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Transuranic Solid Waste Management Research Programs

October-December 1973

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This report presents the status of the Transuranic Solid Waste Management Research and Development Programs at LASL. The previous reports in this series, unclassified, are:

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CONTENTS

I. Transuranic Waste Research and Development Program

A. Criteria Development 1

B. Corrosion Studies

1. Introduction 2

2. Laboratory Experiments 3

C. Radiolysis Studies

1. Introduction 5

2. Laboratory Experiments 6

D. Waste Sorting Studies

1. Introduction 7

2. Retrievable Wastes 9

3. Room Wastes 10

4. Process Relationships 10

5. Assay and Activity Levels 12

E. Survey of Solid Transuranic Wastes 12

F. Temperature Studies of ^{238}Pu Wastes 14

II. Transuranic-Contaminated Solid Waste Treatment Development Facility

A. Incinerator Selection 15

B. Design Capacity 15

C. Air Cleaning Design 15

D. Experimental Plan 15

E. Facility Design 16

F. Process Engineering 16

G. Assay and Accountability 16

III. Evaluation of Plutonium-Contaminated Radioactive Waste Disposal Areas with Respect to Their Potential Hazard and Eventual Disposition

A. Introduction 17

B. Risk Analysis Methodology 18

1. System 18

2. Hazards 18

3. Causal Events 19

4. Model 19

5. Mathematical Formulation 20

6. Illustrative Calculations for a Sample Risk Analysis of a Single Causal Event 22

C. Hydrogeologic Environment of Waste Disposal Areas at Los Alamos 22

D. Abiotic Transfer Processes 23

1. Migration by Self-Diffusion 23

2. Leaching, Transport, and Redeposition 24

E. Biological Transport Mechanisms

1. Introduction 26

2. Terrestrial Environment Transport Considerations 26

F. Radiological Assessment of the Intake of the Transuranic Elements

1. Introduction 33

2. Mathematical Equations Utilized in Evaluating the Radiation Dose from the Inhalation and Ingestion of Transuranic Elements 34

G. Radionuclide Inventory of the Waste Disposal Areas at LASL

1. Introduction 44

2. Location of Waste Disposal Areas 44

3. Radionuclide Inventory of Specific Burial Areas 45

APPENDIX: BIOLOGICAL PARAMETERS UTILIZED IN EVALUATING THE RADIATION DOSE EQUATIONS 51

TRANSURANIC SOLID WASTE MANAGEMENT RESEARCH PROGRAMS

PROGRESS REPORT FOR OCTOBER THROUGH DECEMBER 1973

ABSTRACT

The following report reviews the progress for three transuranic solid waste management research programs funded by the AEC Division of Waste Management and Transportation during the period of October-December 1973. The interim storage criteria, which were reviewed by AEC administrative and contractor personnel, will be revised and submitted to AEC Headquarters for further comment. The experimental conditions for the studies of corrosion in humid air have been revised to better simulate conditions expected in closed-pad storage. In addition to bare mild-steel and stainless-steel samples, mild-steel coupons coated with several rust-inhibiting compounds are being studied. Radiolysis studies of typical waste materials contaminated with ^{238}Pu have thus far produced between 0.56 and 41.01 kPa of gas per simulated year. Waste sorting studies are being used to evaluate the newly developed Multi-Energy Gamma Assay System (box counter). Incinerator type and design feed rate have been selected for the Transuranic-Contaminated Solid Waste Treatment Development Facility, and operational parameters selected. The risk analysis method to be used in the evaluation of past burial practices has been described. Several parameters have been evaluated from literature sources, and equations to estimate organ doses have been derived. A radionuclide inventory of Los Alamos Scientific Laboratory (LASL) disposal areas has been compiled.

I. TRANSURANIC WASTE RESEARCH AND DEVELOPMENT PROGRAM, A412 (R. MULKIN, K. A. PASHMAN, A. ZERWEKH)

A. Criteria Development

The third draft of the Criteria for the Interim Storage of AEC-Generated Solid Transuranic Waste was distributed through the Albuquerque Operations Office to AEC contractors and divisions in November. Comments were received from Idaho Operations, Richland Operations, Albuquerque Operations, Mound Laboratory, Savannah River, Oak Ridge, and the AEC Divisions of Operational Safety and Production and Materials Management. The comments will be evaluated and applicable ones incorporated into a final draft of the Criteria.

Additional work is required to make the definition of terms more specific. More concrete distinctions between the various waste classifications (solid transuranic waste, retrievable and nonretrievable solid transuranic waste, uncontaminated waste) were frequently requested. The definition of combustibility was criticized as being too general.

Significant comments were received for several of the criteria. The most frequently criticized requirement was the necessity of affixing a security seal to the final waste package. Several reviewers felt that this would be an unnecessary effort for

waste materials to be stored in a secured, government-owned area. Questions were also raised concerning the necessity for visual inspection of stored waste packages, the effectiveness of suggested means of implementing fire control criteria, and the practicality of maintaining an effective gas monitoring system. The most significant of the additional criteria suggested for inclusion in the document called for the exclusion of free liquids from the retrievable wastes.

Following evaluation of the comments, selected alterations and additions will be tentatively incorporated into the Criteria. These tentative changes will then be reviewed with AEC Headquarters Waste Management personnel before incorporation into a final draft and issuance of the document.

B. Corrosion Studies

1. Introduction. The transuranic waste storage facilities presently in use at AEC installations are closed after being filled, establishing an environment which at some installations will be characterized by high relative humidities (approaching 90%) and moderate temperatures (around 10-16°C, or about the equivalent of the average annual air temperature for the area). The high humidities in these enclosed storage facilities result from equilibration of a confined volume of air with surrounding moisture-containing soil. Soil containing moisture at values as low as 5-10% of saturation will produce a relative humidity in excess of 90% in a volume of confined air. The relative humidity in the proposed Los Alamos facility, which would be entirely below ground level, is expected to be 90-95%, while that in the facilities at the Atlantic Richfield Hanford Company (ARHCO) and the National Reactor Testing Station (NRTS) may be lower due to the above-ground or partially above-ground construction at these sites, which results in better air movement and soil aeration.

High-humidity environments produce a thin film of moisture on all materials in the storage facility. However, free water should not collect within the storage facility; instead, an equilibrium condition will be maintained between the high-humidity air and the soil moisture.

Temperatures within the facility will vary on a seasonal basis. Annual variations in atmospheric temperature are damped out by soil cover and rarely penetrate more than 1-2 meters. Below that depth soil temperatures will approximate the average annual air temperature. Thus, a below-ground storage facility at the Los Alamos Scientific Laboratory (LASL) covered with 1-2 meters of soil should remain at a fairly constant temperature of 10-12°C. Seasonal variation of this temperature should be no more than a few degrees. At ARHCO and the NRTS the seasonal variation of temperature within the facility will likely be a few degrees greater, but no significant daily temperature variations will occur in these facilities if the earth cover is 1 m or more in thickness (in the absence of free air movement).

Experience gained from observations on the condition of mild-steel waste drums following retrieval from actual burial and drums stored in above-ground locations pending disposal has demonstrated the susceptibility of these containers to breaching from corrosion mechanisms. The containers examined had been buried for up to 18 years in direct contact with soil or stored in above-ground locations up to five years. In view of this susceptibility, a series of laboratory-scale corrosion experiments were started for the following purposes:

a. Determining the basic corrosion mechanisms
b. Determining the waste matrix conditions which accelerate corrosion internal to the drum (Current investigations are discussed later under Section C and D.)
c. Determining the environmental conditions of storage which accelerate corrosion of drum exterior

d. Optimizing environmental storage conditions and protective coatings for the drum to determine if a 20-year life without loss of containment is practical

e. Recommending alternate storage container materials as applicable.

2. Laboratory Experiments. Experiments were conducted to simulate the burial of mild-steel drums in soil from the LASL. Coupons from the mild-steel drum were placed in LASL soils containing water to 10, 50, and 100% of their moisture holding capacities. These studies were terminated at the end of one year. The final samples have been microphotographed and prepared for examination by the electron microprobe. When examination and evaluation of the data from this work is completed, a final report will be prepared.

Comparative studies on stainless-steel coupons are continuing. Insufficient time has elapsed to make a meaningful comparison; but based on data taken from a one-month exposure of these samples to LASL soil saturated to 100% of its moisture-holding capacity, Austenitic-stainless steels show considerably less corrosion than mild steel under the same conditions. If this trend continues, a cost study will be undertaken to determine whether the initially higher price of a stainless-steel container compared with that of a mild-steel container with a 2.29-mm, 50% cross-linked, high-density polyethylene liner is offset by elimination of possible future repackaging or special handling charges.

In order to study the corrosion effects of humid air, drum coupons were exposed to atmospheres of 50% and 100% relative humidity. The experimental setup in use, however, appeared to produce physical inconsistencies with actual conditions. Therefore, to better simulate the conditions found in actual storage environments and eliminate several of the observed experimental problems, the corrosion studies in humid air have been redesigned. The high-humidity environment is established in a truncated 1-liter beaker that has been sealed with a tight-fitting glass lid and stopcock grease. A fine-mesh copper screen cylinder (100-mm-high x 65-mm-diam) holds soil from Mesita del Buey along the sides of the beaker. The soil has been moistened to 25% of its water-holding capacity. The moisture content is being maintained at the established level by adding water when a weight loss indicates loss of water from the soil due to evaporation. An absorbant material ("diaper paper") has been placed in the bottom of the beaker to absorb any free water. The coupon is suspended by a fine wire from the beaker cover to prevent contact with any other surface. The improved experimental system has been diagrammed in Fig. 1. Within two hours

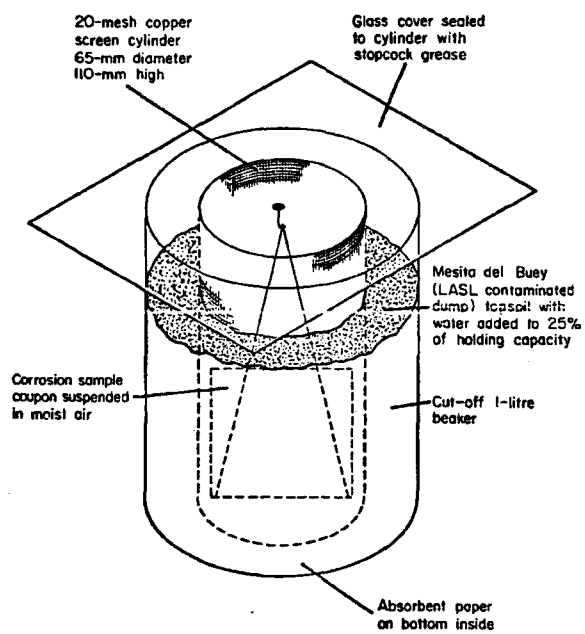

Fig. 1. Sample Container Simulating Covered Pad Storage.

after assemblage, the measured relative humidity of the air in the center void was a constant 98%. The temperature should remain nearly constant at the ambient level (20°C). This temperature, which is higher than that expected in the storage facility, should provide an upper limit for temperature effects since the corrosion rates for mild steel are greater at the higher temperature.

The selection of coupons to be studied at both 50% and 100% relative humidity has been expanded to include a number of protective coatings. The mild-steel coupons now being tested include unprotected samples and sections treated with the following coatings:

a. Komac "Q. C." White Enamel, a baked-on gloss enamel which is currently in use on the waste drums being purchased by LASL
b. Rust Gard, a proprietary rust-preventative coating which may be applied by cloth, spray, dip, or brush. The penetrating film provided is carried in a volatile, flammable solvent.
c. Rustoleum Industrial Coatings (Both of the systems listed below are available in a variety of colors, including black and white.)
   (1) The acid resistant system, a heavy-duty solvent-based vinyl system consisting of a wash primer followed by two coats of acid-resistant finish coat
   (2) The chemical and water-resistant system, a polyamide epoxy system consisting of a "zinc-sele" green primer, an orange primer, and a finish coat
d. Zinc Chromate Underseal, a commonly used rust-resistant primer coating. It is a spray application in approximately 86% hydrocarbon propellant.
e. Hot-dipped zinc, a standard galvanizing process for the protection of ferrous metals against corrosion. These sample coupons were taken from a mild-steel galvanized garbage can having the same wall thickness as the mild-steel waste drums.
f. Bituminous coatings
   (1) Petroleum-based asphalt of the type commonly used for road work. It has excellent penetration and adhesion qualities and has low susceptibility to shock and changes in temperature.
   (2) Koppers Protective Coating Bitumastic No. 300-M, a coal-tar, chemically-cured epoxy designed for the interior and exterior protection of storage tanks. It resists continuous moist heat conditions up to 49°C (120°F) or dry heat to 93°C (200°F). It is expected to withstand considerable physical abuse, including direct impact, abrasion, and flexing, and will not sag or flow at any temperature.

Stainless-steel coupons are also being tested in a comparison study similar to that undertaken for the soil corrosion studies. The number of coupons for each test has been reduced to three, taken from areas on the drum which show the highest rates of corrosion, that is, the side, rib, and top (or bottom). Monthly examination of the coupons, using photomicrographic techniques, will permit selection of samples to be examined more thoroughly with the more sensitive electron microprobe. The data thus obtained will yield relative rates of corrosion (in depth per unit time) for the various samples and permit comparison of corrosion-inhibiting abilities of the coatings being tested.

C. Radiolysis Studies

1. Introduction. A major experimental effort is presently being directed toward determining the effects and the magnitude of the problems created by alpha radiolysis of waste materials. Most radiolysis experiments to date have involved studies of the effects of gamma and beta radiations. Accurate data are required, however, to establish the effects of alpha particles from transuranic radionuclides in typical waste materials.

Radiolytic degradation of several of the materials comprising transuranic-contaminated wastes has been reported to produce various gases.[1] This gas formation suggested the need for an experimental program to determine the extent of the problems arising from the presence of the gases. Among the major concerns are the following:

a. Pressurization of the waste container
b. Increased combustibility hazard from flammable gases in the storage environment
c. Effects of corrosive gases on the waste container
d. Interactions among radiolytic products from different waste types
e. Identification of the products from waste types not previously studied.

If the wastes are to be stored in tightly-sealed containers, an experimental study of the radiolytic production of gases will assist in determining whether pressurization and eventual breaching of the containers can occur. Theoretical calculations for gas production from combustible wastes indicate that pressurization is a possibility. Rough calculations to determine upper limits for rates of gas production have been performed using the following assumptions: 32 kg of the waste material under consideration in a 210-liter drum, contamination level of 2×10^{-4} g of ^{239}Pu per gram of waste material, and complete absorption of the energy of the alpha particles by the waste material. These calculations yield rates of gas production of approximately four liters of H_2 per year from polyethylene, 12 liters of HCl per year from polyvinyl chloride, and three liters of gas (H_2, H_2O, CO, CO_2, hydrocarbons) per year from cellulosic materials. At these gas generation rates, accumulations of between 60 and 240 liters of gas in a 210-liter drum and pressures of 30-140 kPa (5-20 psi) above atmospheric pressure (at STP) are conceivable in 20 years. However, the actual quantities of gas produced in the waste containers will be influenced (and probably reduced) by such factors as the actual amount of energy deposited in the waste material by the alpha particle and the components of the waste matrix (various combinations of the materials considered above). Experimental studies are necessary to determine the effects of these factors and establish design parameters to minimize these effects as practical.

If the waste containers are not to be tightly sealed or are deliberately vented to prevent possible pressurization, it will be necessary to determine the extent of the problem created by the possible accumulation of flammable gases within the storage facility. For example, hydrogen, which results from radiolysis of polyethylene, cellulosics, and perhaps sludges, has a lower explosive limit of 4% concentration in air. In addition, flammable hydrocarbon gases are expected from the radiolysis of organic materials.

Corrosive products of radiolysis have the potential for reduction of the containment ability of the waste container. Thus, experimental studies are being designed to determine whether HCl from polyvinyl chloride and water from various waste materials are produced in sufficient quantities to result in an increased rate of corrosion and eventual breaching of the container. Other studies will ascertain whether two or more products, such as two gases, will

combine and interact unfavorably to create a toxic, corrosive, or explosive mixture within the container or the storage facility (for example, H_2 and O_2 from radiolysis of H_2O).

Little is known about the radiolytic products or the problems created by other types of wastes, such as organic solvents, oils and greases, and liquid waste treatment sludges. The organics will undoubtedly yield flammable gases, but the extent of alpha degradation is uncertain. Motor oils subjected to neutron and gamma radiation are reported to yield 49-127 liters of gas/liter of oil at 10^6 J/kg (10^8 rad),[2] but very little is known about the properties or extent of radiolysis of oils absorbed on solids such as vermiculite or on cellulosic materials.

The radiolysis of sludges is expected to be similar to that of water (which comprises about 60% of the sludge), the major reaction being the production of hydrogen and oxygen gases. The experimental program is expected to determine the possibility of obtaining an explosive mixture as well as the extent of the pressurization problem. The influence upon reaction rate of several factors about which little is known (for example, the alkaline pH of the sludges and the effect of dissolved salts and ions on the rate of gas production) must be determined. The potentially high gas yield and the relative lack of free space in the container present a greater potential for breaching of a container with this waste type.

One other common contaminant of uncertain behavior is nitric acid and/or nitrate salts absorbed on combustibles. Nitric acid causes the breakdown of cellulosics and the release of flammable gases. In addition, alpha particles induce the production of oxygen and nitrate ions.[3] Future experimental studies will indicate whether elimination of nitric acid and nitrate salts will be necessary to remove any potential problem.

2. Laboratory Experiments. The experimental program to study radiolysis effects concerns itself basically with the following areas:

a. The rates of gas production under various conditions
b. The identification of radiolysis products and determination of their relative concentrations
c. The effects of the radiolysis products on the waste materials and on the waste containers.

Small stainless-steel cylinders (300 ml) equipped with 103.4-kPa (15 psi) gauges, needle valves, and 1/4" swagelock connectors to simplify sampling of the accumulated gases (see Fig. 2) have been loaded with samples of several materials found in the transuranic waste stream. The waste materials presently under study include the following:

a. A cellulosics mixture consisting of equal weight percents of paper wipes, paper tissues, a table towel of embossed paper with a polyethylene backing (commonly known as "diaper paper"), cheesecloth, and cotton material from a laboratory smock

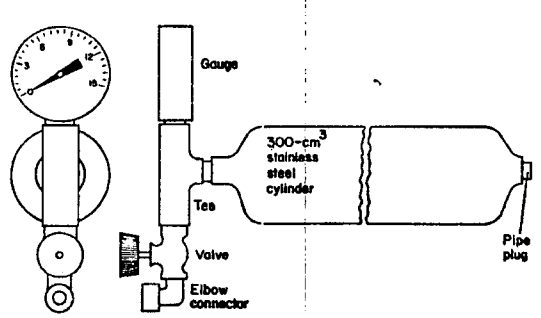

Fig. 2. Experimental Container for Radiolysis Samples.

b. Hypalon drybox glove material
c. Surgeons' gloves (isoprene)
d. Polyethylene bags
e. Tygon tubing
f. Neoprene drybox gloves
g. Lucite sheet plastic.

Other materials which will be added to the experiment include lead-loaded drybox gloves and chemical sludge from the liquid waste treatment plant.

All of the samples have been cut into pieces, spread out in a drybox, and 52.5 g of each contaminated as uniformly as possible with three different quantities of ^{238}Pu as the nitrate. These quantities are 62 mg, 31 mg, and 6.2 mg. The concentrations selected represent a range expected to be present on process-generated wastes going to interim storage. The alpha activity factor was multiplied by 40 in each case by using 80% ^{238}Pu to simulate 20 years of ^{239}Pu exposure in six months.

Data are being obtained by several techniques. Pressure buildup in the cylinders is being monitored, and gas samples are periodically withdrawn from cylinders showing rapid pressurization. The gas samples will be analyzed using mass spectrometry. To date, 23 tests have been started. The cylinder containing the cellulosics mixture with the highest level of contamination (No. 1) was the first to register pressure, 3.5 kPa in the time equivalent of 40 days. In the time equivalent of 2.63 years the pressure had increased to 110 kPa (full scale on the gauge). The gas mixture was sampled, reducing the pressure to 7 kPa, and the sample was submitted to mass spectrographic analysis. The pressure in this cylinder has continued to increase at the same rate, making a total of approximately 190 kPa in the time equivalent of 4.7 years. The lack of pressure buildup in the container of cellulosics contaminated with the middle strength solution (no pressurization at the time equivalent of 4.7 years) introduced the possibility that cylinder No. 1 was an anomaly. A duplicate sample, prepared to check the possibility, showed 3.5 kPa pressure in the equivalent of 40 days. The pressure increased at essentially the same rate as did that in cylinder No. 1, requiring sampling at the time equivalent of 2.74 years. The ten cylinders which have shown measurable pressure increases have been identified in Table I.

At the end of the six-month exposure period, the solid and liquid contents of the cylinders will be examined to identify any degradation products. When the degradation products have been determined and the components of the waste matrix identified and analyzed, the probability of occurrence and resultant consequences of possibly detrimental interactions can be ascertained. Although it is still too early in the experiment to attempt to interpret the data, the formation of gas from radiolytic attack of several of the waste matrices appears to present a significant problem for the proper management of retrievable waste storage facilities.

D. Waste Sorting Studies

1. Introduction. Waste characterization studies are an important part of several of the waste management research and development programs at LASL. Detailed information on wastes generated and sent to retrievable storage is required by both operational personnel and the research staff. Waste management personnel at LASL have identified the need for a more detailed description of the wastes being received in order to optimize waste packaging, handling techniques, and storage facility conditions. The design and use of the new Transuranic Contaminated Waste Treatment Development Facility (TDF) require information concerning the actual amount of combustibles in the waste and the levels of contamination of these materials. A breakdown of combustibles into categories such as cellulosics, plastics (polyvinyl chloride separately), and rubber is also needed for design or selection of feed,

TABLE I

PRESSURES DEVELOPED IN TEST CYLINDERS

Cylinder No.	Contamination Level[a]	Contents	Time to First Readable Pressure[c] (Simulated Yr)	Rate of Pressure Increase (kPa per (Simulated Yr)
1	S	Cellulosics Mixture[b]	0.11	41.01
5	M	Hypalon (drybox glove material)	6.16	0.56
7	S	Isoprene (surgeons' glove material)	3.87	0.89
8	M	Isoprene	1.02	3.37
10	S	Polyethylene (bag material)	0.44	16.27
11	M	Polyethylene	0.24	13.79
19	S	Lucite (polymerized methyl methacrylate resin)	0.27	12.84
20	M	Lucite	0.98	3.53
22	S	Cellulosics Mixture	0.11	40.44
23	S	Cellulosics Mixture	0.11	33.27

[a]S = Strong (62 mg Pu/52.5 g matrix)

M = Middle (31 mg Pu/52.2 g matrix)

[b]Kleenex, diaper paper, cotton laboratory smock material, cheesecloth, Kimwipes, paper towels

[c]First readable pressure = 3.5 kPa (0.5 psi)

combustion, and offgas cleaning equipment.

A program to sort, identify, and assay actual wastes produced at plutonium-handling facilities is serving as a means of obtaining quantitative information on waste characterization. Such quantitative data, combined with the qualitative information obtained from questionnaires and personal interviews (identifying waste types, specific properties, chemicals, handling and storage procedures), serve as input to the research and development experimental programs. Experiments to determine the extent and products of radiolytic degradation of wastes and problems of internal corrosion from chemicals in the waste require identification of commonly occurring components of the waste matrix. The information will also be used to determine the ability of any chemical component to complex the transuranic contaminant and increase its mobility through the environment.

A waste sorting experiment has been instituted at LASL. The sorting procedure consists of introduction of waste materials into a hood, assay of small packages of waste to determine the activity level, separation of the assayed material into non-retrievable (<10 nCi/g, handled in a hood) or retrievable (>10 nCi/g, handled in a glovebox) streams, inspection and

identification of the matrix components, and removal from the sorting line for disposal. Both process-generated and room wastes from the plutonium metal fabrication building and process wastes from other Laboratory sites are being examined. These studies are providing the following types of information:

a. An accurate breakdown of both room and process-generated wastes into combustibles and noncombustibles. Materials within these groups are classified as cellulosics, plastics, rubber, glass, metals, and others. Quantities of materials in these different waste subgroupings will be used in risk analyses of retrievable storage facilities (in particular, the amounts of different combustibles present) and in determining the heat content of materials to be incinerated in the waste treatment facility.

b. The average activity or contamination level of the different waste subgroups. These data will be useful in determining the magnitude of the problem of radiolytic gas production in different waste matrices, especially for the more highly contaminated materials found in process-generated wastes. In addition, the distribution of activity among the various waste types serves to identify categories which contain the majority of the contamination. This information will serve as a basis for waste sorting at the point of origin of the waste, thus possibly reducing retrievable waste volumes, which require more costly handling and storage.

c. Identification of waste types and volumes as a function of origin. Data obtained are being used to identify sources of particular waste types and to rate waste management practices. Waste management personnel may utilize such information to analyze their practices for increased efficiency and decreased waste volumes, contamination levels, and costs. (Such a program of detailed waste characterization, undertaken by Rocky Flats in 1970, was reported to have resulted in a significant decrease in the volumes of operational waste through implementation of improved waste management practices.[4])

d. A distribution of activity per waste package as a function of origin. At LASL, room waste is administratively designated as being contaminated below the 10 nCi/g level, process-generated waste above 10 nCi/g. However, prior to the development at LASL of the pancake counter for assay of transuranic wastes at the 10 nCi/g level, there was no practical way to verify these administrative designations. The information obtained from the sorting and assay of both the retrievable and nonretrievable waste streams may be useful in determining the impact of the 10 nCi/g level, now used to distinguish retrievable and nonretrievable wastes, on the volumes of waste generated. Analysis of the effects of changes in this limit on the amounts of transuranic material going into the disposal or storage environment and on the costs of disposal versus storage will also be available.

2. Retrievable Wastes. The waste sorting experiment has been in progress for six months. The composition of retrievable wastes from routine plutonium-handling operations being studied at

Los Alamos during the quarter is given in Table II. The volume percent is based on uncompacted waste packaged in 115-liter drums. The waste materials sorted averaged 51 wt% noncombustibles, 49 wt% combustibles. The plastic that was accumulated during the sorting operation consisted mainly of plastic bags from bag-out operations. Less than 10% of the plastic consisted of items such as tubing, laboratory plastic-ware, and metallographic mounts. The volume of rags and cheesecloth generated in the processing area is small compared with the volume of plastic. Most of the cellulosic wastes are incinerated for recovery of the plutonium. This waste stream is small enough for one recovery incinerator, burning a maximum of 25 liters (approximately 2 kg) per day, to handle easily. The resulting leached ash heels, or process solids, comprise less than 1 wt% of the retrievable waste stream. The weight of rags that go directly to retrievable storage is less than 5% of the total cellulosic wastes generated. The packing density of metal and glass wastes was quite low in the absence of crushing and compaction. Potential volume reduction methods for these waste types include flattening metal cans used to package and transport materials, such as ash and graphite (resulting in a volume reduction factor of approximately five), and crushing glass containers used to transfer liquids (requiring a solution to the safety problem of shattered glass).

3. Room Wastes. Room wastes from the plutonium chemistry and metallurgy areas are collected in polyethylene bags in cardboard boxes placed inside 115-liter drums having flame-retardant lids. When a container appears to be full, the bag is taped shut, the box is removed from the drum, and the box lid is taped closed. The closed boxes are checked for "measurable" quantities of plutonium in a neutron counter before release to radioactive waste disposal. Since hand compaction of the potentially contaminated material is not permitted at LASL, the density of room wastes is generally quite low. Records of weights, source by area, and other descriptive information were maintained on these boxes during the quarter. Amounts of waste generated and average densities of the material are listed in Table III.

The volume of the room-generated waste stream will be reduced substantially when the presently approved LASL unpackaging area is completed. Most items such as surgeons' gloves and drybox gloves can then be issued directly with all packaging material remaining outside the process area. For example, the uncompacted boxes and tissue paper from one case of surgeons' gloves generate one box of room waste. Since the average rate of use is 18-24 cases of gloves per month, this waste stream could be reduced 10% by unpackaging this one item outside the process area. Still more room trash could be eliminated by having offices outside the process area. Charts, computer runs, and other office wastes make up at least 10% of the present room trash volume.

4. Process Relationships. Initial investigations to relate amounts and types of wastes generated to specific processes have begun. Ash leaching, ion exchange, and metal alloy processing have been studied; the composition of the retrievable solid waste stream from the ash leaching process is listed in Table IV. This process involves leaching off-site incinerator ash three times with nitric acid and calcium fluoride. The metal shipping containers comprised most of the metal in this waste stream. Most of the glass resulted from a routine change-out of the dissolver pots, an activity which occurs infrequently. A survey of an ash leaching operation handling LASL incinerator ash, which is generated in an adjacent glovebox, would show almost no scrap metal waste, since all feed would be delivered in reusable containers. Information obtained by this type of study, in combination with questionnaire-interview data, will result in a

TABLE II

COMPOSITION OF RETRIEVABLE WASTE STREAM

OCTOBER - DECEMBER 1973

Material Type	Composition: Average Weight Percent	Composition: Typical Volume Percent
Metal	28	22
Plastic	29	32
Rubber	8	10
Cellulosics	12	26
Glass	22	10
Process Solids	1	-

TABLE III

GENERATION RATES AND DENSITIES OF ROOM WASTES

OCTOBER - DECEMBER 1973

	October	November	December	Average
Boxes Generated	218	128	180	175
Volume (m^3)	14.8	8.9	12.2	12.0
Weight (kg)	1 262	749	1 038	1 016
Density (kg/m^3)	85	84	85	85[a]

[a]Equivalent to 5.3 lb/ft^3

TABLE IV

COMPOSITION OF RETRIEVABLE WASTE STREAM

FROM

ASH LEACHING OPERATIONS

Material Type	Composition, Wt%
Metal	23
Plastic	14
Rubber	7
Cellulosics	11
Glass	14
Process Solids	31

useful summary of retrievable wastes being generated by various AEC contractors. Such information might then be used to modify processes for waste minimization.

5. Assay and Activity Levels.

a. Pancake Counter (Low-Energy Assay System). At Los Alamos, the plutonium fabrication and recovery areas operate under administrative control which designates process-generated wastes as retrievable and room-generated wastes as nonretrievable (unless recoverable or retrievable quantities of contamination are detected by a neutron counter). Various aspects of the sorting studies have demonstrated that, with a few easily corrected exceptions, this designation is generally an adequate means of segregating retrievable and nonretrievable wastes.

To establish relative activity levels of wastes removed from typical glovebox operations, several items with known histories were studied. A glovebox glove, which had been cleaned by wiping thoroughly with wet cheesecloth, was assayed in the pancake counter. The measured activity of the package was over 1000 nCi/g. In a second study, several one-liter ice cream cartons full of dry chemicals were introduced into the bulk fusion operation. The chemicals were transferred to mixing jars, and the empty cartons transferred to the nearest bag-out station. Contact with plutonium-contaminated surfaces occurred only when the containers were set on the glovebox floor and when they were handled by three or four glovebox gloves during the transfer operation. Nevertheless, assay of the bag-out package showed a plutonium content of 700 nCi/g. This type of study confirms the administrative designation that process-generated wastes are above the 10-nCi/g level.

Room or area waste represents the least contaminated waste stream in plutonium-handling areas. During the quarter, approximately 100 boxes of routinely generated room waste were opened, sorted, and measured in the pancake counter. No box of regular room waste was found to contain more than 10 nCi/g. Although no box of regular room waste assayed above the 10-nCi/g level, a few items resulting from maintenance operations (such as valve changes) were more highly contaminated. Operating procedures have been changed to assure that such material is no longer placed into containers of room waste.

b. Multi-Energy Gamma Assay System (MEGAS). The waste sorting system has been used to aid in evaluation of the newly developed box counter, or Multi-Energy Gamma Assay System (MEGAS). Four boxes of waste from the plutonium processing facility were first assayed in the MEGAS (results in Table V). They were then sorted, repackaged into smaller packages, and assayed in the pancake counter. A comparison of the levels determined by the two assay systems is given in Table VI. The discrepancy between the two results obtained for Box 10 is due to the inherent limitation of the pancake counter to assay plutonium contamination in the presence of dense materials. These materials, consisting of metal items and wet cheesecloth, resulted from a maintenance job involving changing a contaminated valve. The materials in Box 1 consisted of several types of waste from glovebox operations involved in processing scrap from the analytical laboratories, salts from the electrorefining of plutonium metal, and $NaF-K_2S_2O_7$ fusion of residues. Boxes 31 and 34 contained typical room waste with little or no contamination. This round-robin assay program will continue in order to further determine the accuracy of the new MEGAS counter.

E. Survey of Solid Transuranic Wastes

As a supplement to waste sorting studies, a waste survey questionnaire has been developed to provide information for the waste characterization program. A previous survey, which encompassed the major AEC waste-generation sites, provided general

TABLE V

ASSAY OF ROOM WASTES IN MEGAS COUNTER

Box No.	Mass, kg	Pu, mg	Am, μg	Total, nCi/g
10[a]	12.1	403.	11.	2 400.
1[a]	6.91	133.	88.	1 400.
31[b]	5.81	0.14	0.02	<2.
34[b]	8.14	0.04	0.01	<0.4

[a]Error ± 30%

[b]Error ± 50%

TABLE VI

COMPARISON OF ASSAY RESULTS OF PANCAKE AND MEGAS COUNTERS

Box No.	MEGAS Counter, nCi/g	Pancake Counter, nCi/g
10	2 400	80
1	1 400	1 183
31	2	0.4
34	0.4	Background

information on the relative composition of the waste streams, on estimates of overall volumes produced, and on various aspects of the waste-handling procedure.[5] The need for more detailed information on the composition of the wastes, to be used in both chemical characterization and physical interaction studies, has led to the development of a second survey questionnaire. The new questionnaire stresses the relationship between the process and the waste generated. The information sought will be useful not only for present studies, but also for future attempts to reduce waste volumes through optimization of the processes producing the wastes.

Three possible methods of employing the questionnaire are presently under consideration. One method which has proven to be satisfactory is the "personal interview" with the operator. The interview system has been tested with two operators from the plutonium processing facility at Los Alamos. The discussions, which required 1 to 1-1/2 hours, supplied most of the answers desired in appreciable detail. Any information found to be lacking when the interview was summarized was obtained from a short telephone inquiry.

A minor modification in the wording presented in the introduction of the questionnaire permitted the testing of another survey procedure. The modified questionnaire was distributed to five plutonium processing operators to be answered and returned with no interview being conducted. This procedure produced much less satisfactory responses in a significantly longer time period (one to two weeks.) Obtaining the remaining information would require an interview similar to that discussed above.

The results of the distribution system suggest the third possible method of utilizing the questionnaire, a combination distribution-interview procedure. The

questionnaire could be distributed to the operators involved about a week before a planned interview. This procedure would permit time to obtain the answers to the questions, thus perhaps shortening the interview time required. At the same time, the personal interview should minimize both the information omitted and the need for further discussions. Evaluation of the three systems has begun, and a decision on the method of choice for use both at LASL and at other AEC installations will be forthcoming.

F. Temperature Studies of ^{238}Pu Wastes

Plutonium-238 self-heating was measured for containers of contaminated wastes. The wastes, packaged in small plastic bags, had been removed from larger containers of ^{238}Pu contaminated materials in temporary storage at LASL. These packages were placed in small containers, which were then clustered in an 8-liter tin-plated can surrounded by vermiculite in a 115-liter drum. Thermocouples were positioned at several locations, including the center of the arrangement of small containers (see Fig. 3).

The maximum temperatures attained by the package (thermocouple 1) are plotted against the grams of plutonium in the package in Fig. 4. The data obtained indicate that temperatures above 80°C may occur in containers of wastes contaminated with ^{238}Pu being placed into retrievable storage. Preliminary information suggests that because of possible thermal degradation effects on various components of the waste matrix (for example, polyvinyl chloride), storage of more than 20 g of ^{238}Pu in one or a few containers insulated by vermiculite may create special containment problems in retrievable storage.

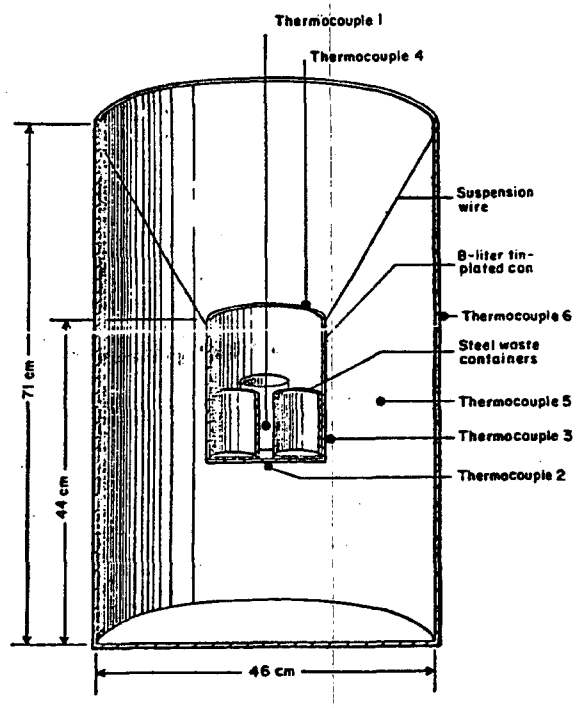

Fig. 3. Experimental Arrangement for Measuring Self-Heating of ^{238}Pu Wastes.

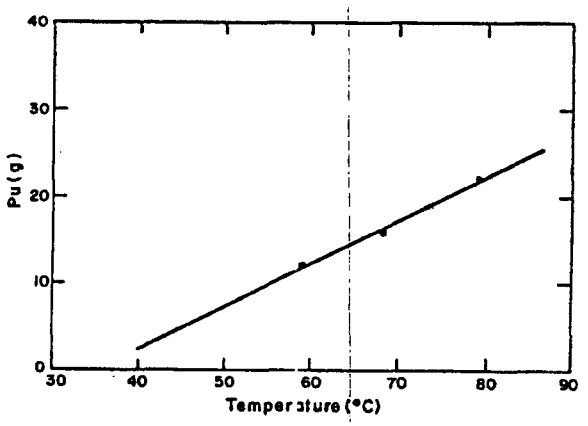

Fig. 4. Maximum Temperatures Attained in ^{238}Pu-Contaminated Wastes in a Storage Unit.

II. TRANSURANIC-CONTAMINATED SOLID WASTE TREATMENT DEVELOPMENT FACILITY, A413 (J. E. HERCEG, S. L. LAMBERT, M. E. MCLAIN, C. J. UMBARGER)

A. Incinerator Selection

The Task Force organized in the first quarter of FY74 to assemble available data on the various types of incinerators used or being considered for the processing of transuranic-contaminated wastes and to review the choice of incinerator type as the candidate process for the Transuranic-Contaminated Solid Waste Treatment Development Facility (TDF) completed its assignment. The information from this effort is presently being edited into a document which will be of use by others considering incineration as a waste treatment process.

Data gathered for this study resulted in narrowing the choice of incinerator types available for the TDF. Most incinerators were eliminated because of moving internal parts, sealing problems, or special design features which make equipment scale-up rather difficult. The two remaining candidates included the British Windscale design utilizing a low temperature tubular combustion chamber and the standard dual chamber controlled-air type incinerator. The Windscale unit was ruled out because of the inherent difficulty of scaling up to a larger, practical size and the uncertainties of corrosion problems. As a result, the controlled-air incinerator was reconfirmed as the candidate process for the TDF on the basis of greater operational flexibility, ease of combustion rate control, minimum particulate emission due to low turbulence in the primary combustion chamber, combustion efficiency, and availability of commercial equipment.

B. Design Capacity

Based upon a detailed evaluation, the design feed rate for the candidate process was reduced from 90 kg/hr to 45 kg/hr. This reduced rate is appropriate for reasonable facility scale-up from this rate, anticipated practical waste processing requirements, and required operational costs.

C. Air Cleaning Design

Although a stated purpose of the TDF is to explore new developments in air cleaning technology, it was felt that the initial installation of a potentially inadequate developmental air cleaning system with sufficient operational latitude would be detrimental to the incinerator evaluation and study. Therefore, the initial air cleaning train design will rely on the proven techniques of dry cyclones and venturi scrubbers following spray cooling of the hot flue gas and finally, HEPA filters. Data has been obtained which indicates that the mean particle size in the flue gas from a controlled-air incinerator burning paper should be on the order of 0.9 μm with a total particulate loading of less than 0.2 g per standard cubic meter (0.1 grain per scf).[6]

An optimized air cleaning train for the incinerator will be developed as part of the experimental program once the incineration process is understood and emissions from the incinerator established.

D. Experimental Plan

A preliminary experimental plan was prepared as a guide for process instrumentation design. Operational parameters to be monitored in the incinerator operation will include:

1. Composition of feed material, gross heating value, and moisture content
2. Flow rates, pressures, and temperatures of internal incinerator gases
3. Composition of incinerator flue gas as determined by analyses for CO_2, O_2, CO, HCl, NO_x and unburned hydrocarbons
4. Particulate loading, particle-size distribution, and chemical composition of the particulate entrained in the flue gas
5. Chemical composition, volume,

and density of ash and the relationship to initial waste volume and density

6. Efficiency of air cleaning components as measured by chemical and particulate analyses of flue gases at each stage of the air cleaning train
7. Distribution of radioactivity in the process components when transuranic-contaminated wastes are burned
8. Presence in the ash and other effluents of elements characteristic of materials of the process vessel construction as an indication of excessive corrosion rates.

Other measurements of the operating process will be made as will be determined by identified needs and feasibility.

E. Facility Design

The design criteria for the TDF were reviewed and updated to take into consideration the changes resulting from the 45 kg/hr design feed rate and to assure, based upon currently available information, proper sizing of the facility to accommodate the unit operations proposed for the TDF. The final design criteria were issued by LASL as report number ENG-9-Rp-65, dated November 30, 1973. At this time facility design was ready to be turned over to an outside architect and engineering firm for Title I and Title II engineering pending release of funds.

Initial funds for the Title I design were released December 26, 1973, with the stipulation that the facility design be reviewed to assure that maximum research and development flexibility be incorporated into the design of the TDF. This review was initiated by LASL at the end of December 1973.

Preparation of the Preliminary Safety Analysis Report (PSAR) for the TDF was initiated in December.

F. Process Engineering

The unit operations for the candidate process were reviewed in detail to assure that adequate space requirements, necessary utilities, and proper material flow through the TDF were provided in the design. Conveyance of waste materials to the head end of the sorting line was simplified by eliminating the need for drum conveyors and substituting a battery-powered forklift.

Because of the uncertainties in adapting commercially available shredders to assure the containment required for plutonium handling and to reduce maintenance problems in a plutonium-contaminated environment, a decision was made to purchase a shredder and associated materials handling equipment to experimentally resolve these design problems prior to completion of the TDF. Specifications for this equipment are being prepared. Additionally, suitable space is being sought at LASL to permit such experimentation with nonradioactive wastes. Tentative space for this work has been identified in the basement of the existing Occupational Health Laboratory (OHL).

G. Assay and Accountability

The development of instrumentation for plutonium assay has continued to progress. A pancake configuration scintillation detector (a modified FIDLER system using a 127-mm-diam x 3.18-mm-thick NaI(Tl) crystal), which has been in operation for more than three months, has demonstrated exceptional reliability in monitoring sorted, low-density wastes. Using only a 10-sec counting period, this system can measure ^{239}Pu via the L series x rays (13-20 keV) of its uranium daughter at concentrations of 1 nCi/g at the 3σ level above background. Americium-241 is also determined by its 60-keV gamma ray and is summed with the ^{239}Pu content to obtain a total activity per gram measurement.

The other facet of the instrumental development effort is the development of

instrumentation for the assay of plutonium L x-ray emission of uranium and the higher energy gamma rays from plutonium and other transuranic elements. Known as the Multi-Energy Gamma Assay System (MEGAS), it is presently arranged for measuring the plutonium content of 0.06-m^3 (2 ft^3) cardboard waste boxes below the 10-nCi/g level. The larger diameter and thickness of the detector (127-mm-diam x 51-mm-thick NaI (Tl)) gives the system a wide dynamic range, permitting the counting of the low-energy L x ray at low plutonium concentrations and the higher energy gamma rays for larger quantities of plutonium or other transuranic elements.

Actual performance of the pancake and MEGAS systems are discussed in Section I, D.

The MEGAS has a mechanical apparatus for box rotation and translocation so that all portions of the container can be scanned by the detector. Integral with the MEGAS is a weighing device using a strain gauge which will determine the mass of each package while being scanned for transuranic material content. A minicomputer interface is under construction for automation of the MEGAS transuranic waste monitor. Study efforts are now underway to adapt the demonstrated detection techniques as a practical operational waste management tool.

III. EVALUATION OF PLUTONIUM-CONTAMINATED RADIOACTIVE WASTE DISPOSAL AREAS WITH RESPECT TO THEIR POTENTIAL HAZARD AND EVENTUAL DISPOSITION, A414 (D. E. MCCURDY, M. L. WHEELER, M. D. MCKAY, R. K. LOHRDING, J. A. MOHRBACHER)

A. Introduction

For several decades, transuranic-contaminated solid wastes have been placed in shallow burial grounds at several of the AEC contractor facilities. Concern over the potential consequences to the environment of release of the long-lived transuranic elements stored within these areas has stimulated studies at LASL to define the potential impact of past waste management burial practices and the risk the existing sites pose to future populations. The primary objective of this study is to determine the potential detriment that the solid waste disposal areas, as they exist in their present condition, present to the environment and to the public. In determining this detriment, the time period of concern is that required for the radioactive materials to decay to some innocuous level. For the transuranic elements this time period may extend over many hundreds of thousands of years.

One approach to evaluating the problem is to break up this overall time frame into three periods. The first time period of concern would be that time during which the Atomic Energy Commission can be expected to maintain supervision over the waste disposal areas. A speculative guess as to the length of this short-term period would be approximately 100-200 years into the future. During this period, the AEC would provide, through administrative action, security measures, waste management practices, geologic surveys, and monitoring programs, a reasonable assurance that no significant amount of radioactive material would migrate to any point outside the boundary of a particular site.

The second time period is that during which the sites will be under lessened administrative control due to the closing of the sites, or the possible return of the areas to public use. The time frame for the evaluation of the adequacy of the sites for this long-term storage is on the order of 5 000 years, based on information which can be utilized to describe previous climatic changes within a given area of the country and to anticipate future variations. The variables of human and animal population distributions and land use changes during this period cannot be safely predicted.

The third time period extends beyond the 5 000 year period. Only highly

speculative assertions can be made about any of the factors influencing the long-term adequacy of the burial sites during this period.

Within these various time frames, the evaluation of the suitability of the sites as safe and efficient means of removing hazardous materials from the immediate environment of man is a very complex problem, employing the efforts and interaction of many scientific and technological disciplines. One technique which has been utilized to evaluate similar problems in the nuclear industry is the concept of risk analysis.

Risk analysis, applied to the evaluation of the impact that the solid waste disposal areas pose to man, involves defining the potential mechanisms by which the material can be released, quantifying the transport of the material through the ecological systems to man, and estimating the radiation dose incurred by man as well as the appropriate consequences of such exposures. Due to the variety of waste management practices utilized over the past years for the burial of the waste and the differences in the hydrogeological characteristics of the burial sites throughout the country, the sites may have several potential modes of release of radioactive material to man's immediate environs. Each potential release mechanism would normally be evaluated in terms of its probability of occurrence through an event-tree analysis and the consequence of such releases estimated. In the final analysis, however, the many uncertainties associated with the understanding of various disposal practices and environmental transport mechanisms will result in risk estimates with inherent uncertainties. Even with these uncertainties this type of approach is considered to be the most applicable to the problem.

This report describes some of the concepts and methodologies to be utilized in the evaluation of the potential impact of the waste burial areas as well as some of the parameters involved in the abiotic and biotic transmission of radioactivity and the radiation dose assessments.

B. Risk Analysis Methodology

The basic steps to be carried out in this risk analysis are:

(1) Define the scope of the system under consideration
(2) State the hazard(s) to be considered
(3) Identify the causal events (those which ultimately produce the consequence)
(4) Formulate a model relating the causal events to the hazard(s)
(5) Compute the risk using probability functions associated with the causal events.

1. System. The system initially under consideration comprises the buried low-level solid waste at LASL, Area G, together with a geographic area surrounding the site, otherwise known as Mesita del Buey. Since the concern is with the potential release of radioactive material from the site, its movement through the surrounding area, and its ultimate effect on man, the consequences of many mechanical and biological phenomena must be examined. These processes are discussed in Section D and E in connection with abiotic and biotic transport of radioactive material through the environment. In this section only the general properties of the processes are considered.

2. Hazards. The hazard under consideration is the potential detriment that may result if radioactivity is released from the waste burial environment. The ultimate hazard would be the incurrence of a significant radiation dose by an individual or by a population group exposed to the released transuranic wastes. There would also be the subtle economic consequences or detriments associated with the rehabilitation of an area, which is directly related to the total risk involved in the disposal of the transuranic-contaminated wastes from

the burial area.

3. Causal Events. Risk analysis is concerned with both hazard and probability of occurrence. As a result, many highly unlikely situations which could cause a release of radioactive material from the burial area must be considered because of the magnitude of the accompanying hazard. Table VII is a list of some possible events which may cause a release.

Besides the probability of occurrence, the probability distribution must be related to each cause. This distribution describes the amount of radioactive material released. The two factors which determine the amount of material released are the amount of material in the containment area and the severity or intensity of the cause.

Accountability and inventory records make possible a fairly accurate estimate of the amount of material buried at Area G (see Section G). For the catastrophic events in Table VII an examination of historical records is in progress so that intensity histograms can be constructed and probabilities of occurrence estimated. Meteorological records will be used in a similar way. Theoretical considerations and experimental laboratory techniques are being used to associate intensities and amounts released.

4. Model. The purpose of the model is to facilitate the calculation of the dose to man from a release in order that the risk analysis might be performed. The compartmental diagram in Fig. 5 depicts the pathways for potential movement of radioactive material to man. The figure illustrates that the process of the movement of the material from the disposal site and its potential contact with man would have to occur in four basic steps:

(1) Material is released from the containment area.
(2) Material is moved and distributed over the area by predominantly mechanical (abiotic) processes.
(3) Material interacts with the environment in predominantly biological (biotic) modes.
(4) Material reaches man.

Figure 6 in Section E presents the model in greater detail. The transport process is separated into biotic and abiotic classes so that the particular types of mechanisms involved and the consequences of these mechanisms can be better studied. For the most part the abiotic systems account for the wide distribution of the radioactive material over the area, whereas the biotic systems usually have a local influence. The classifications of abiotic

TABLE VII

SOME RELEASE CAUSES

Lightning	Flooding
Tornadoes	Meteorite Impact
Dust Devils	Aircraft Impact
Strong Winds	Spontaneous Combustion
Earthquakes	Containment Leaks and Spills
Landslides	Operational Accidents
Volcanic Action	Low-Level Intentional Releases
Soil Migration	Arson and Sabotage
Erosion	Future Excavation

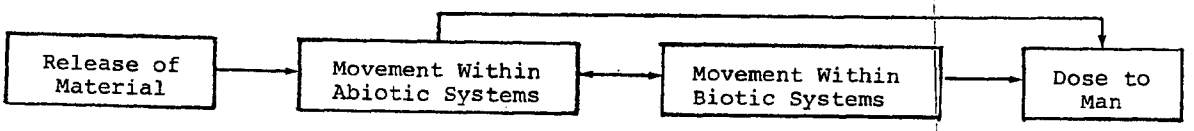

Fig. 5. Compartmental Diagram of Flow of Contaminated Material to Man.

systems are given in Table VIII.

In general, the release causes in Table VII can be thought of as discharging material into one or more of the abiotic transport systems in Table VIII. A discussion of the properties of these systems appears in Section D.

The biotic systems are formed from the components given in Table IX. A description of the components, their interaction with each other, and their interaction with the abiotic systems is given in Section E. An examination of the geographic area under consideration will dictate which components are put together in the different parts of the area to form the local biotic systems.

The movement of radioactivity between all abiotic and biotic systems is represented by the arrows in Fig. 5. Although the complexity of the total system detailed in Fig. 6, Section E, is hidden in Fig. 5, it still must be dealt with. However, each abiotic system and each biotic system can be handled individually in its relationship to the other systems, once they are all identified.

5. Mathematical Formulation. The following is a mathematical formulation of the compartmental model outlined in Fig. 5. The formulation is for a release due to a single cause. It is assumed that the release has occurred, or begun, and that the time following the release is partitioned into time intervals (periods) indexed by $t = 0,1,2,\ldots, T$. The length of the intervals is not specified, nor are they required to be of equal length. The abiotic systems are indexed on $j = 1,2,\ldots, J$, and the biotic systems are indexed on $k = 1,2,\ldots, K$. The unknown variables to be determined are:

y_{jt} = Amount of material in abiotic system j at the beginning of time period t,

z_{kt} = Amount of material in biotic system k at the beginning of time period t,

x_t = Amount of material introduced to man during time period t. (1)

TABLE VIII

ABIOTIC TRANSPORT SYSTEMS

Atmospheric System
Ground Surface System
Below Ground System
Surface Water System
Groundwater System

TABLE IX

COMPONENTS OF BIOTIC SYSTEMS

Terrestrial Plants
Terrestrial Animals
Soil
Aquatic Plants
Aquatic Animals
Water

The known functions and variables are:

Q_t = Amount of material released during time period t,

$c_{jt}(Q_t)$ = Amount of material entering system j due to the released amount Q_t during time period t,

$h_{jj'}(y_{j,t-1})$ = Amount of material moving from abiotic system j to abiotic system j′ during time period t,

$u_{jk}(y_{j,t-1}, z_{k,t-1})$ = Amount of material moving from abiotic system j to biotic system k during time period t,

$v_{kj}(z_{k,t-1})$ = Amount of material moving from biotic system k to abiotic system j during time period t,

$a_k(z_{k,t-1})$ = Amount of material introduced to man from biotic system k during time period t,

$b_j(y_{j,t-1})$ = Amount of material introduced to man from abiotic system j, not affected by any biotic system, during time period t. (2)

For the purpose of this derivation it is assumed that an artificial biotic system exists to "collect" the material depleted by radioactive decay. Also, it is assumed that all migrations, degradations, etc., attributed to any time interval occur instantaneously. The functions are constructed by examination of the particular processes involved and the distribution of the material within the systems, taking into account the natural degeneration of the material. The basis for the construction of these functions is given in Sections D, E, and F. The relationships that must be satisfied follow.

It is assumed that at period t = 0 the release to the abiotic systems begins, with no initial contamination of the abiotic or biotic systems or man. Therefore,

$$y_{j0} = c_{j0}(Q_0) \qquad \text{all } j,$$

$$z_{k0} = 0 \qquad \text{all } k,$$

$$x_0 = 0 \quad ,$$

and

$$u_{jk} = v_{kj} = a_k = b_j = 0 \quad \text{all } j,k \text{ for } t = 0. \qquad (3)$$

For subsequent time periods the following relationships hold: The amount of material in abiotic system j at period t is the amount in it at period t-1, plus the amount transferred to it from other abiotic and biotic systems, less the amount transferred to other abiotic and biotic systems and man. Hence,

$$\begin{aligned} y_{jt} = {} & y_{j,t-1} + c_{jt}(Q_t) \\ & + \sum_{j' \neq j} h_{j'j}(y_{j',t-1}) \\ & + \sum_k v_{kj}(z_{k,t-1}) \\ & - \sum_{j' \neq j} h_{jj'}(y_{j,t-1}) \\ & - \sum_k u_{jk}(y_{j,t-1}, z_{k,t-1}) \\ & - b_j(y_{j,t-1}) \end{aligned} \qquad (4)$$

all j,
t = 1,2,...,T.

Using similar reasoning, the amount of material in biotic system k is given by

$$z_{kt} = z_{k,t-1} + \sum_j u_{jk}(y_{j,t-1}, z_{k,t-1}) - \sum_j v_{kj}(z_{k,t-1}) - a_k(z_{k,t-1}) \qquad (5)$$

all k,
t = 1,2,...,T.

Finally, the amount of material reaching man is

$$x_t = \sum_k a_k(z_{k,t-1}) + \sum_j b_j(y_{j,t-1})$$

t = 1,2,...,T.

For any particular set of source term values $Q_0, Q_1, \ldots, Q_{t-1}$ the hazard to man associated with time period t is defined

by the hazard function $H_t(x_t)$. This function associates an intake (or dose) quantity x with a specific detriment or hazard to man, for example, a 50-year dose commitment. The total hazard after T periods is

$$H(Q_0, Q_1, \ldots, Q_{t-1}) = \sum_{t=0}^{t-1} H_t(x_t) \quad . \qquad (6)$$

In general, however, the source terms Q_t should be considered as random variables with probability density functions $g_t(Q_t)$. In view of this, the hazard, H*, associated with the release is defined as the expected value of H with respect to g_t. That is

$$H^* = \sum_{t=0}^{t-1} H_t(x_t)\ g_t(Q_t)dQ_t, \qquad (7)$$

where the x_t's depend upon the values of the Q_t's.

Recalling that the preceding formulation is for a release due to a single cause, the risk associated with the total system is now formed. Letting H_c^* denote the hazard associated with cause c and p_c denote the probability of its occurrence, the risk is defined as

$$\text{Total Risk} = \sum_c H_c^*\ p_c \quad . \qquad (8)$$

6. Illustrative Calculations for a Sample Risk Analysis of a Single Causal Event. Suppose that the risk associated with the occurrence of a single dust devil three months before the harvest season for a particular field is to be calculated.

Assume that the probability of the dust devil occurrence is, approximately, p = 0.15 and that the expected amount of material, Q, that could be released to the atmosphere is 40 μCi.

Supposing that 10% of the released amount, Q, is deposited on a field, and that the fraction of this amount remaining after 90 days is $e^{-\lambda 90}$, then the amount on the field at the harvest time is

$$z = (0.10Q)\ e^{-\lambda 90} \quad .$$

If the processing of the harvest removes 60% of the material, then the amount, a, which reaches man is

$$a = (0.40)\ (0.10Q)\ e^{-\lambda 90},$$

and for $1/\lambda$ = 15 days

$$a = 9.9 \times 10^{-5} Q\ \mu\text{Ci}.$$

Defining the hazard as the total dose to man, the expected hazard, H*, over all possible values of released amount Q is

$$H^* = (9.9 \times 10^{-5})(40)\ \mu\text{Ci} \cdot \frac{\text{rem}}{\mu\text{Ci}}$$
$$= 4.0 \times 10^{-3}\ \text{rem}.$$

Finally, the associated risk in μCi to man for the hypothetical dust devil example is given by

$$\text{Risk} = H^*\ p$$
$$= (4.0 \times 10^{-3}\ \text{rem}) \cdot (0.15)$$
$$= 6.0 \times 10^{-4}\ \text{rem}.$$

C. Hydrogeologic Environment of Waste Disposal Areas at Los Alamos

Solid waste disposal areas at Los Alamos are located on a series of finger-like mesas, extending southeastward from the main mass of the Jemez Mountains. These mesas were formed by the erosion of deep, narrow canyons in a series of ash-flows and ashfalls of rhyolite tuff. The mesas vary from a few hundred meters to over a kilometer in width. The separating canyons are 30-60 m deep near the disposal areas.

The individual flows vary in thickness, but tend to average about 10 m. The various geologic units containing the tuff have a total thickness of over 300 m and overlie a sedimentary formation containing the regional groundwater aquifer. The tuff is unsaturated throughout its entire thickness, except beneath the canyon bottoms. In these areas, intermittent ephemeral streams have produced localized

saturated alluvium. The zone of saturation extends a few meters into the underlying tuff. Various investigations have indicated that there is no discernible recharge to the regional groundwater supply by water from these perched water tables.[7,8]

As indicated, the regional groundwater table is over 300 m below the surface of the mesas. The tuff is quite porous, averaging about 40% void space, with a specific gravity of about 1.5. In spite of the high porosity, the tuff is relatively impermeable when saturated, being more comparable to clay than to sand in that regard.[9]

The tuff is jointed at irregular intervals by fractures apparently related to the cooling of the ash deposits. These fractures extend through individual flow units and in some areas stand open as much as 100 mm. Considerable attention has been given to the impact of these fractures on the migration of moisture from the surfaces of the mesas to the adjacent canyons or to the regional water table. As yet, no firm conclusions have been reached.

The present climate of Los Alamos is quite dry, with an annual average precipitation of about 460 mm. The evaporation rate is typical of a high desert area and, as evidenced by the absence of perennial streams, is equal to or greater than the average precipitation. However, considerable surface runoff is associated with the melting of winter snow accumulation and with high intensity summer storms. These periods of high moisture availability provide the principal source of moisture for infiltration into the soil and underlying tuff.

D. Abiotic Transfer Processes

Within the hydrogeologic environment of Los Alamos, contamination can be transferred from the disposal area to the biosphere by a variety of mechanisms, both biotic and abiotic. Where possible, each of these mechanisms will be quantified and described in a form compatible with the mathematical structure of the risk analysis previously described. In some instances these descriptions will be relatively simple, taking the form of a single equation. In others, a complex modeling approach may be required to adequately describe the process.

For analytical convenience, the abiotic transfer processes have been divided into several categories, as listed in Table VIII. Those mechanisms operating on or below the surface of the ground can be generally described as hydrogeologic in nature. Listed in Table X are the hydrogeologic processes currently under consideration.

TABLE X

POSSIBLE MECHANISMS FOR HYDROGEOLOGIC TRANSPORT OF CONTAMINANTS OUT OF DISPOSAL AREAS

1. Migration through soil water by self-diffusion
2. Leaching, transport, and redeposition by migrating soil water
3. Earth movements resulting from seismic activity or major impact by falling objects (meteors, airplanes, etc.)
4. Erosion of filled areas by wind or water, resulting in surface transport of contaminants

1. Migration by Self-Diffusion. Transuranics are present in waste material either as solid material or as contaminants in liquids or solids, and not as volatiles. Thus, self-diffusion will only occur due to migration of molecular size particles through soil solutions. Self-diffusion of plutonium in soils of the Hanford site has been studied by Hajek.[10] He concluded that plutonium could be expected to move approximately 85 mm in 2.4×10^5 years by this process. The soils used in his experiments were moistened to 20% water content by volume, much wetter than the observed 1-10%

moisture content typical of the tuff beneath burial areas at Los Alamos. It is safe to conclude that self-diffusion of plutonium is not an important mechanism for movement of contamination out of disposal areas at Los Alamos.

2. Leaching, Transport, and Redeposition. The rate of transport of plutonium out of a burial area by migrating soil moisture is dependent on many factors. The chemical form of the isotope and its solubility in water must be determined. The movement rate of soil moisture through and out of the waste material must be established. The migration pathway through the tuff around and beneath the disposal area will be controlled by existent moisture gradients and hydraulic conductivity variations. Finally, the redeposition rate of plutonium, either by precipitation or adsorption reactions within the pores of the tuff and fill material, must be specified.

Accurate determination of all of these factors requires data not presently available for the Los Alamos area. However, extrapolation from other investigations coupled with additional field investigations will permit an analysis of this transfer process.

The amount of dissolution of plutonium from waste is strongly affected by the chemical form of plutonium present and by the chemistry of the leaching solution. In general, soluble salts increase the solution rates, as do organic complexing agents. Only one study which investigated plutonium leaching from waste material was located.[5] All others examined removal from soil material containing plutonium in various forms. In general, solutions representative of naturally occurring water leach very little plutonium from waste, except for an initial quantity associated with realignment of ion-exchange equilibria. Many authors comment that plutonium leaching from soil appears to be more affected by solubility than by exchange reactions.

Plutonium adsorption by soils is equally complex, being strongly affected by pH, as well as the concentrations of other metallic ions. Plutonium appears to be present as a polymer in most solutions and is highly sorbed by natural materials. In general, it behaves as a cation at pH values up to about 12, at which point it exhibits strong anionic properties.

Dissolution and adsorption of plutonium can be quantitatively described in terms of a distribution coefficient (ratio of material concentration on solid phase to concentration in liquid phase), K_d. This coefficient can be evaluated from information provided by several authors. Table XI presents values of K_d, together with data on dissolution, obtained from various authors.

It is evident that a wide range in values exists for both dissolution and adsorption reactions depending on soil type and experimental design. The data from Christenson[12] is only approximate, since the calculation of K_d involved many assumptions by this author. Accordingly, a midrange value of 10^3 will be used for the adsorptive distribution coefficient.

The data on dissolution is even less consistent. However, a small percentage value appears to be associated with solutions low in dissolved salts and organic solvents. A value of 2×10^{-4} will be used for the dissolution coefficient, K_s. This expresses the fraction of material removed through dissolution from a given volume of waste by an equivalent volume of migrating water.

These coefficients, when combined with known moisture flow rates, can be used to compute migration rates of plutonium through porous materials. The relation between the velocity of the contaminant and that of the carrying solution is as follows:

TABLE XI

SOLUTION PERCENTAGES AND DISTRIBUTION COEFFICIENTS

FOR

PLUTONIUM IN WASTE AND SOIL

Author	Solution Type or Concentration	Percent Leached	Distribution Coefficient, K_d
Hajek[10]	Tap water 1 M $NaNO_3$	0.1%,20 col.vol. 3.5%,20 col.vol.	10^4
Waterbury[5]	Distilled water	$Pu(NO_3)_4$,50-90% PuO_2, 0.05-0.1%	-
Rhodes[11]	150 μg/liter	-	1 000 (pH 5.4-7) 150 (pH 8.4-13) >2 000 (remainder of pH range)
Christenson[12]	Approx 8 μg/liter	-	540 (pH 7)
Knoll[13]	Organic solvents	25-100%,50-5 col. vol.	-

$$\frac{V_i}{V_w} = \frac{1}{1+\frac{\rho}{\varepsilon}k_d} \qquad (9)$$

where

V_i = contaminant velocity

V_w = solution velocity

ε = porosity of medium

ρ = bulk density of medium.

For the tuff in the Los Alamos area $\rho = 1.46$ and $\varepsilon = 0.43$ (Ref. 9). Using a K_d value of 10^3,

$$\frac{V_i}{V_w} = 2.9 \times 10^{-4} \ . \qquad (10)$$

Thus, plutonium in solution will be transported at a rate nearly 3×10^{-4} slower than that of migrating soil moisture. The concentration of the plutonium in solution at the beginning of the leaching process, C_i, is

$$C_i = K_s C_w \qquad (11)$$

where

K_s = solution coefficient

C_w = plutonium concentration in waste matrix.

Plutonium concentrations in waste material can be estimated from existent data on waste volumes and the curie content of the waste obtained as part of the waste inventory discussed in this report.

Little information is available on actual movement rates of moisture either through the tuff or the fill material surrounding the waste. This determination is complicated by the heterogeneous nature of the tuff. Considerable data have been obtained on moisture content of the upper 20-40 m of the tuff.[7,8,9] Data on moisture content of the fill material overlying and surrounding the waste is generally not available. A recent investigation obtained samples of fill material in the upper 3-4 m of disposal pits. These

samples are presently being analyzed for moisture and radionuclide content. Some information is available on saturated hydraulic conductivities of the tuff, and a portion of the data presented by Abrahams[9] has been used to calculate unsaturated conductivities. Further work is planned on defining the actual amounts, pathways, and movement rates of migrating soil moisture. This information, when coupled with the concept of relative migration rates of plutonium to that of soil moisture, will permit a description of the rate and direction of the migration of plutonium out of the disposal pits and shafts.

E. Biological Transport Mechanisms

1. Introduction. Radioactive material in a waste burial pit can be thought of as being in an abiotic environment with little chance of interacting with the biotic components of an ecosystem unless the material is released by one of the mechanisms described earlier. However, once the material is released from the confines of the waste pit or shaft it can be transported through the various ecosystems to man. A general outline of the critical pathways by which man may be exposed to such releases is presented as Fig. 6. It is assumed that soil is the main reservoir or pool of radioactivity for most of the pathways under consideration. The flow diagram depicted in Fig. 6 can be segregated into essentially three principal transport categories: atmospheric, terrestrial, and aquatic. Many of the transport mechanisms within the broad categories can interact with each other and thus should not be thought of as individual entities, incapable of influencing the other transport processes.

Although the transport of radioactivity through the environment is a time dependent process, little is known concerning the relative time frame of the processes. In lieu of the necessary information to mathematically describe such mechanisms, the transport of the transuranium elements through the various pathways outlined will be considered instantaneous. The only exception to this procedure is the foliage surface contamination ⟶ herbivore ⟶ man pathway, in which a loss rate due to weathering is applicable. In view of the time frame under consideration for the hazard evaluation, that is, 50 to 100 000 years, biological transport processes of the order of even a year can be thought of as instantaneous. Of course, each biological component of a given pathway will discriminate against the incorporation of the transuranium elements to some degree. The extent to which the radioactive material flows from one trophic level to a higher level or from an abiotic media, such as soil, to a biotic component of a given pathway will be evaluated through the use of discrimination or concentration factors. Individual concentration factors, defined as the ratio of μCi/g of the recipient to the μCi/g of donor at equilibrium or as the fraction of incident contaminant retained by the recipients,[14] will be documented for each abiotic or biotic link within a transport process.

2. Terrestrial Environment Transport Considerations.

a. The Uptake of the Transuranium Elements from Soil by Terrestrial Plants. In reviewing the literature on the uptake of the transuranium elements by plants, one can generally classify the reported information by laboratory and field experiments. Laboratory experiments are usually designed to prevent any foliage surface contamination due to airborne suspension of the contaminated soil. Therefore, information obtained from laboratory experiments is directly applicable to the evaluation of plant uptake under conditions where deep-rooted plants are growing on a waste burial area covered with a layer of uncontaminated soil. Estimates on the initial plant uptake can also be made for the case where contaminated water is used for flood or rill irrigation of agricultural crops.

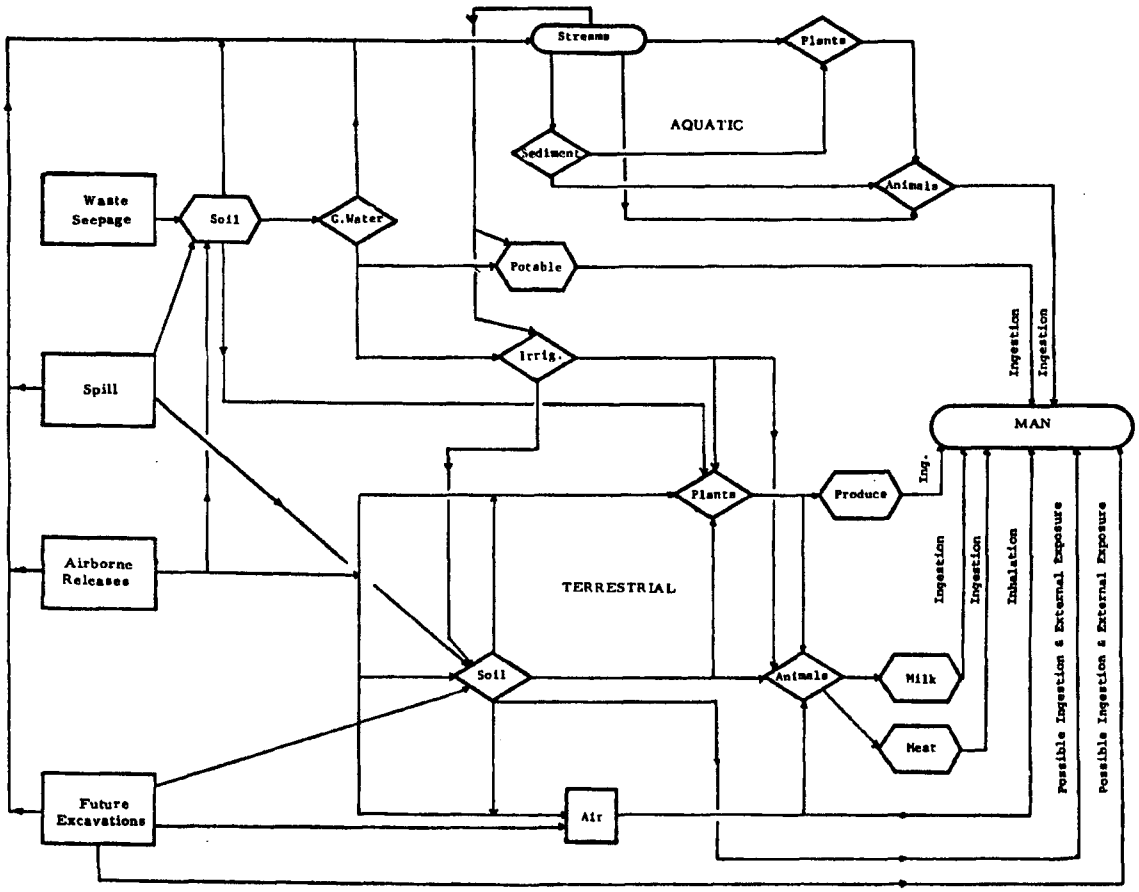

Fig. 6. Potential Pathways by Which Man May be Exposed to Radioactivity from Buried Wastes.

Plant uptake data from field measurements, normally documented in terms of a concentration factor (μCi/g dry plant per μCi/g dry soil), is related to both root uptake and to surface contamination from resuspension processes. This information is particularly important in evaluating the contamination of vegetation growing on contaminated top soil for extended periods of time. In relating laboratory uptake data to field data, it is found that the observed concentration factor (CF) from field data is several orders of magnitude greater than that from laboratory data. Aside from the many subtle variations in laboratory and field conditions, a good portion of the apparent increase in plant contamination is due to surface contamination. The degree to which surface contamination contributes to the plant contamination has been inferred by Fowler et al.[15]

Laboratory Studies

It has been generally recognized that the plant uptake of transuranium elements from the soil via the root system is very limited. The discrimination factor for plutonium and americium from soils to plants ranges from 10^{-3} to 10^{-5} depending on plant species and vigor, soil type, and chemical form or solubility of the deposited material.[16-21] Americium and, especially, plutonium are tenaceously held by the soil,

preventing plant incorporation, particularly if salts, acids, detergents, and organic compounds are absent. Plutonium apparently does not selectively concentrate in any one part of the plant shoot,[15] although Jacobson et al.[20] found that plutonium became fixed within or on the surface of the root system. Soil characteristics, such as soil structure, organic content, pH, and the amount and type of clay present, influence the availability of plutonium to some extent but probably by no more than a factor of ten.[16]

The uptake of plutonium as a function of oxidation state from calcium bentonite suspensions (100 mg/liter) into three-week-old barley plants was studied by Jacobson and Overstreet.[20] Even though the study was for a very limited absorption period (24 hours) the results indicated that barley leaf incorporation of plutonium did vary according to oxidation state. The investigators reported a plant uptake selectivity of $Pu^{3+} < Pu^{4+} < Pu^{6+}$. It was noted that the variation in the amount incorporated at the different oxidation states appears to be associated with the amount of clay adsorption, that is, the greater the clay adsorption, the lower the leaf uptake. The root system sorption was found to be greatest for the oxidation states having the least clay sorption. The reported fractional uptake of the initial plutonium dose by plant leaves ranged between 4.5×10^{-6} (Pu^{3+}) and 10^{-4} (PuO_2^{2+}).

In 1955, Rediske et al.[22] studied the uptake of fission products by plants using the Neubauer seedling test. Essentially, the test involved growing 100 barley seedlings in 100 g of soil contaminated with a radionuclide to a concentration of about 0.1 μCi/g. The seedling growth exhausted the available nutrient supply such that the radionuclide concentration in the leaf portion reached a maximum in about 18-20 days. Data on the uptake of ^{239}Pu into the barley plant from Ephrata loamy soil (pH 7.3) indicated a concentration factor of 9×10^{-4}. This value is approximately an order of magnitude greater than the CF value reported by Wilson and Cline for barley grown on contaminated Ephrata fine sandy loam (1.3×10^{-4}) (Ref. 19).

Studies have also been performed to determine the distribution of the fission products in the plant organs.[22] Beans, tomatoes, Russian thistle, grasses, and wheat were grown in a contaminated nutrient solution and the fruits and leaves analyzed separately for radioactivity content. For typical fission products, the concentration factor for the fruit portion of the seed plants was less than that for the leaves, but by no more than a factor of ten. No specific plant organ studies were performed using the transuranium elements. The measured CF value for plutonium incorporation in the leaves of beans, tomatoes, Russian thistle, and barley grown in the nutrient solution did not vary by more than an order of magnitude (2×10^{-4} to 2×10^{-5}).

The uptake of ^{239}Pu in bean plants as a function of nutrient solution pH was also investigated by Rediske et al.[22] In this study, the CF value, as measured in the aerial portion of the plant, increased from 10^{-4} to 10^{-3} when the pH was adjusted from 7 to 4.

The variation in the plant uptake of $Pu(NO_3)_4$ with soil type was evaluated by Wilson and Cline.[19] Barley was grown using a modified Neubauer technique for 18 days in three different soils spiked with ^{239}Pu. For the three soil types investigated (Ephrata fine sandy loam, Milville silt loam, and Cinebar silt loam), the fractional amount absorbed by the barley and the concentration factor values varied by only a factor of five. The investigations did point out that the uptake of Pu^{4+} from the acidic Cinebar soil was greater than the uptake from the alkaline Ephrata or the calcareous Milville soils. The concentration factor values ranged between 2.2×10^{-4} (Cinebar soil) and 4.0×10^{-5} (Milville soil).

Similar studies on the incorporation of $Pu(NO_3)_4$ into oat seedlings as a function of soil type were conducted by Cummings and Bankert.[23] Oat seeds (Avena sativa) were planted in nine different soil types and following a three-week growing period, the shoots were collected and analyzed for ^{238}Pu. Soil moisture, as well as atmospheric conditions, were maintained at a constant level during the study. Based upon the data presented, there did not appear to be any obvious correlation between plant uptake and the soil characteristics of cation exchange capacity, organic matter, and pH. The percent uptake of the initial plutonium soil dose ranged between 7×10^{-6} and 2.3×10^{-4} for the nine soils tested.

Cline, in evaluating the incorporation of americium and plutonium nitrate into barley from Cinebar and Ephrata soils, found a substantial difference in the plant uptake of the two elements.[24] The concentration factor for americium was 15-30 times greater than that of plutonium for the two soils. For barley grown in Cinebar soil contaminated with ^{239}Pu, the measured CF was 2×10^{-4}. Similar studies for the Ephrata soil yielded a CF only a factor of two less than that of the Cinebar soil. Both studies were performed using the Neubauer technique.

Investigations on the uptake of transuranium elements (Np, Pu, Am) as nitrates from Burbank loamy soil by tumbleweed (Salsola kali) and cheatgrass (Bromus tectorum) have been performed by Price.[17] The laboratory studies simulated normal field conditions and were carried out over a period of two months with some NH_4NO_3 fertilization. In both plant species, the plant uptake was related to the radionuclide applied, with neptunium being absorbed the greatest followed by americium and plutonium in decreasing absorption ability. For $Pu(NO_3)_4$, the observed concentration factors for tumbleweed and cheatgrass were 4.6×10^{-5} and 1.7×10^{-5}, respectively.

Studies conducted by Romney et al. indicated that the concentration of plutonium in plants grown on contaminated soil increased with time.[25] In a five-year cropping sequence of ladino clover (Trifolium repens) grown on Nevada Test Site soil heavily contaminated by previous weapons tests, the concentrations of ^{239}Pu in dry plant tissue increased by a factor of 7.3 between the years 1958 and 1962. The investigators attributed the increase in plant uptake to several possibilities: (1) the development of the root system which would create better contact between the roots and the plutonium fallout particles, and (2) the increase in the availability of the residual plutonium for root uptake due to the natural chelation of the plutonium by decaying roots. This latter possibility is supported by data acquired in another experiment where Romney et al. showed that the uptake of plutonium in alfalfa could be enhanced as much as tenfold through the addition of the chelating agent DTPA (diethylenetriaminepentaacetic acid).[25]

The concentration factors for the soil-to-plant pathway (by means of the root system only) which will be utilized in the environmental transport models have been summarized in Table XII. The values presented were obtained by taking the geometric mean of the numerical data presented in Table XIII for soil media (not nutrient solutions) and are in no manner absolute. The data for the nutrient solutions were not incorporated into the averaging process because it is very unlikely that such soil solutions could exist in a field environment. It should be pointed out that the use of a geometric mean does bias the data toward a lower numerical value than that obtained by an arithmetic mean. Also included within the table are the ranges of the concentration factors found in the literature for neptunium, plutonium, and americium. These CF ranges will be utilized in sensitivity studies of the risk analysis models.

TABLE XII

RANGE AND GEOMETRIC MEAN OF CONCENTRATION FACTORS

DERIVED FROM

SEVERAL LABORATORY STUDIES OF Np, Pu, and Am

Nuclide	Concentration Factor (Plant to Soil) Range	Geometric Mean
Np	1.3×10^{-2} to 1.1×10^{-1}	3.8×10^{-2}
Pu	1.7×10^{-5} to 3.0×10^{-4}	9.3×10^{-5}
Am	6.0×10^{-4} to 3.0×10^{-3}	1.5×10^{-3}

b. The Incorporation of the Transuranium Elements by Animals.

Several possible modes of entry exist by which terrestrial animals can incorporate the transuranium elements into their bodies. Herbivores and small animals may ingest radioactivity by foraging on contaminated vegetation, drinking contaminated water, or consuming radioactive soil while eating or grooming the fur. Inhalation may also represent an important avenue of intake for burrowing animals and for animals living in an environ where considerable resuspension is evident. Predators, however, would probably derive most of their intake from the consumption of smaller prey animals which contain radioactivity in their internal organs or on their pelts. Man enters into this terrestrial food web by consuming the meat from certain sport game and domestic grazing animals and the milk from dairy herds.

Ingestion has been considered to be the primary route of entry involved in the food-chain transport of residual plutonium in the environment.[26] Even though ingestion may be an important mode of entry, a review of the laboratory studies with rats and pigs has shown that the plutonium absorption from the gastrointestinal (GI) tract is very limited.[27] Experiments conducted with rats indicate that plutonium absorption from the GI tract can vary from 0.001% to 1.9% depending on such parameters as the valence state of the administered plutonium, the age of the animal, and the presence of chelating agents. Weeks et al. found that plutonium in very acidic solutions, in the form of a citrate, or in the hexavalent state, was absorbed to a considerably greater extent than $Pu(NO_3)_4$ (Ref. 28). In chronic-feeding experiments with rats, the average absorption of plutonium (pH 2, $Pu(NO_3)_4$) from the gastrointestinal tract was 0.002 8%. In these experiments it was shown that the concentration of plutonium solution administered had no effect on the fractional amount absorbed from the GI tract. Earlier experiments with rats by Katz et al. indicated that the amount of ^{238}Pu nitrate (pH 2) absorbed from the GI tract and distributed throughout the total body, soft tissue, and skeleton was 0.002 61%, 0.000 21%, and 0.002 34% respectively.[29]

In single-feeding experiments with pigs, Weeks et al. determined that the GI absorption of plutonium nitrate was approximately the same as that measured for rats, 0.002 2% (Ref. 28). Nine days after the intragastric administration, three pigs were sacrificed and the plutonium tissue distribution was measured in each specimen. The percent of the administered dose found in the various tissues was the following:

TABLE XIII

LABORATORY STUDIES CITING SOIL-TO-PLANT CONCENTRATION FACTORS FOR THE TRANSURANIUM ELEME

Nuclide	Chemical Form	Soil Type	Plant Species	Fractional Uptake	CF, Plant to
^{237}Np	$Np(NO_3)_5$	Hanford soil (Burbank loamy sand)	Tumbleweed	2.1×10^{-2}	1.1 x 1
	Above	Above	Cheatgrass	1.2×10^{-3}	1.3 x 1
^{239}Pu	PuO_2^{2+}	Calcium bentonite suspensions	Barley (3 weeks old)	1.0×10^{-4}	-
	Pu^{3+}	Above	Above	4.5×10^{-6}	-
	Pu^{4+}	Above	Above	2.4×10^{-5}	-
	$Pu(NO_3)_4$	Ephrata	Barley	1.9×10^{-6}	1.3 x 1
	Above	Milville	Above	6.0×10^{-7}	4.0 x 1
	Above	Cinebar	Above	2.2×10^{-6}	2.2 x 1
	Above	Hanford soil (Burbank loamy sand)	Tumbleweed	9.3×10^{-6}	4.6 x 1
	Above	Above	Cheatgrass	1.9×10^{-6}	1.7 x 1
	Fallout	Nevada Test Site	Ladino clover	2×10^{-5}	1.0 x 1
Pu	$^{239}Pu(NO_3)_4$	Cinebar	Barley	-	2 x 1
	Above	Ephrata	Above	-	1 x 1
	$^{238}Pu(NO_3)_4$	Fox (Ohio)	Oats	7×10^{-8}	-
	Above	Blanton (Florida)	Above	1.0×10^{-7}	-
	Above	Lakeland (Florida)	Above	6.1×10^{-7}	-
	Above	Adamsville (Florida)	Above	1.2×10^{-6}	-
	Above	Ruskia (Florida)	Above	5.3×10^{-7}	-
	Above	Leon (Florida)	Above	4.0×10^{-7}	-
	Above	Congaree (Alabama)	Above	2.2×10^{-6}	-
	Above	Ochlockonee (Alabama	Above	3.3×10^{-7}	-
	Above	Amito (Alabama)	Above	2.3×10^{-6}	-
^{239}Pu	-	Nutrient solution	Bean leaves	-	3 x 1
	-	Above	Tomato leaves	-	2 x 1
	-	Above	Russian thistle leaves and stem	-	2 x 1
	-	Above	Barley leaves	-	2 x 1
^{241}Am	$Am(NO_3)_3$	Hanford soil (Burbank loamy sand)	Tumbleweed	3.2×10^{-4}	1.4 x 1
	Above	Above	Cheatgrass	7.2×10^{-5}	6.0 x 1
	Above	Cinebar	Barley	-	3 x 1
	Above	Ephrata	Above	-	2 x 1

skeleton -	0.001 5
liver -	0.000 36
other soft tissue -	0.000 33.

In addition to the work by Weeks et al.,[28] on the gastrointestinal absorption of plutonium in various forms, Baxter and Sullivan[30] have confirmed the fact that plutonium citrate is much more readily absorbed in rats than the nitrate. The investigators also demonstrated that the effect of DTPA chelation is to increase plutonium absorption by another factor of five over that observed with the citrate administration to rats. The presence of citrates or chelating agents not only affects the fractional amount of plutonium absorbed from the gastrointestinal tract but also influences the amount and the distribution of plutonium within the body.

The most current review of the uptake of the transuranium elements and their body distribution has been performed by the Task Group of Committee Two of the International Commission on Radiological Protection and is published as ICRP Publication 19 (Ref. 27). For plutonium, the Task Group summarized their review accordingly: "Following ingestion of relatively insoluble material, such as PuO_2, the absorbed fraction may be taken to be about 10^{-4}%. For more soluble compounds (for example, plutonium nitrate) the absorption is also low due to hydrolysis; the absorbed fraction may be assumed to be about 3×10^{-3}%." In view of the many parameters which influence the absorption of plutonium from the GI tract, it is felt that these numerical values represent the best approximations which can be applied to the pathway model under consideration for plutonium.

The only reported investigations found concerning the absorption of neptunium from the GI tract of animals were documented by Ballou et al.[31] In their experiments with administering high dose levels to rats, the investigators found that the citrate complex was more readily absorbed than the nitrate form (by approximately a factor of two). The percent of the nitrate dose absorbed from the GI tract following an acute (24 hours) administration was reported as 0.12 ± 0.06. The investigators also found that the absorption of neptunium varied according to oxidation state. When neptunium was administered in three different oxidation states (IV,V,VI) as a citrate, the absorption increased as a function of the valence from 0.30% for Np^{4+} to 2.3% for Np^{6+}. Studies on the organ distribution of neptunium were not conclusive, but their findings did suggest that the general behavior of neptunium is not greatly different from that of plutonium.

Further studies are required to provide reasonable estimates on the organ distribution of the transuranic elements within animals. Organ distribution information is required to evaluate the amount of radioactivity ingested from the consumption of meat from sport game and grazing animals. Additional literature searches will be conducted along these lines during the next quarterly reporting period.

c. Forage and Agricultural Produce Contaminated by the Deposition of Airborne Radioactivity. The important food-chain considerations in the event of an airborne release of radioactivity are the air ⟶ forage ⟶ cow ⟶ milk ⟶ man pathway and the air ⟶ agricultural produce ⟶ man pathway. These pathways must be evaluated separately from the other soil-to-plant contamination pathways since the surface contamination of the vegetation caused by the passage of an airplume is time dependent. Investigations by Thompson have shown that the forage removal half-life under these conditions is approximately 13 days.[32] Since the transuranic elements all have very long physical half-lives with respect to this value, the effective removal half-life would also be about 13 days.

For the first pathway shown above, the

mathematical equation describing the amount of radioactivity ingested per day by man drinking the milk from a cow grazing on contaminated forage is

$$q(t)_{milk} = FC\ K_1\ e^{-\lambda_f t}$$

where $q(t)_{milk}$ is the amount of radioactivity ingested per day at some time t following the initial forage contamination, FC is the initial forage contamination level in $\mu Ci/m^2$, λ_f is the effective forage removal constant (day^{-1}), and $K_1 = Gf_m b$. In the equation describing K_1, G is the amount of pasture grazed per day, f_m is the fraction of the radioactivity consumed daily which appears in a liter of milk, and b is the milk ingestion rate constant (1 liter per day).

The estimate on the amount of pasture grazed per day by cattle is 45 m^2/day and was taken from Burton.[33] This value, however, is highly variable and would depend upon the vegetation density of a given area. The time of year may also influence this value, especially if dairy herds are put on grain or other feeds during severe weather conditions.

The fractional amount of radioactivity consumed by a dairy cow appearing in a liter of milk has been examined by Sanson for acute doses of PuO_2 and $PuCl_4$ (Ref. 34). In either form, the maximum concentration in milk was about 10^{-6}% of the dose per liter of milk, and this value was attributed to fecal contamination. The highest milk concentrations were found after the first day and before the fifth day, but this was also true of the concentration of plutonium in the feces. The author did state that the milking technique used in the study was probably at least as good as that used in commercial dairy herds, so the concentration of plutonium measured in the milk can be taken as an indication of the minimum concentration of plutonium in any form occurring in milk after ingestion by a cow.

The amount of radioactivity available for ingestion by the contaminated agricultural produce pathway can be estimated by the following equation:

$$q(t)_{A.P.} = FC\ b\ f\ e^{-\lambda_f t}/\rho$$

where $q(t)_{A.P.}$ is the amount of radioactivity available for ingestion per day at the time t following the initial contamination event and during the harvesting of the crop, FC is the initial contamination level ($\mu Ci/m^2$), λ_f is the effective forage removal constant (day^{-1}), b is the typical dietary ingestion rate of the crop in grams per day, f is the decontamination factor obtained during the processing of the crop for market, and ρ is the crop density in g/m^2.

At the present time, most of the parameters mentioned above are under consideration. Parameters such as vegetation density, types of crops, and ingestion rates for the various agricultural products vary according to the area of the country under consideration. It is hoped that some of this information can be obtained from agricultural productivity estimates published by the State and National governments.

F. Radiological Assessment of the Intake of the Transuranic Elements

1. Introduction. According to the International Commission on Radiological Protection,[35] the basic standards for maximum permissible internal exposure are predicted on two different criteria. For bone-seeking radionuclides, such as ^{239}Pu and ^{90}Sr, the maximum permissible internal exposure estimate is based on a comparison with the biological damage produced by a certain quantity of ^{226}Ra and daughter products in the body. This method is a result of calculations which have determined (1) the number of microcuries of a radionuclide deposited in the bone that will deliver the same effective RBE dose as delivered by 0.1 μCi of ^{226}Ra and its progeny, and (2) the number of microcuries deposited in the bone that will result in

damage comparable to that observed from known deposits of ^{226}Ra in the bone. Within the ICRP recommendations for the basic standards of maximum permissible internal exposure for occupational groups, the basic relationship between the radium body burden and the radiation bone dose was stated, "The effective RBE dose delivered to the bone from internal radiation during any 13-week period averaged over the entire skeleton shall not exceed the average RBE dose to the skeleton due to a body burden of 0.1 μCi of ^{226}Ra. This is considered to correspond to a dose rate of 0.56 rem/week in the case of ^{226}Ra (derived from a dose rate of 0.06 rad/week, an RBE of 10 and a relative damage factor of 1)." For the radionuclides not having the bone as the principal site of internal deposition, the criteria utilized to limit the intake have been based on radiation dose standards for the various organs.

Based on the information presented by one investigator,[36] it is not certain that the most appropriate approach to evaluating the biological implications of internally deposited ^{239}Pu is from a radiation dose standpoint. Healy[36] feels that "the basic standard for plutonium-239 absorbed into the body, i.e., outside of the lung or other site of initial deposition, is 0.04 μCi for occupational exposure. This value was derived by biological comparison of the late effects when injected into animals with those of radium for which a significant body of information on the effects in humans exist." Under this criterion, the use of the body burden may be somewhat more appropriate to evaluating the biological hazard associated with the internal deposition of ^{239}Pu.

Although uncertainties may currently exist in adopting an approach to evaluating the hazards of internally deposited ^{239}Pu, the basic mathematical equations describing the uptake and retention of the transuranic elements can be developed to address both approaches. This can be accomplished because the first step in formulating the equations which estimate the radiation dose to the internal organs is to quantify the organ burdens from a specific intake.

2. Mathematical Equations Utilized in Evaluating the Radiation Dose from the Inhalation and Ingestion of Transuranium Elements. The radiation dose equations developed herein are predicated on the basic assumptions and information presented in three publications of the International Commission of Radiological Protection (ICRP). The radiation dose to internal organs due to the ingestion of contaminated material was evaluated using the equations documented in ICRP Publication 2 (Ref. 35). The lung compartment model proposed by the Task Group on Lung Dynamics of the ICRP[38] and its revision stated in ICRP Publication 19 (Ref. 27) formed the basis for the derivation of the equations required to estimate the organ doses due to the inhalation of radioactive airplumes.

For the ingestion route, equations were formulated incorporating the continuous, as well as the short term, intake of contaminated sources of the diet. The most probable sources of ingestion of the transuranium elements for this study include:

(1) Agricultural produce contamination
   a. Aerial deposition resulting from airborne releases during an explosion-fire in a waste area
   b. Use of irrigation water derived from a contaminated source
   c. Uptake of radioactivity from contaminated soil
(2) Cattle and sport game grazing on contaminated forage or vegetation
(3) Contaminated potable water supplies, either surface or groundwater supplies
(4) Aquatic animals living in a contaminated aqueous environment.

The two pathways of exposure will be considered in the evaluation of the organ

doses due to the inhalation of the transuranium elements: direct inhalation of a radioactive plume caused by an explosion-fire in a waste area, and inhalation of resuspended soil particles which have been previously contaminated by an accidental spill or by ground deposition of airborne material released in a fire or by any other catastrophic event.

The internal organs of concern for this study shall be the bone, lung, liver, GI tract, and kidneys. This selection is based upon the ICRP's selection of these critical organs for the intake of the transuranium elements.[35] For the ingestion of the transuranic elements, the radiation dose to the bone, liver, GI tract, and kidneys will be evaluated. Since a major fraction of the material deposited in the lungs is biologically cleared to the blood with subsequent translocation to other internal organs, the radiation dose to the bone, liver, GI tract, and kidneys, as well as the lung, will be estimated for inhalation. For this study, the hilary lymph nodes will not be considered as a critical organ since the main commission of the ICRP has recommended that lymphatic tissue should not be considered the critical organ in inhalation exposures to plutonium.[27]

The important parameters required to evaluate or reduce the dose equations have been presented in the Appendix. The parameters include organ masses, effective organ elimination rate constants for each nuclide, effective absorbed energy per disintegration for each nuclide and organ, fractional amount of the nuclide ingested which is transferred to the organ of interest, the fractional amount in the GI tract transferred to the blood, and the fractional amount in the blood transferred to the organ of interest.

a. Equations Related to Ingestion

(1) Continuous or Chronic Ingestion. The amount of radioactivity in the organ of interest at any time t, subsequent to the start of the ingestion period, can be found by Eq. (12).

$$q_c(t) = \frac{Pf_w}{\lambda_E}\left(1 - e^{-\lambda_E t}\right) \qquad (12)$$

where $q_c(t)$ is the amount of radioactivity (μCi) in the organ at time t, f_w is the fraction of the radioactive material ingested reaching the organ, P is the ingestion rate (μCi/day), which is considered constant, and λ_E is the effective elimination rate constant (day^{-1}) for the organ. The ingestion rate P can be determined by knowing the radioactivity concentration in the food or water and the daily dietary intake (obtained from areal surveys).

The total dose received from the radioactive material deposited in an organ is directly related to the time integral of radioactivity in the organ:

$$\text{Dose (rem)} = \frac{51.2\varepsilon}{m}\int_0^t q_c(t)\,dt \qquad (13)$$

which yields

$$\text{Dose (rem)} = \frac{51.2\varepsilon P f_w}{m\lambda_E}\left[t + \frac{e^{-\lambda_E t} - 1}{\lambda_E}\right] \qquad (14)$$

where 51.2 is a constant related to unit conversion (gram-rad-disintegration/MeV-Ci-day); m is the mass of the organ (grams); $\varepsilon = \Sigma EF(RBE)n$, the effective absorbed energy per disintegration (MeV-rem/disintegration-rad); and t is the time interval (days) of ingestion for which the dose is estimated. Equation (14) is only applicable in the case where the intake rate is a constant and, thus, has very limited use in considering environmental contamination of most dietary sources. One possible application could be the evaluation of the dose resulting from the long-term ingestion of contaminated groundwater supplies.

(2) Acute Ingestion. In the case where ingestion occurs for a finite period

in a person's life ending at time t_1, the radiation dose incurred during the ingestion period and some subsequent internal exposure period ending at t_2, can be evaluated by

$$\text{Dose (rem)} = \frac{51.2\varepsilon P f_w}{m\lambda_E}\left[\int_0^{t_1}\left(1-e^{-\lambda_E t}\right) dt + \int_{t_1}^{t_2}(1-e^{-\lambda_E t_1})\, e^{-\lambda_E t}\, dt\right] \quad (15)$$

which yields

$$\text{Dose (rem)} = \frac{51.2\varepsilon P f_w}{m\lambda_E}\left[t_1 + \frac{e^{-\lambda_E t_1} - 1}{\lambda_E}\right] + \frac{51.2\varepsilon P f_w}{m\lambda_E^2}(1 - e^{-\lambda_E t_1})(1 - e^{-\lambda_E t_2}) \quad (16)$$

where t_1 is the length of time of continuous ingestion (days) and t_3 is the length of time after the termination of the ingestion period for which the dose shall be evaluated (days). Normally, the radiation dose is evaluated for 50 years; therefore, $t_1 + t_3 = 50$ years.

A somewhat more convenient method of determining the dose to an organ from ingestion is the application of dose commitment factors. The dose commitment factor stems from the concept of "dose commitment," the total dose received by an individual during the lifetime due to the intake of radioactivity for a specified length of time. In the case of the transuranium elements, which are tenaciously retained in most of the body organs under consideration, one must evaluate the total dose to the organs during the lifetime of the person or during the time period which the radionuclide is present in the organ. Normally, for acute ingestion periods, the dose commitment is stated for 50 years. The dose commitment factor is the dose resulting from the intake of one microcurie of radioactivity, evaluated over a certain length of time. The equation formulated to calculate the dose commitment factor is

$$\text{D.C. (rem/}\mu\text{Ci) ingestion} = \frac{51.2\varepsilon f_w(1-e^{-\lambda_E t})}{m\lambda_E} \quad (17)$$

where the terms ε, f_w, m, and λ_E are the same as previously defined and t is the time period over which the dose is estimated (normally 50 years for short ingestion periods).

For chronic ingestion, where the ingestion rate may vary due to source term fluctuations, the use of the dose commitment factors is somewhat more complicated. If the ingestion period is 10 years, one must sum the dose values obtained from multiplying the dose commitment factors for exposure periods of 50, 49, 48,...41, 40 years by the total amount of radioactivity ingested during the 1st, 2nd, 3rd,... 9th, 10th years, respectively, in order to evaluate the 50-year dose. This procedure appears to be rather tedious if the ingestion period is long, but employment of a computer for data reduction simplifies the calculations. Since the radioactive source term for most of the dietary inputs considered in this study will vary with time, the use of the dose commitment factor for the various exposure periods will most likely be employed to derive the 50-year dose from the ingestion route.

(3) Gastrointestinal Tract Considerations. The lower large intestine of the GI tract is the critical organ for the ingestion of the soluble uranium compounds,[35] and is the organ, other than the lung, receiving the greatest dose from the inhalation of insoluble plutonium and americium. Due to the

relatively short transit time associated with material passing through the GI tract (on the order of days), the use of a 50-year dose commitment value for this organ is not applicable. Since the isotopes of uranium, plutonium, and americium have very long half-lives, the dose to the lower large intestine (LLI) would be received in the time period known as the mean transit time of passage through the LLI. The mean transit time of passage has been estimated by ICRP 2 and later revised by Eve[39] as 0.75 and 1.0 day, respectively.

The radiation dose to the GI tract (LLI) can be evaluated by the equation below:

$$\text{Dose (rem/}\mu\text{Ci)} = \frac{25.6 f_L \varepsilon \tau}{m} \qquad (18)$$

where f_L is the fraction of the material ingested reaching the LLI (which equals 1 - fractional amount removed by the small intestine, f_1), and τ is the transit time (day) through the LLI. In this study, the transit time recommended by Eve[39] will be employed in the dose calculations. The term 25.6 originates from the fact that the dose is distributed over the surface layer of the LLI (that is, 1/2 of 51.2). The mass of the contents of the LLI (m) has been documented as 135 g (Ref. 39).

b. Equations Related to Inhalation

(1) Basic Lung Model. The lung models utilized to formulate the radiation dose equations were proposed by the ICRP Task Group on Lung Dynamics[38] in 1966 and have been modified in ICRP Publication 19 (Ref. 27). Publication 19 deals specifically with the metabolism of the compounds of plutonium and other actinides, and much of the information presented in the text has been incorporated in the basic lung models and forms the basis for the formulation of the radiation dose equations.

The assessment of the radiation dose to the lung and the other internal organs from the inhalation of radioactive material is complicated by the fact that the deposition and retention of respirable material are influenced significantly by the particle size and chemical form of the material as well as the individual's breathing rate. All of these parameters have been dealt with in the newly proposed lung model of the ICRP, but there is still some question concerning the solubility of many of the radioactive compounds.

The basic lung models, depicted as flow diagrams, have been presented as Figs. 7 and 8. For the purposes of modeling, many of the specific details of the lung model have been eliminated in this discussion, but the information is readily available from Refs. 27 and 38. Basically, the lung model is delineated into three physiological lung compartments depicting both deposition and clearance processes. The compartments are defined as follows:

1. Nasopharynx (N-P) - This compartment is commonly referred to as the upper respiratory tract. It begins with the anterior nares, through the anterior pharynx to the level of the larynx or epiglottis.
2. Trachea and Bronchial Tree (T-B) - Anatomically this section includes the trachea and bronchial tree down to and including the terminal bronchioles.
3. Pulmonary Compartment (P) - This region incorporates the respiratory bronchioles, alveolar ducts, atria, alveoli, and alveolar sacs.

In the models presented as Figs. 7 and 8, D_1 is the total amount of material inhaled, D_2 is the material in the exhaled air, and D_3, D_4, and D_5 represent the amounts of material deposited in the N-P, T-B and pulmonary regions, respectively. Ordinarily, D_3, D_4, and D_5 are expressed in fractional amounts of D_1. The retention or deposition of material in the three compartments is dependent upon the activity, median aerodynamic diameter of the

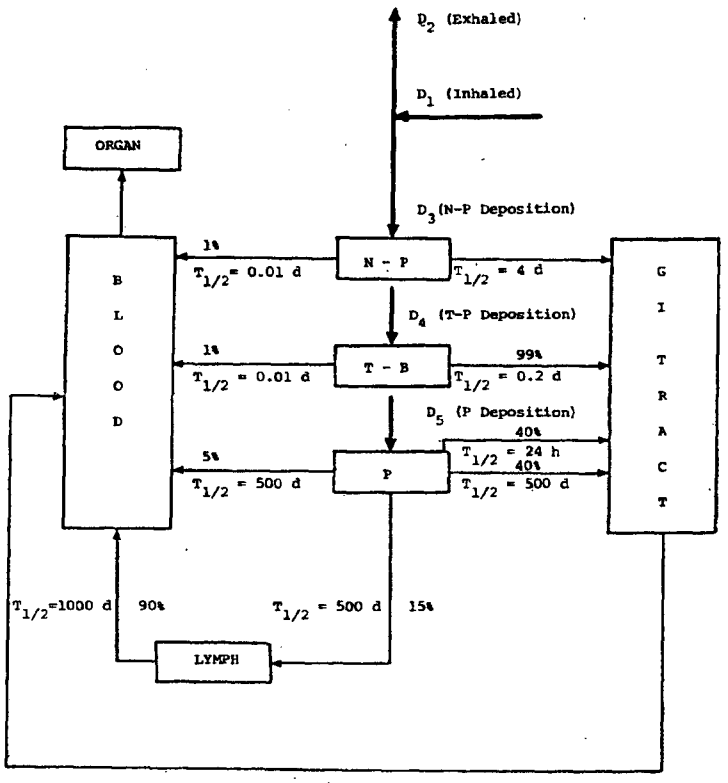

Fig. 7. Schematic Model of the Lung Deposition and Clearance Processes for the Transuranium Elements Falling in the Class Y Compound Category.

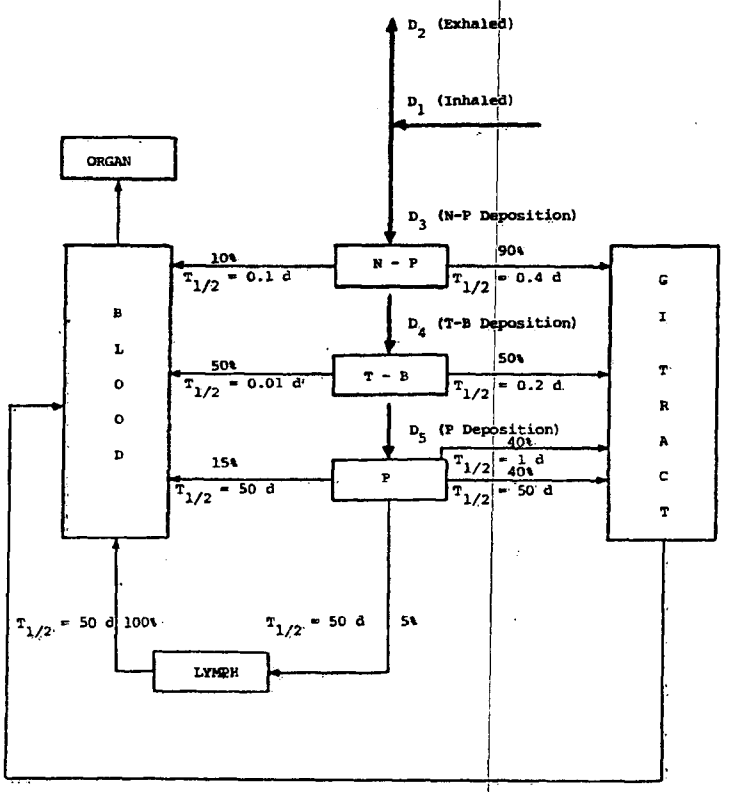

Fig. 8. Schematic Model of the Lung Deposition and Clearance Processes for the Transuranium Elements Falling in the Class W Compound Category.

particles inhaled, and the breathing rate of the individual. Figure 9 can be used to obtain the deposition in the nasopharyngeal and pulmonary compartments (D_3 and D_5) for any given particle size assuming a moderate breathing rate of 20 liters per minute. The deposition in the tracheobronchial compartment, D_4, is assumed to be constant at 8% of the amount inhaled, D_1, irrespective of particle size.

Two different lung models have been presented because the clearance processes and retention of deposited material for the various compartments vary according to the chemical nature of the radioactive compound inhaled. The Task Group on Lung Dynamics classified the various compounds by retention time in the lung. The majority of the compounds of plutonium and the other actinides fall into Class Y (retained for years) or Class W (retained for weeks). Carbides, oxides, hydroxides, and lanthanide fluorides are specified as Class Y, whereas nitrates, lanthanide halides, and carbonates fall into Class W.

Inspection of the transport rates and biological processes of the lung models reveals that the majority of the material reaching the blood is directly transported, although a portion of the material reaching the blood originates from the lung to GI tract clearance mechanisms. In addition, a major fraction of the material in the lung is translocated from the lung within five days after deposition.

(2) Radiation Dose to the Lung Following an Acute Inhalation Exposure. Close examination of the lung models presented indicates that the material deposited in the nasopharynx and tracheobronchial compartments is cleared very rapidly and thus, would represent a very small fraction of the time-integrated amount of radioactivity in the lung. Therefore, the main lung compartment of interest giving rise to the lung dose would be the pulmonary compartment. According to the models presented,

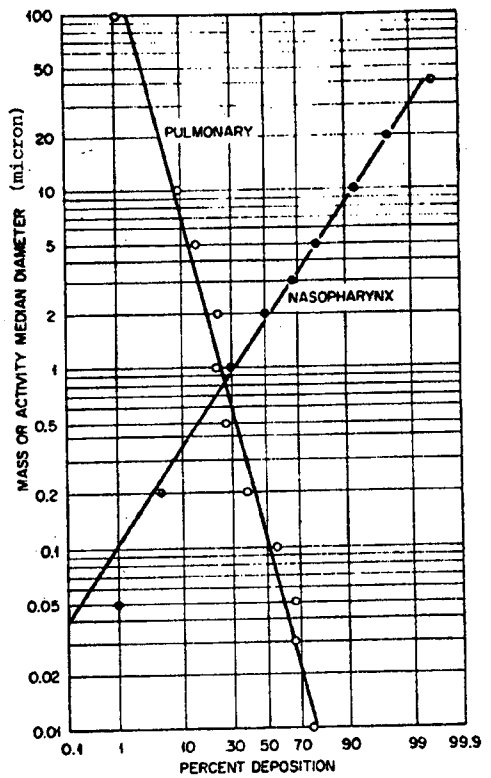

Fig. 9. The Deposition Estimate for the Nasopharyngeal and Pulmonary Compartments While Breathing at 20 Liter/Min (Taken from Ref. 40).

60% of the Class Y material deposited in the pulmonary compartment will be eliminated with a 500-day half-life, whereas 60% of the Class W material in the pulmonary region will be cleared with a 50-day half-life. The remaining 40% of either material (Class Y or W) deposited in this compartment is cleared to the GI tract with a 24-hour half-life.

If one considers only the pulmonary compartment for long-term retention, then the amount of radioactivity in the lung at any time, t, following an acute exposure is:

$$q_L(t) = f_p f_L q_o \, e^{-\lambda_L t} \qquad (19)$$

where f_p is the fractional amount of the inhaled radioactivity (q_o) deposited in the

pulmonary compartment, f_L is the fractional amount of the deposited material undergoing long-term retention, and λ_L is the effective elimination rate (1.39×10^{-3} day^{-1} for Class Y compounds and 1.39×10^{-2} day^{-1} for Class W compounds). The fractional amount of the inhaled material deposited in the pulmonary compartment is a function of particle size and can be estimated from Fig. 9. The total amount of radioactive material inhaled (q_o) during the exposure period is determined by:

$$q_o = B\ C\ t (\mu Ci) \qquad (20)$$

where B is the breathing rate (0.02 m^3/min),
C is the average plume concentration ($\mu Ci/m^3$),
t is the exposure interval (min).

Assuming a lung mass of 1 000 g and an effective absorbed energy of ε, the radiation dose to the lung can be determined by:

$$\text{Dose (rem)} = \frac{0.0512 \varepsilon f_p\ f_L q_o (1-e^{-\lambda_L t})}{\lambda_L} \qquad (21)$$

where t is the time interval (days) over which the dose is evaluated. The dose commitment factor for the lung would be

$$\text{D.C. (rem/}\mu\text{Ci)} = \frac{0.0512 \varepsilon f_p f_L (1-e^{-\lambda_L t})}{\lambda_L} . \qquad (22)$$

(3) General Radiation Dose Equations for Organs Other Than the Lung and the GI Tract.

The radiation dose to the bone, liver, and kidneys from the inhalation of the transuranium elements can be evaluated by relating the amount of radioactivity in these organs originating from the various lung clearance processes. The best approach in developing the necessary equations for such clearance processes is to determine the amount of material translocated from the lung compartments to the blood since a certain fraction of the material in the systemic system is transferred to the organs of interest. According to the models presented in Figs. 7 and 8, there are two clearance processes by which material in the nasopharynx and tracheobronchial regions reaches the lung. These are (1) direct transfer to the blood, and (2) transfer to the GI tract with a fractional amount of this material subsequently being transferred to the blood. For the pulmonary compartment, three processes are involved: the two processes previously mentioned for the other two lung compartments and the clearance from the lymph nodes.

Basically, the dose equations for Class Y materials can be developed accordingly: Rate of change in the number of microcuries in organ (dq_c/dt) = Input Rate - Elimination Rate, where the input rate is the cumulative rate of all compartments translocating material to the blood, either directly or indirectly.

$$\text{Input Rate} = \underbrace{f_1 f_2\ 0.99\ q_{N-P}\ e^{-\lambda_{N-P_1} t}\ \lambda_{N-P_1} + f_2\ 0.01\ q_{N-P}\ e^{-\lambda_{N-P_2} t}\ \lambda_{N-P_2}}_{\text{N-P Region Input}}$$

$$\underbrace{+ f_1 f_2\ 0.99 q_{T-B}\ e^{-\lambda_{T-B_1} t}\ \lambda_{T-B_1} + f_2\ 0.01 q_{T-B} e^{-\lambda_{T-B_2} t}\ \lambda_{T-B_2}}_{\text{T-B Region Input}}$$

$$+ f_1 f_2\ 0.40\ q_P\ e^{-\lambda_{P_1} t}\ \lambda_{P_1} + f_1 f_2\ 0.40\ q_P\ e^{-\lambda_{P_2} t}\ \lambda_{P_2} + f_2\ 0.05 q_P\ e^{-\lambda_{P_3} t}\ \lambda_{P_3}$$

P Region Input

$$+ \frac{f_2 0.135 q_P \lambda_{PN}}{\lambda_N - \lambda_{PN}} \left(e^{-\lambda_{PN} t} - e^{-\lambda_N t}\right) \lambda_N$$

Lymph Node Input (23)

$$\text{Elimination Rate} = -q_c\ \lambda_E \qquad (24)$$

where

f_1 is the fractional amount transferred from the GI tract to the blood,

f_2 is the fractional amount transferred from the blood to the organ,

q_{N-P}, q_{T-B}, q_p are the initial amounts of material deposited in the nasopharynx (N-P), tracheobronchial (T-B), pulmonary (P) regions, respectively,

λ_{N-P_1} is the biological elimination rate constant for the N-P region to GI transfer (day^{-1}),

λ_{N-P_2} is the biological elimination rate constant for the N-P region to blood transfer (day^{-1}),

λ_{T-B_1} is the biological elimination rate constant for the T-B region to GI transfer (day^{-1}),

λ_{T-B_2} is the biological elimination rate constant for the T-B region to blood transfer (day^{-1}),

λ_{P_1} is the biological elimination rate constant for the P region to GI transfer describing the short-term component (day^{-1}),

λ_{P_2} is the biological elimination rate constant for the P region to GI transfer describing the long-term component (day^{-1}),

λ_{P_3} is the biological elimination rate constant for the P region to blood transfer (day^{-1}),

λ_{PN} is the biological elimination rate constant for the P region to lymph node transfer (day^{-1}),

λ_N is the biological elimination rate constant for the lymph node to blood transfer (day^{-1}),

q_c is the amount of radioactivity in the organ of interest,

λ_E is the effective elimination rate constant for the organ of interest (day^{-1}).

Note that all organ input rate constants from the various lung compartments were defined in terms of the biological elimination rate constants. In the case of long-lived transuranium elements, the effective elimination rate constants for the lung model are essentially equivalent to the biological elimination rate constants.

The amount of radioactivity in the organ of interest at any time, t, following lung deposition can be determined by combining Eqs. (23) and (24) and integrating.

$$q_c(t) = \frac{K_1\lambda_{N-P_1}(e^{-\lambda_{N-P_1}t} - e^{-\lambda_E t})}{\lambda_E - \lambda_{N-P_1}} + \frac{K_2\lambda_{N-P_2}(e^{-\lambda_{N-P_2}t} - e^{-\lambda_E t})}{\lambda_E - \lambda_{N-P_2}}$$

$$+ \frac{K_3\lambda_{T-B_1}(e^{-\lambda_{T-B_1}t} - e^{-\lambda_E t})}{\lambda_E - \lambda_{T-B_1}} + \frac{K_4\lambda_{T-B_2}(e^{-\lambda_{T-B_2}t} - e^{-\lambda_E t})}{\lambda_E - \lambda_{T-B_2}}$$

$$+ \frac{K_5\lambda_{P_1}(e^{-\lambda_{P_1}} - e^{-\lambda_E t})}{\lambda_E - \lambda_{P_1}} + \frac{K_6\lambda_{P_2}(e^{-\lambda_{P_2}t} - e^{-\lambda_E t})}{\lambda_E - \lambda_{P_2}}$$

$$+ \frac{K_7\lambda_{P_3}(e^{-\lambda_{P_3}t} - e^{-\lambda_E t})}{\lambda_E - \lambda_{P_3}} + \frac{K_8\lambda_{PN}\lambda_N(e^{-\lambda_{PN}t} - e^{-\lambda_E t})}{(\lambda_E-\lambda_{PN})(\lambda_N-\lambda_{PN})} \qquad (25)$$

$$- \frac{K_8\lambda_{PN}\lambda_N(e^{-\lambda_N t} - e^{-\lambda_E t})}{(\lambda_N-\lambda_{PN})(\lambda_E-\lambda_N)}$$

where $K_1 = f_1 f_2\ 0.99 q_{N-P}$

$K_2 = f_2\ 0.01 q_{N-P}$

$K_3 = f_1 f_2\ 0.99 q_{T-B}$

$K_4 = f_2\ 0.01 q_{T-B}$

$K_5 = f_1 f_2\ 0.40 q_P$

$K_6 = f_1 f_2\ 0.40 q_p$

$K_7 = f_2\ 0.05 q_p$

$K_8 = f_2\ 0.135 q_p$.

When the equation is evaluated for the time measured in days, and since $\lambda_E << \lambda_{N-P_2}$, λ_{T-B_1}, λ_{T-B_2}, Eq. (25) reduces to

$$q_c(t) = \frac{K_1\lambda_{N-P_1}(e^{-\lambda_{N-P_1}t} - e^{-\lambda_E t})}{(\lambda_E - \lambda_{N-P_1})} + (K_2+K_3+K_4)e^{-\lambda_E t}$$

$$+ \frac{K_5\lambda_{P_1}(e^{-\lambda_{P_1}t} - e^{-\lambda_E t})}{\lambda_E-\lambda_{P_1}} + \frac{K_6\lambda_{P_2}(e^{-\lambda_{P_2}t} - e^{-\lambda_E t})}{\lambda_E-\lambda_{P_2}}$$

$$+ \frac{K_7\lambda_{P_3}(e^{-\lambda_{P_3}t} - e^{-\lambda_E t})}{\lambda_E-\lambda_{P_3}} + \frac{K_8\lambda_{PN}\lambda_N(e^{-\lambda_{PN}t} - e^{-\lambda_E t})}{(\lambda_E-\lambda_{PN})(\lambda_N-\lambda_{PN})}$$

$$- \frac{K_8\lambda_{PN}\lambda_N(e^{-\lambda_N t} - e^{-\lambda_E t})}{(\lambda_E-\lambda_N)(\lambda_N-\lambda_{PN})} \quad . \tag{26}$$

The dose to the organs of interest for any time interval, t (in days), following a short-term inhalation can be found by:

$$\text{Dose (rem)} = \frac{51.2\varepsilon\int_0^t q_c(t)}{m}\,dt \tag{27}$$

where the constant 51.2 and ε and m are the same as previously defined.

$$\text{Dose(rem)} = \frac{51.2\varepsilon}{m}\left[\frac{K_1(1-e^{-\lambda_{N-P_1}t})}{\lambda_E - \lambda_{N-P_1}} - \frac{K_1\lambda_{N-P_1}(1-e^{-\lambda_E t})}{\lambda_E(\lambda_E-\lambda_{P_1})}\right.$$

$$\frac{(K_2+K_3+K_4)(1-e^{-\lambda_E t})}{\lambda_E} + \frac{K_5(1-e^{-\lambda_{P_1}t})}{\lambda_E-\lambda_{P_1}} - \frac{K_5\lambda_{P_1}(1-e^{-\lambda_E t})}{\lambda_E(\lambda_E-\lambda_{P_1})}$$

$$+ \frac{K_6(1-e^{-\lambda_{P_2}t})}{\lambda_E-\lambda_{P_2}} - \frac{K_6\lambda_{P_2}(1-e^{-\lambda_E t})}{\lambda_E(\lambda_E-\lambda_{P_2})} + \frac{K_7(1-e^{-\lambda_{P_3}t})}{\lambda_E-\lambda_{P_3}} - \frac{K_7\lambda_{P_3}(1-e^{-\lambda_E t})}{\lambda_E(\lambda_E-\lambda_{P_3})}$$

$$+ \frac{K_8\lambda_N(1-e^{-\lambda_{PN}t})}{(\lambda_E-\lambda_{PN})(\lambda_N-\lambda_{PN})} - \frac{K_8\lambda_{PN}\lambda_N(1-e^{-\lambda_E t})}{\lambda_E(\lambda_E-\lambda_{PN})(\lambda_N-\lambda_{PN})} - \frac{K_8\lambda_{PN}(1-e^{-\lambda_N t})}{(\lambda_N-\lambda_{PN})(\lambda_E-\lambda_N)}$$

$$\left. + \frac{K_8\lambda_{PN}\lambda_N(1-e^{-\lambda_E t})}{\lambda_E(\lambda_N-\lambda_{PN})(\lambda_E-\lambda_N)}\right] \quad . \tag{28}$$

For Class W compounds the same equation can be used to estimate the dose, but the constants K_1 through K_8 must be changed to reflect the differences in the fractional amount of material cleared from the various compartments to the GI tract, blood, and lymph. The appropriate constants are stated below:

$$K_1 = f_1 f_2\ 0.90\ q_{N-P}$$
$$K_2 = f_2\ 0.10\ q_{N-P}$$
$$K_3 = f_1 f_2\ 0.50\ q_{T-B}$$
$$K_4 = f_2\ 0.50\ q_{T-B}$$
$$K_5 = f_1 f_2\ 0.40 q_p$$
$$K_6 = f_1 f_2\ 0.40 q_p$$
$$K_7 = f_2\ 0.15 q_p$$
$$K_8 = f_2\ 0.05 q_p\ .$$

G. Radionuclide Inventory of the Waste Disposal Areas at LASL

1. Introduction. An essential portion of the risk analysis is a determination of the type and quantities of radionuclides in existing disposal areas. Data on the transuranics is necessary to evaluate the concentration of the various nuclides in the waste areas. Analysis of the hazards associated with exhumation of the waste must consider all radionuclides that would be encountered in such an operation.

A preliminary survey was made of the radionuclide content and composition of solid wastes disposed of at Los Alamos since the beginning of activity at the Laboratory in the early 1940's. Records of solid waste disposal are highly variable in quality. In general, no records were kept until the mid 1950's. Detailed records as to content and composition of wastes were not kept until 1959. The quality of records has improved steadily since that time. Information on waste disposal is available in several forms. Daily entries in logbooks form the basis of most information; numerous memoranda and other file information are also available. Lack of manpower and time has so far prevented direct use of this information. Extensive use has been made of material accountability records for inferring the amounts of certain nuclides disposed of prior to the initiation of detailed record keeping. Monthly and biannual summaries of the daily logs prepared by waste management personnel provided the bulk of the information presented in this report. The completeness of this inventory is largely limited by that of the summaries.

A large variety of radioactive waste types have been disposed of at Los Alamos. The bulk of the material is room-generated trash, generally contained in cardboard boxes or wooden crates. A large variety of special disposal operations have been performed, ranging from the shaft disposal of a a cylinder containing a few grams of tritium-contaminated solution to the demolition and burial of entire buildings. During the early years of the Laboratory, liquid wastes were disposed of directly to the ground. In later years these liquids or sludges resulting from waste treatment were placed in drums for burial. Liquid wastes containing americium and plutonium contamination were mixed with cement and disposed of in drums or directly into disposal shafts. An attempt has been made to delineate the various types of disposal used for the material included in this inventory, for example, disposal shaft, pit, and absorption bed. Confirmation of the delineation will require completion of a detailed examination of the daily logs presently in process and is expected to last several months.

2. Location of Waste Disposal Areas. In general, two types of disposal operations have been conducted at Los Alamos. In the first several years of operations of the Laboratory, expediency dictated rapid and, by present standards, casual disposal of contaminated wastes. These wastes were generally quite low in activity. The technical areas generating radioactive waste operated their own burial areas, and no

Laboratory-wide record keeping or supervision existed. The reduction in research pressure following the war resulted in an increased awareness and concern for the adequacy of disposal techniques. A waste disposal section was organized, and with it were created various organized burial areas. In addition to areas receiving Laboratory-wide generated waste, several areas have been used for special or one-time disposal operations. The various burial operations known to contain radioactive material are indicated on Fig. 10. The letter designation is one utilized by the Waste Management Section for reference convenience and does not necessarily convey any information about the chronology of use. Each area will be discussed separately in the following sections.

3. Radionuclide Inventory of Specific Burial Areas.

a. Area A. This was the first disposal area receiving waste under

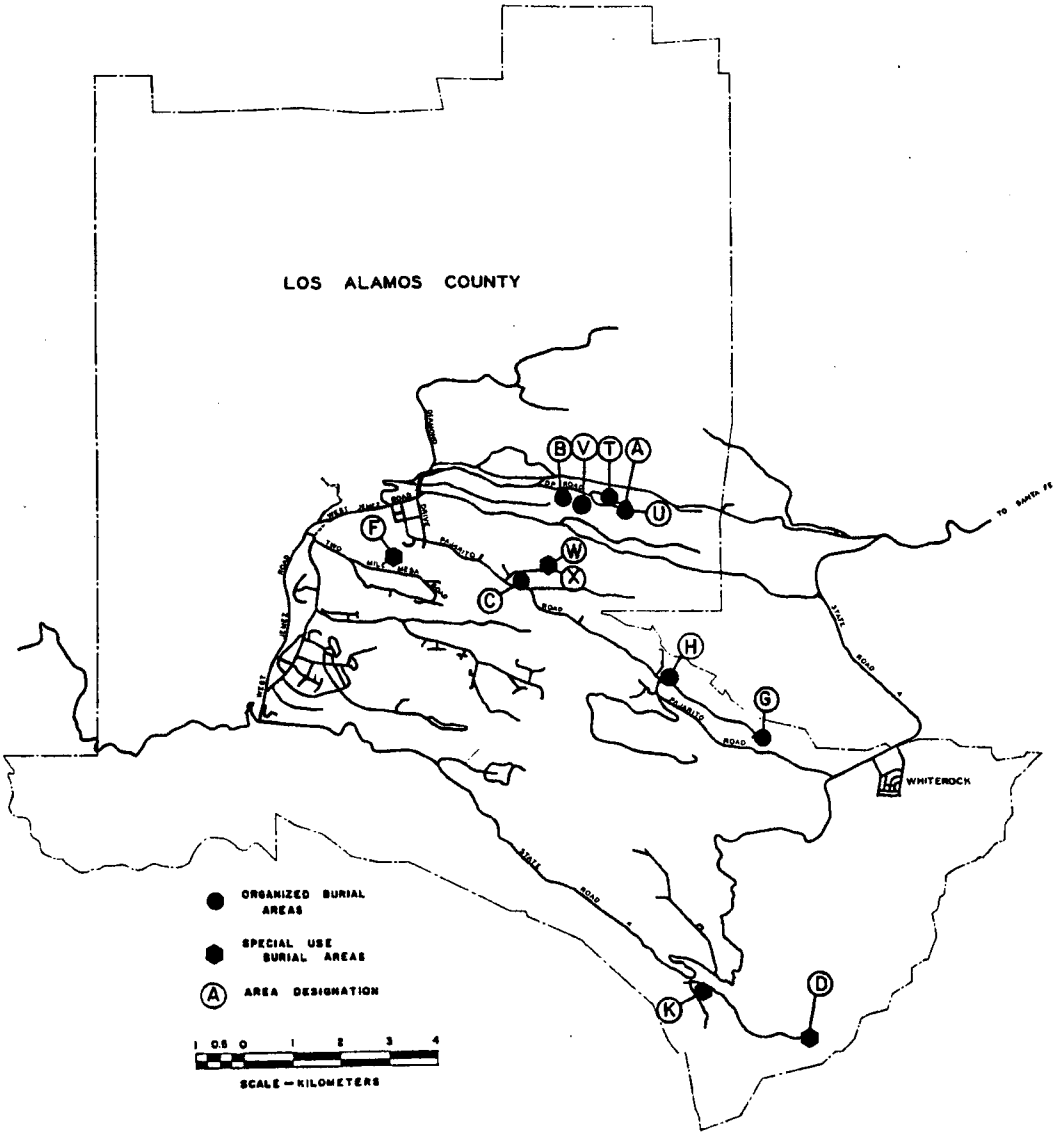

Fig. 10. Location of Radioactive Waste Disposal Areas at LASL.

the supervision of an organized waste disposal section. It was operated from 1945 to 1946. Waste was placed in pits excavated in the volcanic tuff, and the material covered with the crushed material removed from the pit. No records were kept of the types and volumes of waste placed in these pits. However, it is known that little, if any, transuranic material was placed in these pits.[41] Plutonium was an extremely scarce material during the years this area was used, and every effort was made to recover it from waste material. This area undoubtedly contains trace quantities of many of the radionuclides present in fission products and irradiated material that are known to exist in other disposal areas, but it is not presently possible to identify them specifically. The pits in Area A were also used for the disposal of chemical wastes.[41]

b. Area B. This area was opened in 1946, and its use continued through 1948. It was operated in the same manner as Area A, utilizing trenches for burial of all waste, including some chemical materials such as gas cylinders of HCl, H_2S and HF (Ref. 41). No records exist on the types or volume of material placed in this area. The general inferences concerning radionuclides in Area A apply also to Area B. Concern over the proximity of waste disposal operations to residential areas prompted the opening of a new disposal area and the closing of Area B.

c. Area C. This area was first opened in 1948. Trench disposal techniques were utilized for room-generated trash, with separate pits used for radioactive and chemical wastes. Wastes containing unusual quantities of activity were placed in vertical shafts, varying from 6-20 m deep. A total of six pits existed in this area, all of which were filled by late 1959. Use of the disposal shafts in this area continued through 1969.

Records of the types and volumes of waste placed in Area C prior to 1950 are incomplete. Beginning in 1951 data were recorded on the curie content of sludge generated by the liquid treatment facility and buried in this area.[42] The curie content of some isotopes placed in this area can be inferred from material accountability records, which indicate the amount of various materials removed from the inventory and considered to have been disposed of as waste.[43] Some of this material was released in liquid effluents, and some escaped to the atmosphere. Thus, these records only provide an upper limit on the material actually placed in the disposal area.

The first complete records of curie content and composition of wastes placed in Area C begin in 1960 and apply only to the material placed in disposal shafts. However, the summary records used for determining the content and composition of the wastes were not specific as to the shaft used for disposal. Beginning in 1965 shafts were in use in both Area C and Area G, and this dual usage continued through 1969. An estimate has been made of the amount of material placed in shafts in the respective areas during this time. The data on activity and nuclide composition of wastes in pits in Area C are presented in Table XIV, and those on the wastes in shafts in Area C are summarized in Table XV.

d. Area D. This disposal area is located within Technical Area 33. Radioactive wastes generated by weapons testing were disposed of locally due to the site's remoteness from the remainder of the Laboratory. Area D contains several shafts approximately 15 m deep that were used for subsurface detonation of explosives containing ^{238}U and ^{210}Po. The shafts were last used in 1949 and are known to be contaminated with ^{238}U. One shaft was opened for inspection in 1954 and then backfilled with crushed tuff. In addition to the shafts, this area contains several pits known to contain kilogram quantities of ^{238}U (Ref. 47). No accurate estimate of

TABLE XIV

RADIONUCLIDE CONTENT OF MATERIAL PLACED IN DISPOSAL PITS

AND

ABSORPTION BEDS, AS OF JANUARY 1973[a]

	Pits		Absorption Beds		
	C	G	T	U	V
Isotope	1958-1959	1959-1972	1945-1951	1945-1968	1945-1961
^{3}H		4[e]			
^{90}Sr-^{90}Y		2 962[f]			<0.1
^{227}Ac				1	
U[b]	25[d]	48[d]			
^{238}Pu		15[c]			
^{239}Pu	26[c,d]	332[d]	10[e]		<0.1[e]
^{241}Am	149[c]	2 074[c]			
TOTAL	196	6 468	10	1	<0.1

[a]All values in curies, decay corrected

[b]Includes isotopes 234, 235, 236, 238

[c](Ref. 42)

[d](Ref. 43)

[e](Ref. 44)

[f](Ref. 45)

the curie content of these wastes can be made at this time.

e. Area F. This disposal area is located on Two Mile Mesa and was used once in 1946 for local disposal of wastes prior to the organization of a Laboratory-wide disposal section. Wastes were placed in shallow pits or trenches, but information on the types and quantities of radionuclides disposed of is generally not available. A memo dated February 13, 1964, reports on the presence of an estimated 30 mCi of ^{137}Cs within this disposal area.[48]

f. Area G. This is the primary disposal area used by Los Alamos Scientific Laboratory since 1959. The area contains six large pits (30 m x 180 m x 8 m) and eight smaller pits of varying dimensions, all of which have been used for routine burial of Laboratory-generated radioactive wastes. The records describing the material placed in these pits generally do not contain information on the curie content of the

TABLE XV

RADIONUCLIDE CONTENT OF MATERIALS PLACED IN SUBSURFACE DISPOSAL

AS OF JANUARY 1973[a]

Isotope	Disposal Shafts C(46) 1960-1969	Disposal Shafts G(46) 1965-1972	Disposal Shafts T(44) 1968-1972	Total in Pits and Absorption Beds, Table XIV	Total - All Disposal Areas
^{3}H	1400	98 449		4	99 853
^{22}Na	40	46			86
^{60}Co	20	229			249
^{90}Sr-^{90}Y	31	306	2	2 962	3 301
^{137}Cs		6	2		6
^{233}U	5	5	7		17
U[b]	<0.1			73	73
^{238}Pu		4	13	15	32
^{239}Pu			70	368	438
^{241}Am			1 256	2 223	3 479
Fission Products	50	77			127
Induced Activity	200	2 458			2 658

[a]All values in curies, decay corrected

[b]Includes isotopes 234, 235, 236, 238

material, but the isotopic composition is generally indicated. Uranium- and plutonium-contaminated wastes are placed in separate pits. Americium-241 is known to be present in the pits, occurring in association with plutonium in drums of sludge generated by liquid treatment facilities.[42] A reliable estimate can be made of the curie content of the various isotopes using material accountability data.[43] Other radioisotopes, such as tritium, are known to be present in the disposal pits in unknown quantities.[37] A summary of the available data on activity and nuclide content of wastes in disposal pits in Area G is presented in Table XIV.

In addition to the pits, Area G contains over 70 disposal shafts used for burial of intermediate and low-level contaminated wastes. Records on the types and activity of these wastes are generally quite good.[46] A summary of the known

TABLE XVI

MINOR NUCLIDES IN AREA G DISPOSAL SHAFTS

^{24}Na	^{91}Y	^{144}Ce	^{227}Ac
^{32}P	^{105}Ag	^{147}Pm	^{232}Th
^{51}Cr	^{114}In	^{152}Eu	^{240}Pu
^{57}Co	^{131}I	^{182}Ta	^{242}Pu
^{59}Fe	^{133}Xe	^{191}Au	^{244}Cm
^{65}Zn	^{140}Ba	^{210}Po	^{252}Cf
^{85}Kr			

activities of material in these shafts is presented in Table XIV. In addition to the isotopes listed in Table XV, unspecified but small quantities of the nuclides listed in Table XVI are also present in the disposal shafts.[46]

The data presented in Tables XIV and XV were obtained from biannual summaries prepared by the Waste Management Section. For the sake of completeness, a review of the original logbooks detailing the daily operations of the disposal areas is presently under way. It is expected that this review will result in some modifications to the information presented here. However, the record keeping and summaries thereof have been sufficiently complete that no significant changes are expected.

g. Area H. This area contains two shafts used for the disposal of contaminated classified material. The classified nature of the material has generally restricted the reporting of the composition and activity of the radionuclides present. However, the material is reported as being generally low level.[37]

h. Area K. This disposal area is located in close proximity to Area D and was operated similarly for the local disposal of wastes generated at Technical Area 33. Wastes containing an estimated several hundred kilograms of ^{238}U were placed in pits in this area between 1949 and 1955 (Ref. 47). In addition, a shallow pit in the area was used for disposal of tritium-contaminated solutions between 1950 and 1959. Septic tanks in the area have received liquid wastes contaminated with ^{235}U and ^{238}U. One additional septic tank received two emergency releases of plutonium-contaminated liquid in 1961. No estimate of the curie content of these various wastes has been made.

i. Area T. This area has been utilized for waste disposal in two different ways. From 1945 to 1951 covered absorption beds were used for subsurface disposal of liquid wastes resulting from recovery of plutonium. Beginning in 1968 liquid effluents were mixed with cement and placed in vertical shafts. The two disposal operations are discussed separately

in this section.

Absorption beds. Four pits 30 m long, 6 m wide, and approximately 2 m deep were dug in the tuff and partially backfilled with large gravel. Liquid wastes containing plutonium were discharged to these pits beginning in 1945. The use of the pits was discontinued in 1952 with the completion of a liquid waste treatment facility. Hydrofluoric acid used in the plutonium recovery operation is known to have been present in the waste discharge. Some tritium is also known to have been present.[47] A summary of the radioactivity discharged to the pits is presented in Table XIV (Ref. 44). When the use of the pits was discontinued, they were completely backfilled with earth and crushed tuff.

Disposal shafts. The operation of the liquid treatment facility at TA-21, near Area T, resulted in the production of a sludge residue contaminated with plutonium and americium. For many years this material was mixed with concrete and placed in steel drums for disposal at Area C and Area G. In 1968, the operation of a pug mill was instituted, which mixed the waste material with cement. This cement was then pumped directly into unlined vertical shafts approximately 20 m deep and 2 m in diameter. The shafts were augered into the tuff using a large diameter bucket auger. This practice continued through 1973, when techniques were developed for the disposal of the contaminated cement slurry in a fashion that would permit retrieval on a 20-year basis. Numerous radionuclides are known to be present in the contaminated slurry, among them ^{90}Sr, ^{238}Pu, ^{239}Pu and ^{241}Am (Ref. 49). A complete summary of the activity placed in the shafts through 1972 is presented in Table XV.

j. Area U. This area contains several absorption beds similar to those in Area T. The beds were used for subsurface disposal of contaminated liquid wastes between 1945 and 1968. The primary radionuclide present in these wastes was ^{210}Po. No records were kept of the amount discharged. However, the short half-life of the material (1.19×10^7 s) will have produced a decay of the material to innocuous levels by 1972. During 1953 approximately 2.5 curies of ^{227}Ac were discharged to the pits.[44] As indicated in Table XIV, a portion of this activity remained undecayed as of the end of 1973.

k. Area V. This area was used for the disposal of contaminated liquid waste resulting from laundry operations during the years 1945 to 1961, using absorption beds similar to those in Area T. An estimated total of three curies of ^{89}Sr, ^{140}Ba, and ^{140}La were present in the liquid discharged to the beds.[44] This activity had decayed to a very low level by the end of 1972. In addition, small quantities of ^{90}Sr and ^{238}Pu were contained in the waste.[44] The remaining non-decayed activity of these two contaminants is given in Table XIV.

l. Area W. This area is used for the subsurface storage of two coolant tanks associated with the LAMPRE reactor dismantled in 1963. Two stainless-steel tanks, each containing 110-150 liters of irradiated sodium, are encased in carbon-steel sleeves and located in separate vertical shafts about 30 m deep. The sodium is known to be contaminated with ^{58}Co, ^{137}Cs, ^{182}Ta, and ^{239}Pu (Ref. 50). The total activity present in the tanks is not known.

m. Area X. This site, in close proximity to Area W, is being used for the subsurface storage of the LAMPRE reactor vessels. The vessels were buried in 1964, containing an unspecified amount of ^{235}U. Other activation products are also expected to be present.

n. Summary. Records on the subsurface storage and disposal of radioactive waste are of varying quality. No complete records of waste volume, curie content, and composition were kept until 1960. After that date the records contain

information on the curie content of material placed in disposal shafts and on specialized material placed in disposal pits. Quantities of certain materials present in various disposal areas can be estimated from material accountability records.

The various disposal areas at Los Alamos contain a recorded decayed total of 107 046 Ci, of which nearly 99 000 Ci is tritium. Strontium-90 and ^{241}Am are the second and third largest contributors to the activity, while ^{239}Pu comprises a relatively small fraction of the activity (428 Ci). However, all of the isotopes present in the pits, shafts and absorption beds are of concern when considering the ultimate disposition of the disposal areas. Many of the isotopes are short-lived and may be of concern only in the event of a decision to exhume the waste for transferral to a more permanent repository. Many of the wastes, such as ^{239}Pu, are of concern principally because of their biological toxicity and long half-lives.

APPENDIX

BIOLOGICAL PARAMETERS UTILIZED

IN

EVALUATING THE RADIATION DOSE EQUATIONS

TABLE A-I

FRACTION OF MATERIAL IN THE GI TRACT TRANSFERRED TO BLOOD[a]

Element (Soluble Form)	Fraction from GI to Blood, f_1
U	10^{-4}
Pu	3×10^{-5}
Am	10^{-4}
Pu (insoluble)[27]	10^{-6}

[a](Ref. 35)

TABLE A-II

FRACTION OF MATERIAL ENTERING THE BLOOD TRANSFERRED TO THE ORGAN OF INTEREST[a]

Element	Organ	Fraction Reaching Organ, f_2'
U	Bone	0.33
U	Kidneys	0.11
Pu	Bone	0.45[b]
Pu	Liver	0.45[b]
Pu	Kidneys	0.02
Am	Bone	0.25
Am	Liver	0.35
Am	Kidneys	0.03

[a](Ref. 35)

[b](Ref. 27)

TABLE A-III

FRACTION OF MATERIAL INGESTED TRANSFERRED TO THE ORGAN OF INTEREST[a]

Element (Soluble Form)	Organ	Fraction Reaching Organ of Interest, f_w'
U	Bone	3.3×10^{-5}
U	Kidneys	1.1×10^{-5}
Pu	Bone[b]	1.4×10^{-5}
Pu	Liver[b]	1.4×10^{-5}
Pu	Kidneys	6×10^{-7}
Am	Bone	2.5×10^{-5}
Am	Liver	3.5×10^{-5}
Am	Kidneys	3×10^{-6}

[a](Ref. 35)

[b](Ref. 23)

TABLE A-IV

EFFECTIVE ABSORBED ENERGY PER DISINTEGRATION FOR ORGANS OF INTEREST[a]

Nuclide	ε (MeV/disintegration)				
	Liver	Lung[b]	Kidneys	Bone	LLI
^{233}U	-	50	50	250	0.49
^{235}U	-	46	46	230	-
^{238}U	-	43	43	220	0.43
^{238}Pu	57	57	57	280	0.55
^{239}Pu	53	53	53	270	0.52
^{240}Pu	53	53	53	270	0.52
^{241}Pu	1.0	0.053	2.5	14	-
^{242}Pu	51	51	51	250	0.49
^{241}Am	57	57	57	280	0.56

[a](Ref. 35)

[b]Insoluble compounds

TABLE A-V

ORGAN MASSES UTILIZED IN EVALUATING THE RADIATION DOSE EQUATIONS[a]

Organ	Mass (grams)
Lungs	1 000
GI (LLI) Tract[b]	135
Bone	7 000
Kidneys	300
Liver	1 700

[a] (Ref. 35)

[b] (Ref. 39)

TABLE A-VI

EFFECTIVE ELIMINATION RATE CONSTANTS FOR THE VARIOUS ORGANS

Organ	Nuclide	Effective Half-Time (day)	Effective λ (day^{-1})
GI (LLI) tract	U, Pu, Am	1	-
Bone	^{233}U	300	2.3×10^{-3}
	^{235}U	300	2.3×10^{-3}
	^{238}U	300	2.3×10^{-3}
	^{238}Pu	1.7×10^{4}	4.0×10^{-5}
	^{239}Pu	3.6×10^{4}	1.9×10^{-5}
	^{240}Pu	3.6×10^{4}	1.9×10^{-5}
	^{241}Pu	4.2×10^{3}	1.6×10^{-4}
	^{242}Pu	3.6×10^{4}	1.9×10^{-5}
	^{241}Am	3.0×10^{4}	2.3×10^{-5}
Liver	^{238}Pu	1.0×10^{4}	6.8×10^{-5}
	^{239}Pu	1.5×10^{4}	4.8×10^{-5}
	^{240}Pu	1.5×10^{4}	4.8×10^{-5}
	^{241}Pu	3.6×10^{3}	1.9×10^{-4}
	^{242}Pu	1.5×10^{4}	4.7×10^{-5}
	^{241}Am	1.3×10^{3}	5.2×10^{-5}
Kidneys	^{233}U	15	4.6×10^{-2}
	^{235}U	15	4.6×10^{-2}
	^{238}U	15	4.6×10^{-2}
	^{238}Pu	1.6×10^{4}	4.3×10^{-5}
	^{239}Pu	3.2×10^{4}	2.2×10^{-5}
	^{240}Pu	3.2×10^{4}	2.2×10^{-5}
	^{241}Pu	4.2×10^{3}	1.7×10^{-4}
	^{242}Pu	3.2×10^{4}	3.2×10^{-5}
	^{241}Am	2.3×10^{4}	3.0×10^{-5}

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