animals but they are generally alveolar in origin. The tumor for which human risk data is given in man is a bronchogenic tumor. There is no concensus yet on how or if the human and animal tumors relate to one another.

(c) Osteosarcomas have never been observed in animals exposed to [illegible] PuO_2 aerosols, however osteosarcomas have developed in animals exposed to [illegible] PuO_2 aerosols. (D Craig, [illegible] Congress 1978). Whether this means that health effects models must also be [illegible] plutonium isotopes or not is unclear.

(d) The effects on animals [illegible] of [illegible] versus distr[illegible] ticles in "hot spots" [illegible] c[illegible] in [illegible].

Attachment A

Dose conversion values currently being used by the Environmental Protection Agency, Office of Radiation Programs:

Nuclide	Lung (mrem/yr per pCi/m^3 conc)
Pu-238	12
Pu-239	12
Pu-240	12
Pu-241	.012
Am-241	3
Cm-242	2
Cm-244	4

These values are [illegible] and are under study and new values are anticipated.

REFERENCES

1. Task Group on Lung Dynamics
   Deposition and Retention Models for Internal Dosimetry of the Human Respiratory Tract.

   Health Physics 12:173-207 (1966)

2. ICRP Publication #19
   The Metabolism of Compounds of Plutonium and Other Actinides.

   Pergamon Press N. Y. 1972

3. Wm. S. Spector Handbook of Biological Data, W. B. Saunders Co., Philadelphia 1956

4. T. F. [illegible] and P. Gross Pulmonary Deposition and Retention of Inhaled Aerosols,

   Academic Press, New York [illegible]

5. D. V. Bates, W. C. [illegible] and [illegible] Use of Xenon-133 in Studying the Ventilation and Perfusion of the Lung. [illegible] Symposium Series [illegible] Dynamic Clinical Studies with Radioisotopes [illegible] 1964

6. [illegible]
   [illegible]
   at [illegible]

   Health Physics [illegible] (1972)

7. E. [illegible]
   in [illegible]
   [illegible]

8. R. O. McClellan, J. [illegible] C. [illegible] H. C. [illegible] Inhaled in Fused Clay Particles-an Experimental Approach, pp 395-416 IBID

9. R. E. Stanley and E. W. Bretthauer. Absorption, Distribution and Excretion of Plutonium by Dairy Cattle-A Preliminary Report Oct. 1973

10. Plutonium in Autopsy Tissue, LA-4875. 1973

APPENDIX II

ENVIRONMENTAL SURVEY RESULTS

ARGONNE NATIONAL LAB, 1973 (ANL-8078)

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Air:	Monthly filter samples: -9 on-site 6 off-site	At one location filter was changed daily, all others were changed at weekly intervals.	Error is between 5 and 20% for most results but approaches 100% at the detection limit. Average on-site concentration Pu-238: 0.85 aCi/m^3 Pu-239: 12.5 aCi/m^3 Average off-site concentration Pu-238: 0.46 aCi/m^3 Pu-239: 9.9 aCi/m^3	Similarities between samples collected by other labs indicate that the plutonium in Argonne samples is primarily from fallout.
Water:	Argonne waste water discharged into Sawmill Creek which flows into the Des Plaines River. Samples collected below outfall on continuous basis and individual samples collected five times weekly.	Plutonium & neptunium analyses were performed on 10-liter samples by a Pu separation method, followed by alpha spectrometry.	Principal radionuclide was tritium. Average Pu concentration: Sawmill Creek: 5.1 fCi/liter Des Plaines River: 1.0 fCi/liter	Plutonium is attributed to fallout.
Soil:	Samples collected downwind of Bldg. 205. Samples collected near buildings in which plutonium had been used. 5 on-site samples 9 off-site samples	Each soil sample consisted of two cores totaling 173 cm^2 in area by 30 cm deep.	Average on-site concentration Pu-238: .17 nCi/m^2 Pu-239: 2.68 nCi/m^2 Average off-site concentration Pu-238: .22 nCi/m^2 Pu-239: 2.64 nCi/m^2	Results for Pu within fallout concentrations as measured by other laboratories.

ATOMICS-INTERNATIONAL, 1973

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Air:	On and off-site locations; number not specified.	Automatic continuous sequential samplers, filters analyzed daily for gross α and β. Sample ~ 20 m^3 per day.	Average gross alpha at Headquarters and NDFL was 7.5 and 7.2 fCi/m^3, respectively.	
Water:	Various reservoirs and creeks.	One liter sample collected. 500 ml evaporated and counted for gross α and β.	Values averaged $<.26 \pm .26$ pCi/liter	
Soil:	144 on-site samples 48 off-site samples.	Top 1/2" of undisturbed soil collected (no area specified). Two-gram aliquot placed in planchet and counted directly for gross α and β.	Gross Alpha (pCi/gm of soil) On-site 0.57 Off-site 0.51	Also provide summary of soil radioactivity data from 1964 thru 1973, broken down to on-site vs off-site.

BATTELLE COLUMBUS LABORATORIES, 1973

Media	Sampling Program	Sampling Techniques & Analysis	Results Reported	Remarks
Water:	Effluent stream monitored at release point after filter bed.	One-liter samples collected weekly. Analyzed for alpha and beta by Eberline Institute Corp.	None included in report.	0.16 mCi of alpha activity released from West Jefferson site.
Air:	Exhaust stacks sampled with continuous air monitors. Samples changed weekly. (No ambient air monitoring network.)	Samples counted for "appropriate radionuclides" and gross alpha and beta.	None included in report.	
Foliage: (grass)	Samples collected at 14 locations, spring and fall.	Samples composited and ashed. Analyzed for gross alpha and beta, Pu, and ^{90}Sr - ^{90}Y, and reported as pCi/gm of ash. (No surface area relationship given.)	Plutonium (pCi/gm of ash): ^{238}Pu Max. 0.005 Min. 0.005 Avg. 0.005 ^{239}Pu 0.018 0.005 0.007 (Results of 14 composites)	

HANFORD, 1973 (BNWL-1811)

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Water	Samples drawn quarterly from two Columbia River locations. A) Vernita B) Richland	Extraction with diethyl ether. Count dried residue with gas flow alpha proportional counter.	Less than 0.001% of radiation concentration guide.	Alpha concentration is 1.5% of radiation concentration guide for an unknown mixture of alpha emitters.
Air:	21 off-site stations--locations not defined.	Analyses made quarterly on composited filters counted for gross alpha and beta concentrations.	Average concentration of alpha plutonium was 1.5 pCi/m^3.	
Soil and Vegetation:	Thirteen stations for routine sampling around the perimeter of the Hanford Reservation. Annual determination.	Samples of the top inch of soil and native vegetation (perennial) were taken at each station.	Average concentration in soil: Pu-238: < 3 fCi/gm Pu-239: 10 fCi/gm Average concentration in vegetation: Pu-238: 1 fCi/gm Pu-239: 2 fCi/gm	The plutonium concentrations are typical of general regional levels for the arid and western states.

LAWRENCE BERKELEY LABORATORY, 1973 (UCID-3651)

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Air:	10 stations on site 4 stations perimeter	4 cfm through 4" x 9" HV-70, continuous sampling, changed weekly.	Gross alpha average on site: 0.4 fCi/m^3 perimeter: 0.6 fCi/m^3	Based on stack sampling, 0.5 μCi of alpha activity released during reporting period.
Deposition:	9 stations on site 4 stations perimeter	15" diameter cylinder. Rinsed with water if dry.	Average alpha on site: 0.04 nCi/m^2 perimeter: 0.08 nCi/m^2	
Sewage:	Two outfalls to municipal system. Sampled by continuous proportional samplers.	Assayed weekly.	Average concentration of alpha: 0.20 pCi/liter.	Total of <63 μCi of alpha activity discharged to sewers (isotopes not specified).
Surface and Tap Water:	Three on-site and two off-site streams sampled weekly.	One quart grab sample weekly.	Average alpha on site: 0.34 pCi/liter off site: 0.52 pCi/liter	
	One tap water location sampled weekly - location not given.	One quart grab sample weekly.	tap water: 0.08 pCi/liter	

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Air:	6 perimeter samplers: 25 cfm 11 off-site samplers: 4 cfm	Weekly samples analyzed for gross alpha. Monthly composite analyzed for Pu-238 and Pu-239 at perimeter sites. Selected composites analyzed for Am-241 Jan-May.	Perimeter average Pu-238: 4.3 aCi/m^3 Pu-239: 1.2 fCi/m^3 Am-241: 63. aCi/m^3	Compared to RCG of 1 pCi/m^3 for insoluble Pu.
Soil:	28 sites in Livermore Valley.	Samples collected to a depth of 0-1 cm.	Pu-238 average: 0.48 fCi/gm Pu-239 average: 5.9 fCi/gm	
	18 sites in San Joaquin Valley.	Samples collected to a depth of 0-25 cm for total deposition.	Pu-238 average: 0.026 nCi/m^2 Pu-239 average: .65 nCi/m^2	
	14 sites in surface drainage ditches and creeks which drain Lawrence Livermore Lab.	Some samples collected to 1 cm and others to 25 cm.	Pu-238 range: 0.035-4.1 fCi/gm Pu-239 range. 0.62-68. fCi/gm	
Sewage:	Weekly samples of 4 sites at the sewage treatment plant.	Gross alpha, Pu-238, Pu-239, and Am-241 analyses on monthly composite.	Pu-239 in effluent averaged 13 and 410 fCi/liter at 2 plants.	0.20 mCi of Pu-239 released to sewer.
SITE 300 SURVEILLANCE (13 miles SE of Livermore)				
Air:	10 on-site samplers: 25 cfm 1 off-site sampler: 1 cfm	Weekly samples except twice per week during the summer.	Pu-238 average: 1.6 aCi/m^3 Pu-239 average: 17 aCi/m^3	
Water:	13 on-site wells, off-site springs, ponds, and creeks.	Gross alpha analyses at different frequencies.	All gross alpha results below detection limit of 1.2 pCi/liter.	

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Air	16 off-site, 10 perimeter, and 10 on-site locations	Samples operated continuously, changed weekly. Composited monthly for Pu-238 and Pu-239 12 sites composited quarterly for Am-241	Off-site average Pu-238 15 aCi/m^3 Pu-239 21 aCi/m^3 Am-241 8 aCi/m^3 Perimeter average Pu-238 18 aCi/m^3 Pu-239 26 aCi/m^3 Am-241 5 aCi/m^3 On-site average Pu-238 10 aCi/m^3 Pu-239 15 aCi/m^3 Am-241 5 aCi/m^3	8.7 mCi of Pu-238, Pu-239, and Am-241 released to the atmosphere in 1973
Water	13 regional water sources within 75 km		Pu-238 average 40 fCi/liter Pu-239 average 140 fCi/liter	Pu analyses performed on water samples are highly suspect because of cross-contamination and/or effluent-contamination problem
	40 perimeter surface and ground water sites within 5 km		Pu-238 average 20 fCi/liter Pu-239 average 10 fCi/liter	
	Los Alamos water supply - 16 wells and 1 gallery.		Pu-238 average 20 fCi/liter Pu-239 average 20 fCi/liter	
	30 on-site surface and ground waters		Pu-238 range 0.01-8.2 pCi/liter Pu-239 range 0-10.1 pCi/liter Am-241 range 0.03-65.6 pCi/liter	
	Industrial wastes from 2 plants.	Composite of each week's effluent analyzed	Site TA-50-1 average. Pu-238 160 pCi/liter Pu-239 11 pCi/liter Am-241 25 pCi/liter Site TA-21-257 average Pu-238 40 pCi/liter Pu-239 30 pCi/liter Am-241: 20 pCi/liter	8.4 mCi released 0.6 mCi released 1.4 mCi released 0.2 mCi released 0.2 mCi released 0.1 mCi released
	Domestic wastes	Semiannually analysis of effluents from technical area and municipal sewage plants	Technical area average Pu-238 .08 ± .08 pCi/liter Pu-239 .09 ± .26 pCi/liter Municipal sewage average Pu-238 .046 ± .008 pCi/liter Pu-239 .03 ± .10 pCi/liter	
Soil and Sediment	Soil and sediment samples collected at same sites as regional water sources	Soil samples collected by taking 5 plugs, 75 mm diameter and 50 mm deep at the center and corners of a 10 meter square area and combined for a composite sample. Sediment samples collected behind boulders of flowing streams or 20 mm deep across the main channel of intermittent streams	Soil average (7 locations) Pu-238 8 ± 6 fCi/g Pu-239 10 ± 4 fCi/g Sediment average (9 locations) Pu-238 15 ± 2 fCi/g Pu-239 17 ± 4 fCi/g	
	Samples collected around the Laboratory and Los Alamos County		On-site soil & sediment range Pu-238 6-120 fCi/gm Pu-239 13-1300 fCi/gm Off-site soil & sediment range Pu-238 5-25 fCi/gm Pu-239 5-560 fCi/gm	
Special Studies	Sediment, vegetation, and rodents	Study of 3 canyons receiving waste from Los Alamos since 1943	Results reported for each sample individually	

TRINITY SITE, Alamagordo, NM (UCLA-406)
Report of Periodic Surveys, 1947 - 1956

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Soil:	Numerous samples collected in fallout area, up to 95 miles downwind (NE) of GZ. Samples collected at different times during the 10-year reporting period.	Nitric acid leach with chemical separation of plutonium from other alpha emitters. Extract counted with alpha scintillation counters.	Concentration of Pu in soil - dpm per gram and µgm per sq. ft. Up to several hundred nCi/m^2 in off-site areas. At 85-90 miles from GZ, values as high as 45 nCi/m^2 were found in 1950.	Serial samples at same locations did not always decrease with time - may be due to sampling variability.
		Some profile samples collected, mostly surface samples--1/2 or 1 inch depths.	Highest values were found on Chupadera Mesa,-- 30 miles NE of site.	
Vegetation:	Samples of grass, juniper, and pine at various locations.		dpm/gram: Results inconclusive -- probably surface contamination.	

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Air:	21 off-site sampling locations.	Continuous weekly high volume air samples of 40 ft^3/min thru Microsorban disk. Po-210 weekly analysis at 2 sites; monthly composite for Pu-238 at 21 sites.	Po-210 average: 1.2 fCi/m^3 Pu-238 average: 18.1 aCi/m^3	RCG for Po-210: 2 pCi/m^3 RCG for Pu-238: 20 fCi/m^3
	5 on-site sampling locations.	Continuous weekly high volume air samples of 40 ft^3/min thru Microsorban disk. Po-210 and Pu-238 analysis weekly	Po-210 average: 1.1 fCi/m^3 Pu-238 average: 534 aCi/m^3	RCG for Po-210: 7 pCi/m^3 RCG for Pu-238: 70 fCi/m^3 68 μCi of Po-210 and 84 μCi of Pu-238 were discharged to the atmosphere.
Water:	5 sites on the Great Miami River.	Samples collected 5 days per week and composited for monthly Pu-238.	Pu-238 average: 1.3 pCi/liter	RCG for Pu-238: 2 nCi/liter
	8 sites of ponds and streams.	Quarterly sampling and analysis for Pu-238.	Pu-238 average: 0.355 pCi/liter	
Foodstuffs and Vegetation:	Locally grown foodstuffs and vegetation sampling including milk, fruits and vegetables, grass and aquatic life.	Evaporate samples to dryness and analyze for Pu-238.	Pu-238 average: Milk: 0.9 fCi/gm Fruits and vegetables: 1.6 fCi/gm Grass: 7 fCi/gm Aquatic life: 4 fCi/gm	
Soil:	One location in each quadrant plus background location.	10 core samples (3.5" diameter by 12" deep) at each location and composited. Pu leached from samples.	Pu-238 average <4 miles: 1.17 nCi/m^2 >20 miles: 1.27 nCi/m^2	
Silt:	8 sites in ponds and streams (same as surface water).	Pu leached from samples.	Pu-238 average: 38 fCi/gm	

NEVADA TEST SITE, 1973 (NERC-LV-539-31)

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Water:	59 sampling sites from community water supplies, wells, springs, streams, lakes, and ponds.	Pu-238 and Pu-239 analyses annually on selected surface water samples.	Pu-238 range: <0.016-<.61 pCi/liter Pu-239 range: <0.012-<.74 pCi/liter	
	Long-term hydrological monitoring program at all active and inactive test areas.	NTS samples analyzed quarterly for Pu-238 and Pu-239.	Range of averages Pu-238: <0.014-<.083 pCi/liter Pu-239: <0.010-<.048 pCi/liter	
Air:	8 selected sites from the Air Surveillance Network.	Pu-239 analyzed on composite of 5 days sampling each month.	No results reported yet for 1973.	Previous results reported for 1965-1972.
Soil:	Part of continuing program started in 1970.	Ten 10 x 10 cm cores 5 cm deep were composited. Samples taken at the intersections of a 5 x 5 mile grid. Analyzed portion that passed thru a 10 mesh screen.	Results plotted as contours along with previous results.	

OAK RIDGE NATIONAL LABORATORY, 1973 (UCC-ND-280)

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Soil:	Nine locations near Perimeter Air Monitoring Stations.	Four samples, approx. 3" diameter and one centimeter thick, collected from a one-square-meter area and composited. Analyzed for plutonium (Note: Not clear whether includes 238 & 239 or just 239).	Plutonium (fCi/gm) Maximum 67 Minimum 12 Average 29	

PANTEX PLANT, 1973

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Water:	Samples collected monthly from one off-site lake and wells on-site.	Samples analyzed for gross alpha and beta, total uranium, plutonium and tritium.	Total plutonium (pCi/l) WS - 0.72 ground water - 0.38	Compared to RCG for uncontrolled area of 2 nCi/liter.
Soil and Vegetation:	Samples collected monthly at 25 off-site locations surrounding plant.	Soil - top 5 centimeters.. Vegetation - native grasses. Soil analyzed for total plutonium by radiometric analysis.	Total plutonium concentration pCi/gm dry soil Minimum 0 Maximum 1.39 Average 0.50 No results for vegetation.	
Air:	Air sampling network consisting of nine off-site continuous samplers surrounding plant. Samples changed weekly.	Samples analyzed for gross alpha and beta, total plutonium, and uranium content.	Plutonium (fCi/m^3) Minimum 0 Maximum 3.53 Average 1.26	Compared to RCG 20 fCi/m^3

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Air	11 air stations on site operated continuously	2 cfm through Gelman Type E glass fiber filters. Analyzed for alpha Jan-June and monthly composite analyzed for Pu July-Dec	Alpha average for Jan-June 6.9 ± 17 fCi/m^3 Pu average for July-Dec. 1 214 ± 99. fCi/m^3	Total alpha released Jan-June and Pu released July-Dec. from plutonium facilities was <77 .09 µCi.
	12 air stations off-site between 2 and 4 mile radius.	Continuous sampling through Delbag Microsorban filter media at 27 cfm, composited monthly	Pu average: <0 053 ± 53% fCi/m^3	RCG for Pu-239 is 20 fCi/m^3
	9 air stations in population centers around Rocky Flats.	Beginning in 3/73, continuously sampling through Gelman Type E glass fiber filters. Analyzed weekly for alpha. Beginning 7/73, samples composited monthly for Pu	Alpha average for Jan-June <2 6 ± 20. fCi/m^3 Pu average for July-Dec <0 264 ± 163% fCi/m^3	
Water	Effluent from 3 holding ponds	Sampled continuously and collected daily and composited for weekly analysis	Pond A-3 U + Pu average 5.61 ± 26% pCi/liter Pu average < 29 ± 37% pCi/liter Pond B-4 U + Pu average 16 58 ± 36% pCi/liter Pu average 7 37 ± 47% pCi/liter Am-241 average <1 79 ± 122% pCi/liter Pond C-1 U + Pu average 1.96 ± 35% pCi/liter Pu average <.18 ± 42% pCi/liter	RCG for soluble Pu = 1600 pCi/liter RCG for americium-241 = 1300 pCi/liter
	Walnut Creek (main effluent stream off-site)	Sampled continuously, collected daily, composited for weekly analysis	U + Pu average 11 43 ± 60% pCi/liter Pu average 3 11 ± 43% pCi/liter Am-241 average <1.31 ± 81% pCi/liter	
	2 reservoirs and 9 tap water locations around Rocky Flats.	Collected weekly and composited monthly	Reservoir U + Pu average 3.32 pCi/liter Pu average. <.06 pCi/liter Am-241 average: < 18 pCi/liter Tap Water U + Pu average <3.87 ± 54% pCi/liter Pu average < 07 ± 34% pCi/liter	
	Additional area reservoirs, lakes, and streams.	30 samples collected 9/73 out to about 20 miles.	U + Pu average 1 82 ± 51% pCi/liter Pu average.. < 31 ± 76% pCi/liter	
Soil.	Continuation of program	60 samples collected each 18 degrees of arc on circles of 1, 2 and 5 mile radius	Results displayed on map.	

Media	Sampling Program	Sampling Techniques and Analysis	Results Reported	Remarks
Air:	Weekly analysis of air filters collected at 8 monitoring stations near plant perimeter and ten stations around a 25-mile radius from plant. Four additional stations at Savannah, and Macon, Georgia and Columbia and Greenville, S.C. serve as background levels.	Beta activity, gamma measurements and gross alpha (uranium and plutonium) activity determinations for all samples.	Alpha activity Plant perimeter: 0.7 fCi/m^3 25-mile radius: .8 fCi/m^3 Distant sites: .9 fCi/m^3 Maximum reported value was 3.8 fCi/m^3 at plant perimeter.	Concentration Guide for alpha emitters is 20 fCi/m^3.
Vegetation:	Grass samples collected at seven locations along plant perimeter and at seven other locations along a 25-mile radius route.	Samples composited for monthly analysis.	Gamma emitting radionuclides were from fallout. Alpha emitters averaged 0.16 pCi/gm (dry wt) at plant perimeter and .12 pCi/gm (dry wt) at 25-mile radius.	
Food:	Farm produce representing four food categories collected at 14 localities.	Sixty samples analyzed by gamma spectrometry. Radiochemical analysis for ^{90}Sr and alpha emitters.	Alpha emitters in food stuffs averaged between 2-10 fCi/gm (wet wt). Maximum concentration of 240 fCi/gm (wet wt) in plums.	
Water:	Four sampling points on Savannah River. Fourteen samples of public water supplies.	River samples collected continuously and analyzed weekly. Public supplies sampled in April and October.	Drinking water alpha emitter concentration varied between nondetectable to 3 7 pCi/liter with an average of 0.8 pCi/liter.	The higher specific activity at Jackson 3.7 pCi/liter is attributed to thorium-228.
Soil:	Four sampling locations near plant perimeter, and 3 locations up to 100 miles.	Depth profiles at each site for ^{238}Pu and ^{239}Pu.	Total plutonium average in top 5 cm of soil is 15.2 fCi/gm at plant perimeter and 17 8 fCi/gm at distant locations.	
	Deposition-10 sampling locations at plant perimeter.	Core samples taken to 15 cm.	^{239}Pu at plant perimeter was 1.78 nCi/m^2 and 1.69 nCi/m^2 at distant locations.	