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TITLE: LIMITS FOR THE BURIAL OF THE DEPARTMENT OF ENERGY TRANSURANIC WASTES

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LIMITS FOR THE BURIAL OF THE DEPARTMENT OF ENERGY TRANSURANIC WASTES

by

J. W. Healy and J. C. Rodgers

EXECUTIVE SUMMARY

Recommended limits are derived for the burial of solid wastes contaminated with transuranium elements as generated in Department of Energy facilities. The limiting exposure to future populations, used as a basis for the calculations, was 0.5 rem/year to the maximum exposed organ of the individual deemed to have the highest exposure to the transuranium elements for each pathway. This dose rate, applied to the dose rate occurring during a 70 year lifetime of the maximum exposed individual, was derived as applicable after review of the recommendations of the National Council on Radiation Protection and Measurements, The International Council on Radiological Protection, The Federal Radiation Council and The Environmental Protection Agency.

Before the actual pathway analyses were done, calculations were made of the dose to an individual from inhalation or ingestion of the transuranium elements and these values were applied to obtain a soil limit based upon an individual living on the area. The ^{241}Am limit is particularly uncertain because of the lack of information on the diet. We assumed that the ^{241}Am uptake from the G-1 Tract was ten times higher than that of plutonium and this resulted in ingestion being the controlling pathway.

Major Pathways considered were:

- 1) Erosion of the wastes into a nearby stream after several thousands of years;
- 2) Leaching of the transuranic waste to an aquifer;
- 3) Intrusion into the waste mass; and
- 4) Exposure of the wastes by erosion, or other mechanism, with people living in the area.

Other pathways reviewed, but not treated in such detail, were movement to the surface by plant uptake, movement to the surface by burrowing animals and severe events, such as flooding, meteor impact or glacial action.

The intrusion pathway was calculated but firm recommendations were not made on this pathway because of differences in the Task Group as to the propriety of considering intrusion as a valid source of exposure. This arises from feelings, on one hand, that warnings are adequate with anyone disregarding them bringing the risk upon himself; and, on the other hand, a distrust of being able to maintain such warnings over centuries. It was the recommendation of the Group that the applicability of this pathway should be reviewed by the decision-makers, perhaps with the advice of individuals skilled in the legal, ethical and sociological aspects of the matter.

The intrusion pathways were calculated to limit the dose rate to artifact hunters. In addition to the exposure during the digging in a search for artifacts, surface contamination limits are derived for solid objects, such as tools or glassware, and lower limits are derived for trash packaged in plastic because of the evidence that current plastics delay the degradation. This leaves the possibility that the intruder could consider the bags of waste as artifacts.

Maximum limits, for individual packages of waste, were derived for two major classes of waste; 1) those in which the contamination level of the individual items varies widely, with much of the waste not contaminated; and 2) those in which the contamination levels are reasonably uniform. Examples of the first class are the routine trash or decommissioning waste and examples of the second class would be incinerator ash or sludges from the treatment of liquid waste. The differences in the two classes results from differences in the degree of self-dilution. For the first class, it was estimated that the average concentration in the waste was 5% of the maximum and an additional dilution of 1/6th occurs from the soil used in layering of wastes and the soil between pits in the burial ground. For the second class, the average is close to the maximum and only the soil dilution is used.

Because the exposure of the wastes and intrusion were the limiting pathways, consideration was given to the use of "deep burial." This was not closely defined because of wide site differences, and it is recommended that, if this option is chosen, the conditions for a particular site be defined by an

analysis of erosion rates and other factors with the goal of making the exposure of wastes improbable.

Limits derived in this report are given in Table I.

It must be recognized that the data available for predicting the doses upon which the proposed limits are based have many uncertainties so that a corresponding uncertainty occurs in the limit. We have examined the key uncertainties in the last section of the report (pp. 110). One key uncertainty is the choice of the dose limit - 500 mrem/y to the organ with the highest dose rate at the end of the life-span of the maximum exposed individual. If it is judged that a higher or lower dose rate is applicable, then the waste limits should be scaled in proportion.

TABLE I
LIMITS DERIVED FOR THE BURIAL OF SOLID WASTES
CONTAMINATED WITH TRANSURANIUM ELEMENTS

Type of Waste	Limit					
	^{238}Pu	$^{239,240}\text{Pu}$	^{241}Am	^{241}Pu	^{242}Cm	^{244}Cm
<u>Shallow Burial</u>						
Class 1 (nCi/g) ^a	3 000	50	5	200	500 000	20 000
Class 2 (pCi/g) ^b	100	2	0.4	10	30 000	900
<u>Intrusion</u>						
Trash-nondegradable packaging (nCi/g) ^d	15	3	3	100	3 000	1 000
Artifacts (pCi/cm ²) ^e	300	60	60	2 000	10 000	20 000
<u>Deep Burial</u>						
Class 1 (nCi/g)	5 000	800	100	4 000	1 000 000	300 000
Class 2 (nCi/g)	300	40	5	200	50 000	10 000

^aContamination distributed with average 5% of maximum.

^bContamination uniform through waste.

^cNot recommended until review indicates applicability of the intrusion pathway.

^dDegradable waste packaged in non-degradable plastic.

^eSurface contamination-averaged over 1 m².

LIMITS FOR THE BURIAL OF THE DEPARTMENT OF ENERGY TRANSURANIC WASTES

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ABSTRACT

Potential limits for the shallow earth burial of transuranic elements were examined by simplified models of the individual pathways to man. Pathways examined included transport to surface streams, transport to ground water, intrusion, and people living on the burial ground area after the wastes have surfaced. Limits are derived for each pathway and operational limits are suggested based upon a dose to the organ receiving the maximum dose rate of 0.5 rem/y after 70 years of exposure for the maximum exposed individual.

I. INTRODUCTION

In the early days of the Manhattan Project, controlled shallow earth burial grounds were established at the individual sites for disposal of items which were contaminated with radioactive materials and were no longer useable. This was done initially for security reasons and was continued in order to isolate the wastes in a controlled area to avoid any potential hazard to the public that access to such landfills would entail. The burial practices have continued with improvements at many of the sites although those projects in more heavily populated areas have shipped their wastes to other places for burial.

In 1970, the Atomic Energy Commission (AEC) required that special consideration be given to wastes contaminated with transuranic elements if the level exceeded 10 nCi per gram of waste.¹ This level was derived by comparison with the highest levels of radium in deposits in the earth.² In September 1974, the Nuclear Regulatory Commission published a rulemaking,³ subsequently

withdrawn, which required commercial wastes greater than this level to be delivered to the Energy Research and Development Agency (ERDA) for placement in a repository. Essentially, these actions establish an upper limit for transuranic elements on wastes that could be buried.

In the early development by the AEC of the present Department of Energy (DOE) burial grounds there was a presumption that the area would be controlled by the Federal Government until the radioactive wastes had decayed to non-hazardous levels. For certain of the transuranic elements this would imply control for many thousands and tens of thousands of years.

On September 29, 1975, Dr. Richard W. Roberts, Assistant Administrator for Nuclear Energy in ERDA, requested Dr. J. L. Liverman, Assistant Administrator for Environment and Safety in ERDA, to "...establish an official ERDA health and safety viewpoint identifying those TRU wastes that should not be disposed of by simple land burial and, therefore, would require long term control such as afforded by a Federal Repository." (Emphasis added.) The letter of request is reproduced as Appendix A.

In response, the Los Alamos Scientific Laboratory was commissioned to perform the study with the assistance of a special Task Group composed of experts in a variety of fields. This Task Group consists of:

S. I. Auerbach, Director, Environmental Sciences
Division, Oak Ridge National Laboratory.

John Corey, Savannah River Laboratory.

J. O. Duguid, Environmental Sciences Division,

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In addition, Bruce Wachholz, Environment and Safety, Department of Energy served as a main contact with the sponsors.

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considered here and the beta-gamma activity requires additional consideration as to shielding and leachability.

This section is a brief discussion of the current wastes and waste handling methods. A more detailed discussion is given in Appendix B.

A. DOE Sites and Waste Quantities

DOE sites where wastes are generated and buried are listed in Table II. Of these, the Rocky Flats Plant in Colorado, the Savannah River Plant in South Carolina, the Mound Laboratory in Ohio, the Hanford Complex in Washington State, and the Los Alamos Scientific Laboratory in New Mexico are the major places where TRU wastes are generated. The others have smaller quantities incidental to their operation. Several of the other DOE facilities not mentioned in the table such as Ames Laboratory; Atomics International in Santa Susana, California; Bendix in Kansas City; Brookhaven National Laboratory, and others will have very small quantities of TRU contaminated wastes that are usually shipped to commercial burial grounds.

TABLE II

DOE SOLID WASTE GENERATING AND BURIAL SITES

<u>Generating Site</u>	<u>Disposal</u>
<u>Western U.S. (Generally Arid)</u>	
Hanford - Washington State	Buried on Site
Idaho National Engineering Laboratory - Idaho	Buried on Site
Rocky Flats - Colorado	Shipped to Idaho
Nevada Test Site, Nevada	Buried on Site
Lawrence Livermore Lab. - Calif.	Shipped to commercial grounds
Los Alamos Scientific Lab. - New Mexico	Buried on Site
<u>Midwestern U.S. (Significant Rainfall)</u>	
Argonne National Lab. - Illinois	Shipped to commercial or Idaho
<u>Eastern U.S. (Significant Rainfall)</u>	
Oak Ridge - Tenn.	Buried on Site
Mound Laboratory - Ohio	Shipped to commercial or Idaho
Savannah River Plant - South Carolina	Buried on Site

The total quantity of wastes containing transuranium elements at all levels generated at the DOE locations is difficult to obtain because records differentiating these wastes from those containing other contaminants have not been kept before 1970. However, the Panel on Land Burial of The National Academy of Sciences - National Research Council, Committee on Radioactive Waste Management⁴ notes that the 61 604 m³ of all wastes, radioactive or suspected of being radioactive, produced at all ERDA sites in FY 1974, was about 40% of the volume of solid waste produced annually in a college town with a population of 50 000 and a student population of 32 000. This comparison is intended to show that the land areas required are relatively modest and not to provide direct comparison with potential hazard.

B. Waste Matrices

The matrix for solid wastes is considered as the item or material contaminated with the transuranic elements. Such items consist of everything that can be used in research and development, or processing, from wipes to glove boxes or from laboratory glassware to metal lathes. Other components of the waste arise from the processes themselves, such as the sludges resulting from the purification of liquid wastes, the oils used for lubrication and seals, or the fluorocarbons for degreasing. The latter items are normally converted to solid waste by absorbing them on a material such as vermiculite. Items such as High Efficiency Particulate Absorbing (HEPA) filters used in cleaning the air of the processing building or the air from the process stream are important sources of waste. When a facility is decommissioned, there are large quantities of obsolete equipment and construction materials that are included.

The importance of each of the possible matrices differs from one location to the next, depending upon the processes used at the site. Variations with time are to be expected as these processes change and decommissioning activities at a particular site occur.

Wastes can originate in any area where the transuranium elements are handled. It is general practice to consider any item that goes into a room where these elements are used to be potentially contaminated unless it can be shown that the probability of contacting any of the transuranium elements is negligible. It is expected that this practice will continue in the future because of the difficulty and expense of measuring each item to show that it is not contaminated. This results in a large quantity of essentially

uncontaminated trash or equipment with levels well below any limit. As a result, the average concentration going to burial from the, room or from decommissioning of a facility, will be considerably lower than any maximum level established as a limit.

C. Burial Practices

At all ERDA sites the wastes are placed in pits or trenches in the ground and are covered by the material excavated from the pits. A summary of some of the burial factors at each site is given in Table III.⁵

A review of the experience with shallow-earth burial sites, both DOE and commercial, is given in Appendix C.

D. Radioactivity Characteristics of the Transuranium Elements

The transuranic elements discussed in this report are those that are of primary interest to DOE in present and known future operations. Emphasis is

TABLE III
SOME BURIAL AND BURIAL SITE CHARACTERISTICS
AT ERDA SITES

	<u>Trench Size</u> (m)	<u>Distance Between Pits</u> (m)	<u>Minimum Cover</u> (m)	<u>Distance to Water Table</u> (m)	<u>Inter- posed Material</u>
Savannah River	6 wide 6 deep variable length	-	1.2	10-20	Sand and clay
Oak Ridge	3 wide 3-4.5 deep 15 long	1.5	1	2-5	Fractured shale
Los Alamos	8-30 wide 8 deep 120-180 long	4.5	1.5	200-400	Fractured tuff
Idaho	2-30 wide 4 deep	-	1	60-300	Sand and clay
Hanford	1.5-5 wide 4-8 deep variable length	-	2.5	100	Sand and clay

placed on the alpha emitting isotopes of plutonium, ^{238}Pu , ^{239}Pu , ^{240}Pu and ^{241}Am , a daughter of ^{241}Pu . ^{242}Cm and ^{244}Cm are considered only as parents of plutonium isotopes while ^{233}U is not included because of the lack of known major plans for separation of this nuclide from irradiated thorium.

A discussion of the decay chains for these nuclides is included in Appendix B. Table IV presents some of the characteristics of the important nuclides considered here.⁶

The alpha emitting plutonium isotopes decay to uranium isotopes that occur in nature. However, the half-lives of the daughters are long so that their concentrations are considerably lower than the concentrations of the original plutonium parents. The primary potential for exposure from daughters occurs in the decay of ^{241}Pu to ^{241}Am resulting in an increase in alpha activity for the first 70 years after purification of the ^{241}Pu .

TABLE IV^a
DECAY CHARACTERISTICS

PARENT				DAUGHTER		Ci Daughter per Ci of parent ^b
Isotope	T _{1/2} (Years)	Emission	Specific Activity (Ci/g)	Isotope	T _{1/2} (Years)	
^{238}Pu	87.75	Alpha	17.1	^{234}U	2.445×10^5	3.6×10^{-4}
^{239}Pu	24 390	Alpha	0.0613	^{235}U	7.1×10^8	3.4×10^{-5}
^{240}Pu	6 537	Alpha	0.0367	^{236}U	2.3415×10^7	2.8×10^{-4}
^{241}Pu	14.4	Beta	103	^{241}Am	433	0.030
^{241}Am	433	Alpha	3.43	^{237}Np	2.14×10^6	2.0×10^{-4}
^{242}Cm	0.45	Alpha	3 280	^{238}Pu	87.75	5.0×10^{-3}
^{244}Cm	18.11	Alpha	80.9	^{240}Pu	6 537	2.7×10^{-3}

^aValues of constants taken from Reference 6.

^bAt time of maximum ratio.

E. The Model Burial Ground

The actual burial grounds at DOE sites vary widely although the burial practices are relatively the same at all sites. Thus, the depth to the water table varies from about 300 meters at Los Alamos to several meters at Oak Ridge; the material in which burial occurs varies widely from consolidated tuff at Los Alamos to sandy soils with admixed clays at Hanford; and the excess of rainfall over evaporation varies from negative at Hanford and Los Alamos to high at Savannah River and Oak Ridge.

As a result we have defined a model burial ground that combines many of the worst features of each site for purposes of this generic study. Thus, the results of many of the pathway analyses are expected to be conservative for many of the sites but more realistic for others.

In the derivation of the criteria to be used, we have defined two types of burial situations that will be considered in the derivation of limits - shallow earth burial and deep earth burial. The difference lies largely in the accessibility to exposure of the wastes by later intrusion or erosion. Present practice, with cover over the waste of about 2 m, is defined as shallow earth burial. Deep burial is defined as burial at a depth such that the wastes are not expected to be exposed by erosion and the probability of inadvertent intrusion is very low. The exact depth will depend, to some degree, on the nature of the area and the material in which the wastes are buried but would seem to be on the order of 10 meters or more for most places. (Note that deeper depths, such as could be obtained by mining or caves, would not be excluded.) The same basic model will be used for both with the only difference being the depth of the top of the waste below the surface.

The burial ground is taken to be a square 1 000 m on a side with an area of about 100 hectares. At a burial density of 3 500 m³ per acre⁷, this would give a total capacity for the burial ground of about 860 000 m³ or about 30 000 000 cubic feet. The wastes at the bottom of the pits are assumed to lie immediately above the aquifer which is 10 m deep with a bulk velocity in the aquifer of 30 cm/day. Porosity of the aquifer will vary. The pits are 4 m deep with spaces left between the pits so that, of the total volume of the burial ground, about 1/5th of the volume including the two meter cap, is taken up by wastes.

For sites where the water table is some distance below the burial pits especially if the intervening material has a high ion exchange coefficient and distributed flow through soil particulates, the conservatism in the above model

is significant. This is due to the delay of the contamination front occasioned by the absorption of transuranics on the sediments. If rainfall is insufficient to penetrate to the water table, even greater conservatism is present. Flow rates in aquifers lower than the 30 cm/day do occur as well as rates considerably in excess of this. It may be noted that if the flow rate is considerably smaller, one possibility may be an indication of low permeability so that the development of wells to provide significant quantities of water may be more difficult.

Overall, as a basis for developing generic limits applicable to present sites, we believe the above simple model to be appropriate.

III. CRITERIA FOR RECOMMENDATIONS

The basic approach in this document is to use pathway analysis to estimate potential radiation doses to individuals in future generations as a result of postulated releases of transuranium elements from the burial ground. From these doses, estimates of limitations on quantities and/or concentrations of transuranium elements in the waste are derived. Criteria are required for both the dose that could be permitted if the postulated scenarios develop and for the consideration to be given to potential exposures in the far distant future from the longer-lived isotopes of certain of the transuranium elements. The criteria to be used are discussed and derived in this section.

A. Long-Term Considerations

The long half-lives of ^{239}Pu and ^{240}Pu result in quantities buried remaining in existence for thousands of years with a gradual decrease over periods of tens of thousands of years. The potential risk that these materials may impose on people in future generations is a central issue in the disposal of radioactive wastes and the same issue will undoubtedly become of greater importance for non-radioactive wastes under the National Resources Recovery and Conservation Act. The National Environmental Policy Act of 1969 (NEPA) set forth a national goal to: "fulfill the responsibilities of each generation as trustees of the environment for succeeding generations". This phrase is sometimes quoted to indicate that the intent of Congress was to make this responsibility a paramount objective. However, the Act also states that it is the policy of both government and private organizations "-- to use all practicable means and measures - - - in a manner calculated to foster and

promote the general welfare, to create and maintain conditions under which man and nature can exist in productive harmony, and fulfill the social, economic and other requirements of present and future generations of Americans." (Emphasis added) It is, then, specified that all practicable means consistent with other considerations of National Policy shall be used to fulfill the responsibilities to future generations.

It is apparent that the Congress, in passing this Act, recognized the conflicts between present needs and desires and the responsibilities to the future, and intended that the welfare of future generations be included as a factor in making decisions but that this was not to be an overriding goal. Thus, we cannot use NEPA as a basis for decision, but can use it for general guidance.

The question of risk to future generations, particularly those in the far distant future, is not resolvable by scientific reasoning and even logic is limited in arriving at a decision because of the very large uncertainties as to the future course of events. Instead it is a social question that must be resolved by consideration of the resources that society wishes to allocate to this goal as opposed to other national goals. Eventually the principles derived for the handling of radioactive wastes may be applied to other non-degradable toxic materials. Thus, consideration of the impact of criteria derived for these wastes may include industries and activities far broader than the nuclear applications.

The Environmental Protection Agency has recently proposed draft criteria for discussion.⁸ A brief digest and discussion of these criteria as they apply to our problem follow. However, it must be remembered that these criteria are preliminary and were issued only for discussion.

Criterion 1 "Radioactive material which has no designated resource or product value should be considered radioactive waste ---".

This criterion obviously applies to the transuranium element contaminated wastes that are the subject of this report, although we believe that additional effort is needed in process development to minimize both the quantity of transuranium elements in the waste and the total volume of wastes.

Criterion 2 "Environmental protection determinations for radioactive wastes should be based primarily on an assessment of risk to individuals and populations." The following factors should be examined in the assessment: "a) the total amount of waste in a location and its persistence ---; b) the

potential adverse health effects on human individuals and populations for a reasonable range of future population sizes and distributions and uses of the land ---; c) the projected effectiveness of alternative methods of institutional, engineered and natural barrier controls ---; d) the probabilities of releases of radioactive materials --- through failures of natural or engineered barriers, loss of institutional controls or intrusion ---; and e) the uncertainties in the risk assessments ---."

The use of risk estimates in arriving at limitations for radiation exposure is a policy of the EPA. Such estimates are currently derived using the linear, no-threshold hypothesis from effects noted in humans, usually at relatively high doses and dose rates so that considerable uncertainty exists in the result. In addition to this uncertainty a further difficulty arises in defining a risk that will be considered as acceptable. Efforts to do this have frequently resulted in very low risks as compared to those encountered from other currently "acceptable" (or at least, accepted) practices because of the emotional nature of the responses and the political need to gain acceptance of a proposal. While we will not use risk as a primary tool in the derivation of exposure limits, we will use currently accepted methods for estimating the risk to the individuals involved in the scenario providing the highest dose.

Criterion 3 "Risk due to radioactive wastes should be deemed unacceptable unless it is justified that more complete isolation is unreasonable in view of economic, social and technical considerations; any potential risks to future generations should be no greater than that accepted by the current generation".

The first part of this criterion essentially calls for a cost-benefit analysis to reduce the future exposure as far as is reasonably practicable. For our study of an acceptable level on a generic basis, this criterion poses problems because such a study requires definition of a particular type of waste and site. However, we will provide a limit that is to be regarded as an upper limit with the cost-benefit analysis for further reduction to be provided with respect to the individual site.

The second part of this criterion, dealing with the risk to future generations, is discussed in the body of the EPA report only by the statement: "Because of the long term implications of many of the waste materials and the ethical responsibility to minimize intergenerational risk transference, it is important that as a limiting case the risk imposed on future generations be no greater than those the producing generation is willing to accept, as expressed

in its own public health protection standards and policies." This criterion seems to be an interpretation of NEPA which does not appear to take into account the requirement to "-- fulfill the social, economic and other requirements of present and future generations of Americans." (Emphasis added) Thus, some provision should be made to consider the other policies of the nation and the economic well-being of the present generation. We would believe that, under some conditions, it would not be advisable to conduct business so as to leave a minimum-risk world to future generations regardless of costs, social disruptions or risks to present generations.

In this context, there appears to be a feeling that future generations will be similar in their characteristics to us. Some speculation as to the future of mankind and the resulting impact of inherited risks may be in order. We can postulate a number of possible scenarios that will have different apparent probabilities of occurrence to different people but which are all possible. Without going into too much detail, we shall consider three scenarios and address qualitatively the impact from the use of present risk standards. For the first scenario, technology and social behavior become more highly developed so that risks from disease, accidents and other of the ills that man is heir to are decreased. This will have two possible effects. 1) Man will be more risk averse because the risks that he is used to are lower.⁹ This means that he will tolerate only risks at a lower level than those we now consider acceptable. 2) However, he will be better able to control those possible residual risks from the disposal of waste and, in fact, may find these residues of benefit to him in ways not now seen. On balance, we believe that the increase in ability to cope with risks of nature, or those presented by wastes containing transuranium elements, will overbalance the increase in risk aversion. In other words, man in this scenario, will be well beyond us in his ability to handle situations that the present generation may leave. In the second scenario, exactly the opposite occurs. That is, due to an increasing population, the depletion of resources, or war, man reverts to a more primitive stage where the diseases that are now under control increase to the point where, on a competitive basis, the risks from the radioactive waste become even smaller in comparison. In this case, again, one could justify a higher risk because the overall impact of the radioactive risk on the total risk picture is smaller.⁹ In the third scenario, conditions remain the same as they are at present for all future generations. In view of the rapid increase in technology and the social sciences in the past

50 to 100 years it appears unlikely that this scenario will develop, but it appears to be the only one in which one could justify using present resources to fully protect future generations.

Criterion 4 "Controls should be applied with a goal of isolating radioactive wastes from the biosphere ---. When institutional control is the method chosen to provide environmental protection of radioactive wastes, no restrictions on customary uses of associated land areas and surface and ground waters --- should be required after 100 years; radioactive wastes that would require protection beyond 100 years should not be isolated by institutional means, but rather by as many physical and natural barriers as is practicable to minimize environmental impact if one or more fails or is accidentally or intentionally breached."

It is doubted that shallow land burial without considerable modification in installing barriers or going to greater depth could ever meet the goal of totally isolating the long-lived transuranium elements. Of course, if this goal is met, then all of the other criteria become redundant. We will proceed upon the assumption that this goal really means that burial shall only occur in areas which have optimal characteristics in preventing migration. It is also noted that the 100 year time period is not necessarily an optimum period and that reasons could be given for a longer time to permit the decay of intermediate lived nuclides or for a shorter time to minimize the period of control. In general the intent of this criterion that control over many centuries should not be required for future burial grounds is a worthy one.

Criterion 5 "Locations for radioactive waste disposal should be chosen whenever practicable such that the action over time of natural forces --- could be will projected to enhance, rather than reduce, environmental isolation."

This criterion speaks to the criteria for the burial site, which may be influenced in DOE by the location of present facilities and the costs and potential risks of shipping the waste to a different site.

Criterion 6 "Certain additional procedures and techniques should also be applied to waste disposal systems which otherwise satisfy these criteria if they provide a net improvement in environmental and health protection; among these are: a) monitoring prior to completion of disposal to determine for timely any unanticipated effects which could result in releases of radioactivity ---; b) procedures or techniques designed to enhance the retrievability of the waste; and c) passive methods of communicating to future people the potential hazards

that could result from an accidental or intentional disturbance of radioactive wastes."

Experience in recovering lost articles from burial sites indicate that retrievability can be accomplished, although at some expense and risk, along with a larger quantity of waste than was originally placed in the burial ground because of the accompanying soil. However, provisions for increased retrievability are frequently expensive and should not be necessary if proper burial grounds are used and limits are applied to the quantities placed in the burial ground to limit exposure, even if leakage does occur. Methods of communicating to future people will require considerable study because of man's proven tendency to steal or badly deface any marker or monument. It should also be noted that, in the case of burial grounds, such marking will serve to call attention to the fact that something is buried there and may invite intrusion. On balance, however we believe that such marking would be useful if a method that is practical in costs, tamper-proof and potentially effective can be devised.

B. Dose Limitations

Recommendations for the limitation of radiation doses have been made for close to 50 years by several organizations: the International Commission on Radiological Protection (ICRP) and the National Council on Radiation Protection and Measurements (NCRP) or its predecessors in name. In these organizations recommendations have been developed by individuals knowledgeable in the field and who, as radiation workers themselves, were willing to live with both the risks and handicaps resulting from their recommendations. The limits and practices that they derived were voluntary in application but were widely used in the derivation of regulations for the control of radiation by many governments, including the U.S. In 1959 the Federal Radiation Council was formed to provide guidance to all federal agencies involved in the use of radiation. The Federal Radiation Council was composed of the Secretaries of key government agencies as well as the leaders of other agencies not of cabinet position, such as the Atomic Energy Commission (AEC). Their work was carried out by inter-agency groups with frequent consultation with individuals in the private sector, and the results were used as advice to the President as to standards for radiation protection to be used by Federal Agencies. In 1970 the functions of the Federal Radiation Council were transferred to the newly

established Environmental Protection Agency (EPA). The EPA carries out studies with their own staff to arrive at a preliminary finding which is then issued in draft form for comment and before final issue.

In this section we will review the pertinent recommendations of each of these groups to arrive at a limit that will be used for the pathway calculations.

In their most recent report on dose limitations, the ICRP¹⁰ has provided a system whereby the dose to any individual organ or tissue is limited to such an extent that the sum of all of the risks to the individual from irradiation of the organs or tissues does not exceed the risk caused by the dose limit to the whole body from external radiation. For the external dose to the public they prescribe a value of 0.5 rems per year to the critical group in the population (i.e. the group that is at greatest risk from the presence of radiation or radioactive materials in their surroundings). They use a value of risk to the general public of 10^{-6} to 10^{-5} per year as an acceptable value and note that such an average risk could be attained even if a few individuals exposed to the dose-equivalent limit could run a risk in the range of 10^{-5} to 10^{-4} per year. They also indicate that dose partitioning, because of multiple sources of exposure, is not really needed because the use of critical groups keeps the dose to the population as a whole low, and, in general, when the practices causing the exposure are few, they cause little exposure outside of the critical group. They do caution however, that the authorities should keep a watch on this situation to assure that there is not overlap in the critical group and that no individual receives a serious overdose as a result of membership in more than one critical group. Thus, they believe that the full value of the 0.5 rems per year could be used in the derivation of the limit for any one procedure. They then state, "Even the few individuals who would actually receive doses of this magnitude over a long period of time would not be subject to risks appreciably above those generally considered acceptable".¹⁰

Limits for the individual organs or tissues are derived to give the same risk as whole body radiation by weighting factors. These factors are given in Table V for the organs or tissues of interest from exposure to transuranium elements.

It should be noted that the factor for bone applies to the bone surface while the liver factor is essentially a default number applied to all organs except those listed because of the lack of information to derive the proper

TABLE V
WEIGHTING FACTORS FOR ORGAN OR TISSUE
DOSES - ICRP-26^a

Organ	Weighting Factor
Lung	0.12
Bone Surface	0.03
Liver	0.06

^aReference 10.

factor. In practice these weighting factors are multiplied by the dose to the appropriate organ or tissue and summed. If the sum is less than 0.5 rem per year, the doses are below the limit. This provides an apparent increase in organ limit since the lung is now allowed 4.2 rem per year and the bone surfaces are allowed 16.7 rem per year if no other organ or tissue is irradiated. However, such organs or tissues are rarely irradiated alone so that consideration of the dose to all organs will begin to bring the values closer to those used formerly by the ICRP and used currently by other bodies. One other change from past practice in the ICRP report was the use of a quality factor of 20 for alpha particles rather than a value of 10.

The NCRP has not systematically evaluated the internal emitter position since the listing of MPC's was issued in 1959.¹¹ However, they did review the basic criteria for radiation protection in 1971.¹² While they did not provide specific limits for organs as did the ICRP, they did provide a limit, for the critical organs (whole-body) of an individual in the population, of 0.5 rem in any one year in addition to natural radiation and medical and dental exposure. That they intended this value to be used for individual organs wherever possible is indicated by a portion of their discussion. "To have no organ or tissue exceed 0.5 rem per year is a reasonable target, but it is arbitrary, of course, and may not always be achievable. At that limit, the incremental radiation received by any individual is at most some four to five times the natural radiation, and is less than twice the existing background in some regions. With an intended limit of 0.5 rem per year for most, if not all, situations, the average incremental dose to population groups from radiation plants and

installations would probably easily stay below 0.1 rem per year - - -. It should be emphasized here that the limit of 0.5 rem per year applies to the integration of contributions from all sources, excluding natural radiation and medical radiation. Obviously, the contribution from any one radiation plant or installation should be a small fraction of this to avoid the need for specific dose allocation."¹²

The Federal Radiation Council¹³ coined several definitions that are currently in wide use, particularly in the U.S. The Radiation Protection Guide was defined as, "the radiation dose which should not be exceeded without careful consideration of the reasons for doing so; every effort should be made to encourage the maintenance of radiation doses as far below this guide as practicable." This definition emphasized the risk-benefit nature of an acceptable dose and was coined to replace the term "Maximum Permissible Dose." The Radioactivity Concentration Guide was defined as "the concentration of radioactivity in the environment which is determined to result in whole body or organ doses equal to the Radiation Protection Guide." For external whole body radiation they derived a value of 0.5 rem per year for individuals in the population above natural background and medical uses of radiation. From this, they further derived a limit for the whole-body irradiation of population groups¹³ as follows: "Under certain conditions, such as widespread contamination of the environment, the only data available may be related to average contamination or exposure levels. Under these circumstances, it is necessary to make assumptions concerning the relationship between average and maximum doses. The Federal Radiation Council suggests the use of the arbitrary assumption that the majority of individuals do not vary from the average by a factor greater than three. Thus, we recommend the use of 0.17 rem for yearly whole-body exposure of average population groups. - - - It is critical that this guide be applied with reason and judgement. Especially, it is noted that the use of the average figure, as a substitute for evidence concerning the dose to individuals, is permissible only when there is a probability of appreciable homogeneity concerning the distribution of the dose within a population included in the average." While the value of 0.17 rem/year for population groups is the same as the one then in existence from the NCRP and ICRP, it is of interest that the FRC derivation is one of monitoring rather than estimate of allowable dose.

The FRC provided guides for ¹³¹I, ^{89,90}Sr and ²²⁶Ra in terms of intake.¹⁴ These were in the form of three ranges with prescribed monitoring and actions

for each of the ranges: 1) well below the guide; 2) about at the guide; and 3) above the guide. It is also of interest that in the derivation of the guides for radium and strontium they used values based upon a conclusion that activities could be carried out at this level rather than upon an acceptable risk.

The EPA has provided several standards aimed at particular operations or situations. Standards for environmental radiation protection for nuclear power operations cover the entire fuel cycle exclusive of the mining of uranium or thorium and waste disposal.¹⁵ As a basis for the standard they chose to use the total population dose commitment because "The proposed expanded development of the nuclear power industry requires - - the use of a broader environmental perspective that more specifically considers the potential radiological impact on human populations of radioactive effluents from this industry, rather than just that on the most exposed individual". In addition, they examined the feasibility and cost of restricting effluents and concluded that: "Thus, the standards generally represent the lowest radiation levels at which the Agency has determined that the costs of control are justified by the reduction in health risk". However, in expressing the standards they changed to a basis of individual dose to members of the public or emission limits because of the uncertainties in the models used and the difficulty in administering controls based upon such models. The standards limit the annual dose equivalent to the whole body to 25 mrem, to the thyroid to 75 mrem and to any other organ to 25 mrem. In addition, the quantities of ⁸⁵Kr, ¹²⁹I and certain long-lived transuranic elements released to the environment per gigawatt-year of power produced by the entire fuel cycle are limited to 50 000 curies, 5 millicuries and 0.5 millicuries respectively. Exceptions are made for portions of the fuel cycle, however, in that radon and its daughter products and waste management activities are not included in this standard. The basis for this action was a balancing of the estimated risk against the benefit of the electricity produced as well as an appraisal of what they thought the industry could do to limit radioactivity releases.

The EPA regulations for drinking water¹⁶ cover both man-made beta and photon emitters and alpha emitters. The beta and photon emitters are limited to a quantity that would deliver a dose equivalent of 4 mrem per year, or less, to the total body or to any organ. This was based upon the fact that present water supplies have less radioactive materials than would produce this dose and the

value was set to limit future controllable discharges. A limit of 5 pCi/L was given for the combined ^{226}Ra and ^{228}Ra with a gross alpha particle limit (excluding radon) of 15 pCi/L.

In a recent proposed standard for transuranics in the environment,¹⁷ the EPA provided a basic limitation of 1 mrad/year to the lung and 3 mrad per year to the bone to be applied to the critical group in the exposed population. These values were based upon an estimated cancer risk of about 7×10^{-5} per lifetime or about 10^{-6} per year to an individual continually exposed over his full lifetime. The primary source of concern here was for transuranium elements in the soils because the emissions from processing plants and reactors are well below these values. The value is expressed in rads because the nuclides of concern are all alpha emitters and it was their belief that using this unit as a basis would allow comparison with other alpha emitters and would not require a change in the standard if the value for the Quality Factor for alpha radiation were to change at a future date. However, using the current NCRP value of 10 for the quality factor,¹² these doses correspond to 10 mrem per year to the lung and 150 mrem per year to the bone (including dose distribution factor).

A general principle in radiation protection since the late 40's and early 50's has been that of keeping exposures to the minimum practicable (or "as low as reasonably achievable" in the terminology of the NRC). This principle derived from the assumption that the effects of radiation follow a no-threshold, linear relationship. All bodies and agencies having to do with radiation protection have included such a statement as an integral part of their recommendations. As a corollary to this, the statement that any exposure should be allowed only if there is a corresponding benefit has become a central tenet. These principles are as much a part of the limits as the numbers given above and must be included in any consideration of an applicable dose.

C. Costs

While we have not attempted to provide a detailed cost analysis for each site in this generic analysis, we cannot completely ignore the potential for making recommendations that are completely unworkable because of the additional costs required. The only current alternative to burial appears to be that of placement of the wastes in the proposed repository. In order to ship the wastes and place them in the repository it currently appears that the following additional steps in waste handling will be required.

- 1) Oxidation of all combustibles to remove sources of fire and gas in the repository;
- 2) Compacting or otherwise reducing the volume of the metal or glass objects;
- 3) Packaging of the wastes to meet DOT requirements;
- 4) Shipment to the repository; and
- 5) Unloading and placing the wastes in the repository.

In addition to the costs of these steps, the capital costs of the repository and of any necessary treatment and shipping equipment must be included. It is possible that the wastes could be shipped to the repository without treatment and a central treatment plant at the repository could be used. This would increase shipping costs and would require the operators of the repository to treat wastes with which they have less familiarity than the operators at the generating location. An additional possibility could be that the combustibles are not oxidized but are placed directly in the repository (perhaps after compaction). This would require the shipment of a greater bulk of material and impose some (but possibly not very great) uncertainty in the performance of the repository.

The Deutch Report¹⁸ (pp. 41) notes that the cost of volume reduction and disposal in suitable deeper geologic formation will cost about \$60 per cubic foot as compared to about \$3 per cubic foot for the present operation of the burial grounds. Thus, if it is assumed that 275 000 cubic feet can be buried per acre, the alternative is equivalent to a cost of up to \$10 million per acre. Table K-7 of the Deutch Report¹⁸ indicates that about 1.25 million cubic feet of low-level waste are generated in the DOE per year plus, from Table K-12, about 250 000 cubic feet of transuranic waste above 10 nCi/g. The cost for the transuranic waste placement in the repository would be about \$15 million per year. Since the low level waste contains some waste contaminated with transuranium elements below 10 nCi/g, a maximum cost to DOE for disposal of all wastes would be about \$90 million per year. It should be noted that these cost figures do not include certain commercial wastes that may, in the future, be transferred to DOE because they contain long-lived radionuclides. Thus, there appears to be no real cost restriction involved in a limit as long as it is not too low.

D. Criteria to be Used

The review of present recommendations for dose limitation and current regulations indicates a wide variation in the radiation doses and corresponding risks permitted for different situations. The proposed EPA guidance for transuranics in the environment¹⁷ of 1 mrad/yr to the lung and 3 mrad/yr to the bone would appear, on the surface, to be relevant to our study. This guidance, however, has not, as yet, been promulgated and there is some question as to whether it applies to waste management. Thus, the EPA considers only a depth in the soil of 1 cm in deriving a soil screening level and, in the implementation of the guidance, they permit restrictions to the use of the area without consideration of the possible exposures at future times. The EPA also permits shallow burial of the contamination removed during cleanup at especially designated locations on site. From this, it is apparent that this specific guidance is inapplicable.

The pathways and scenarios postulated in this study are hypothetical in the sense that one cannot say with any certainty that they will occur and, in general, conservative assumptions are used to estimate the dose if they do occur. While a realistic analysis is preferable, the general level of uncertainty as to future conditions as well as the uncertainty in many of the parameters used in the pathway calculations make this conservatism prudent to assure that doses considerably higher than those adopted in these criteria do not occur.

Under these conditions, a limiting whole body dose of 500 mrem/yr as applied to critical groups as recommended by the ICRP³ appears reasonable. However, the ICRP system differs from the NCRP implicit recommendation⁵ that any organ be limited to 500 mrem/yr. For example, calculations of the inhalation of 1 μ m AMAD particles of ²³⁹Pu indicate that the ICRP weighting factors for organ irradiation would result in a permissible daily inhalation some ten times that required to limit the bone dose to 500 mrem/yr after 70 years. In view of the added complexity of applying the weighting factors to uncertain pathways, the uncertainty of the weighting factors, themselves, and the position of the NCRP as the national group for recommending limitations for the conditions in the U.S., we have chosen to utilize the limit of 500 mrem/yr to the maximum organ after 70 years of exposure. It may be noted that this is essentially a dose commitment calculation because the annual increments of uptake sum to a limiting dose rate at the end of life. However, instead of the use of a fixed time over

which each increment acts, such as 50 or 70 years, the limit expressed in the manner of this report sums each increment over the years of life left to the individual.

In application of this dose rate criterion, we will consider it to apply to the individual having the estimated maximum exposure (maximum exposed individual). This is done because it is possible to define a reasonable series of actions for this individual that will result in exposure. Alternative actions would be to apply the number to a "critical group" as recommended by the FRC¹³ and the ICRP¹⁰ or to formulate an additional criterion based upon population dose. These actions were deemed to be impractical for lack of a clairvoyant to predict the makeup of the critical group or the size and distribution of the population and the impossibility of predicting the actions of people that could result in exposure at times far in the future. It is apparent that control of the dose to the maximum exposed individual will result in the average population dose being lower than that to the maximum exposed person by some factor depending upon the pathway of exposure and its degree of uniqueness to the maximum exposed individual.

One other criterion needed is the length of time over which control of the burial ground is required. A reasonable length of time after which the surveillance and control of access can be eliminated will minimize the burden of future expenditures by the controlling organization and will assure reasonable safety regardless of disruptions in records of governments. We have arbitrarily chosen a 200 year period of control for these government facilities, although surveillance can be lowered at some earlier time. The results of most of the scenarios are not greatly dependent upon the period chosen, the primary difference being some additional decay of ^{238}Pu and ^{241}Am and additional time for the waste to settle. We consider this period to start at the time that the operations in the burial ground stop.

In summary, our criteria for these calculations will be a limitation of the dose to the highest organ of a maximum exposed individual for each of the scenarios and pathways considered. However, in deference to the ICRP weighting factors¹⁰ and the EPA guidance¹⁷ for transuranics, we will include calculations of the lung dose so that conversion to an alternative basis is possible. A control period of 200 years is assumed following stoppage of use of the burial ground.

V. PATHWAY ANALYSES

A. Intake Limitations

In order to simplify the presentation of the pathway analyses, quantities of the individual transuranics that can be taken in by inhalation or ingestion without exceeding the maximum dose equivalent rate of 500 mrem/year are calculated in this section. They are, then, applied directly in the pathway analysis. It may be noted that the tacit assumption is made that the quantity calculated is actually taken in each year, with no allowance for time spent elsewhere or accounting for the possibility that the individual will spend only a portion of his lifetime in the area. Annual average intakes are used as the limiting case because the primary impact is the long-term buildup in the organs and this will be affected only slightly by day-to-day variations in the intake.

1. Inhalation

Inhalation is normally considered to be the most important source of intake (possibly aside from wounds in occupational exposure) because of the relatively low absorption of most of the actinides from the GI Tract. Inhalation will result in a radiation dose to the lung from the actinides deposited in the pulmonary region as well as to other organs, chiefly the bone and liver, from the actinides translocated from the lung to the blood with deposition in other organs.

The ICRP^{19,20} has produced a lung model that describes the deposition in the various regions of the lung as a function of the aerodynamic diameter of the particles. It also describes the retention and translocation to other organs as a function of the chemical nature of the inhaled material. In this model three classes of solubility have been defined: Class Y, in which the lung clearance time is of the order of years; Class W, in which the clearance time is of the order of weeks; and Class D, in which the clearance time is of the order of days. For plutonium, the usual compounds expected are described by Class Y while for ²⁴¹Am the greater solubility of the +3 valence actinides leads to the belief that it may better be represented by Class W. However, it is stressed that the assignment of classes must be considered as uncertain, particularly where the actinides are bound to soil particles - the most likely situation in this study. The transfer coefficients and deposition values used in this report are given in Table VI along with the other values required for the calculation of dose. Values were calculated for three particle sizes, as well as for both

TABLE VI
PARAMETERS USED FOR CALCULATION
OF INHALATION DOSE

1. Organ Values

	<u>Weight</u>	<u>Half-Life</u>
Lung	500g	Class Y 500d Class W 50d
Bone	5 000g	100y
Liver	1 800g	40y

2. Deposition

Particle Size	Tracheobronchial	Nasopharyngeal	Pulmonary
0.1 μ m	0.08	0.01	0.5
1 μ m	0.08	0.3	0.25
10 μ m	0.08	0.9	0.09

3. Organ Transfers

	Class Y	Class W
BP to Blood	0.01	0.1
TB to Blood	0.01	0.5
Lung Retention (Lung $T_{1/2}$)	0.6	0.6
Lung to Blood	0.05	0.15
Lung to Lymph	0.15	0.05
Lymph to Blood	0.9	1

4. Radiological Values

Quality Factor	10
W in Bone	5

5. Alpha Energies^a

²³⁸ Pu	5.5 Mev
^{239,240} Pu	5.14 Mev
²⁴¹ Am	5.48 Mev

^aFrom Reference 6.

Class Y and Class W materials, to indicate the influence of these parameters on the dose.

The dose rates resulting from the inhalation of 1 pCi/day are given in Table VII. It may be noted that both ^{241}Am and ^{238}Pu result in a pattern of dose such that the dose rate in the lung reaches a maximum value in a time of 8-15 years. This results from the radioactive decay of these two nuclides so that, for an initially contaminated area, the quantity of nuclide in the area decreases by a significant amount in 70 years (to 52% of the original quantity for ^{238}Pu and to 89% of the original quantity for ^{241}Am). The dose rates to bone and liver through a 20 year period are shown in Figure 1 for the case of 0.1 μm particles with Class Y behavior in order to illustrate this pattern. It may be noted that the bone dose rate for ^{238}Pu also shows a decrease in the later years over that of ^{239}Pu .

TABLE VII
DOSE RATES FROM INHALATION OF 1 pCi/DAY AT TIME OF
MAXIMUM DOSE OR 70 YEARS (CORRECTED FOR DECAY OF THE NUCLIDE)

Nuclide	Rads/year								
	0.1 μm			1 μm			10 μm		
	Lung	Bone	Liver	Lung	Bone	Liver	Lung	Bone	Liver
Class Y									
^{238}Pu	0.37 ^a	0.47	0.20	0.19 ^a	0.25	0.11	0.07 ^a	0.14	0.06
$^{239,240}\text{Pu}$	0.38	0.80	0.32	0.19	0.41	0.17	0.07	0.22	0.09
^{241}Am	0.39 ^b	0.74	0.30	0.20 ^b	0.40	0.16	0.07 ^b	0.22	0.09
Class W									
^{238}Pu	0.04	0.75	0.30	0.02	0.64	0.26	0.01	0.79	0.32
$^{239,240}\text{Pu}$	0.04	1.2	0.50	0.02	1.0	0.42	0.01	1.3	0.52
^{241}Am	0.04	1.3	0.50	0.02	1.1	0.42	0.01	1.3	0.52

^a At 9 years after inhalation starts.

^b At 12 years after inhalation starts.

Fig. 1: Annual doses to bone and liver from continual inhalation of 1 pCi/day of 0.1 μm particles of Class Y nuclides.

The total doses in a 70 year period are given in Table VIII. The total dose from ^{238}Pu is lowest while ^{239}Pu and ^{241}Am are about the same. ^{241}Pu is not included in these dose tables. While ^{241}Pu will decay to ^{241}Am resulting in dose to the body if inhaled, ^{241}Pu has a half-life of only 14.4 years so that it will essentially disappear before control is lost on the burial ground. For this reason, ^{241}Pu is not considered separately but is limited by the quantity of ^{241}Am that will grow in at later times.

The parameters used for these calculations are those for adults even though some exposure can occur in childhood. There are several balancing features here

TABLE VIII

TOTAL DOSES TO 70 YEARS FROM INHALATION OF 1 pCi/DAY
(CORRECTED FOR DECAY OF THE NUCLIDE)

Nuclide	Reme								
	0.1 μ m			1 μ m			10 μ m		
	Lung	Bone	Liver	Lung	Bone	Liver	Lung	Bone	Liver
<u>Class Y</u>									
^{238}Pu	21	20	9.2	10	11	5.0	3.8	6.0	2.8
$^{239,240}\text{Pu}$	26	27	12	13	15	6.7	4.6	8.1	3.7
^{241}Am	26	27	12	13	15	6.6	4.7	8.2	3.7
<u>Class W</u>									
^{238}Pu	2.2	34	16	1.1	29	13	0.4	37	17
$^{239,240}\text{Pu}$	2.6	46	21	1.3	39	18	0.5	49	22
^{241}Am	2.7	45	20	1.3	39	18	0.5	48	22

which have not been well explored. The inhalation rate of children is lower than that of adults but, at the same time, the weights of the organs are lower so that a smaller quantity will cause a higher dose. In addition, children are known to be more susceptible to some forms of radiation to given organs. In view of the many other uncertainties in this study, we will consider the doses calculated for adults to be appropriate.

For later application in the pathway studies, these dose rates can be converted to allowable concentrations using standard values of intake of air and a limiting dose rate of 0.5 rema/year. In this study we used a total intake of air of $20 \text{ m}^3/\text{day}^{21}$. The plutonium compounds were considered to be Class Y while ^{241}Am , a trivalent actinide, was considered to be Class W. In all cases, the bone is the limiting organ because it builds to the highest dose rate in the assumed 70 years lifetime of man. On these assumptions, the air concentrations maintained over the lifetime of man to result in a dose rates to bone of 500 mrema per year at any time in his life are given in Table IX. A similar concentration limit is included for lung because present information indicates that lung may have a higher risk per unit of dose²² and the EPA preliminary

TABLE IX

AIR CONCENTRATIONS TO RESULT IN 500 MREM/Y ORGAN DOSE TO
INDIVIDUAL EXPOSED TO THIS AIR FOR 70 YEARS
(PLUTONIUM CLASS Y - AMERICIUM CLASS W)

	nCi/m ³					
	Particle Size					
	0.1 μ m		1 μ m		10 μ m	
	Lung	Bone	Lung	Bone	Lung	Bone
²³⁸ Pu	0.07	0.05	0.13	0.1	0.4	0.2
²³⁹ Pu	0.07	0.03	0.13	0.06	0.4	0.1
²⁴¹ Am	0.6	0.02	1.3	0.02	2.5	0.02

guidance for transuranics in soil includes both bone and lung dose rate limitations.¹⁷

The discontinuity in values between the plutonium isotopes and ²⁴¹Am is due to the use of different ICRP Task Group solubility classes. The use of Class W for americium emphasizes strongly the bone dose at the expense of the lung dose. This results from the high absorption from the tracheobronchial, nasopharyngeal and pulmonary region for the Class W material. If situations are found where the ²⁴¹Am behavior is closer to Class Y, the lung dose from the concentrations given in this table will be underestimated by about a factor of 10 while the bone dose will be overestimated. However, since the bone is the critical organ in all classes, the use of Class W for ²⁴¹Am will be conservative in our estimates.

2. Ingestion

While ingestion is not normally considered to be a controlling route for transuranics because of the relatively low transfer from soils to plants to meat to man or from soils to plants to man, the large quantities of foodstuffs ingested can, if contaminated, add to the dose to the bone and liver received from inhalation of transuranium elements.

The question of the absorption of the transuranics from the GI Tract to the blood and then to other organs has been raised lately because of the high uptake values used by the EPA in their proposed guidance on transuranics in the

environment¹⁶ and the contention that plutonium will be oxidized to the +6 state in the chlorination of water with resultant high uptake from the GI Tract.²³ As a result, a review of the data available on uptake from the GI Tract was done and is given in Appendix D. In summary, it was found that recent studies by Sullivan²⁴ showed that the previous data were faulty and the newer data indicated little, if any, increased uptake for plutonium in the +6 state. The review in Appendix D indicated that the previous ICRP value of 3×10^{-5} for the GI absorption was reasonably good in light of present data, although values were derived of 5×10^{-5} for the transfer to bone and 1×10^{-5} to liver for plutonium and 1×10^{-4} to bone and 5×10^{-5} to liver for ²⁴¹Am. Following the preparation of Appendix D it was found that the ICRP²⁵ had decided to use values for the GI absorption during occupational exposure of 10^{-4} for "soluble" plutonium and 10^{-5} for "insoluble" plutonium. For americium, the value used by the ICRP was 5×10^{-4} . The distribution of 45% to bone and 45% to liver was retained. Since these values are in reasonable agreement with those derived in Appendix D, the ICRP numbers for insoluble plutonium and americium were accepted for use in this study.

Another question that arises is whether the GI uptake at very low plutonium intakes is greater than those at higher intakes used in many of the experiments to determine uptake. In early studies on uptake of plutonium, Katz et al.²⁶ and Weeks et al.²⁷ administered chronic doses of ²³⁹Pu and ²³⁸Pu to rats over a nine month period. The total quantity of plutonium ranged from 0.0016 ug to 140 ug with no indication of an increase in uptake with lower quantities administered. Thus, it appears reasonable to believe that the effect of quantity taken into the body is not of great importance if it exists at all.

The possibility of enhanced uptake from transuranium nuclides incorporated in foodstuffs because of their changed chemical state and possible binding with protein or other components of the food is now under investigation. It is a difficult question to study because of the low uptake of plutonium in foodstuffs and the low uptake from the GI Tract resulting in difficulties in obtaining foodstuffs with sufficient activity to allow measurable quantities of the transuranium nuclide to be absorbed. Healy²⁸ arbitrarily assumed that incorporated plutonium would be absorbed from the GI Tract ten times higher than unincorporated plutonium. However, some data are now becoming available from the experiments of Sullivan^{29,30,31}. These data are discussed in Appendix D with a summary of the ratios between the incorporated plutonium and the controls

given in Table D-XXXV of this Appendix. The results are scattered but the only real indication of an increased uptake occurs with the feeding of a 2 day old rat that had been administered plutonium intraperitoneally or the feeding of the ileum of a young rat that had previously been fed plutonium. In view of the indication that the possible effect is uncertain, is of a relatively low magnitude and the probability that a sizable portion of the plutonium on foods (particularly plant foods that are not protected by a shell or peel) is on the surface and is not incorporated, we will not increase the uptake for the transuranium elements in foods.

The quantity of a transuranic element in the bone or liver following constant administration of 1 pCi/day is:

$$q = \frac{0.45 u I}{\lambda_B} e^{-\lambda_R t} \left(1 - e^{-\lambda_B t} \right), \quad (1)$$

where q = The quantity in the bone or liver (pCi);
 0.45 = Fraction of that entering the blood that goes to bone or liver;
 u = Uptake from the GI Tract to the blood;
 I = Intake = 1 pCi/day;
 λ_B = Biological elimination constant in the liver or bone (days⁻¹);
 λ_R = Radioactive elimination constant of the nuclide (days⁻¹).

The dose in bone and liver was calculated using conventional equations with the radiological factors from Table VI. Once again, the bone was limiting in terms of the 0.5 rem/year limit chosen and doses were evaluated for this organ. The doses increased continually throughout life so that the limiting dose is reached at the end of 70 years. These dose rates from an intake of one pCi/day are given in Table X.

TABLE I
DOSE RATES TO BONE
AFTER 70 YEARS OF CONTINUED
INGESTION OF 1 pCi/DAY

Nuclide	Dose Rate (rema/y)
^{238}Pu	5.4×10^{-4}
$^{239,240}\text{Pu}$	8.7×10^{-4}
^{241}Am	4.2×10^{-3}

These may be converted to allowable intake per day not to exceed 0.5 rema/y to the bone by dividing the calculated dose rate into 0.5. These results are given in Table II.

3. Derived Limit for Soils

The potential for contamination of soils is an important intermediate step in several of the pathways from the burial site to man. We have, therefore, chosen to consider this step in some detail so that it can be incorporated into the analysis without repeating the detail at each point. Thus, this derivation corresponds to the air and water derived limitations of the previous sections, although the models for calculation of the dose from soils are not as well developed so that the estimates of dose resulting from the soil to man are considerably more uncertain.

The two main methods of movement of transuranium elements from the soil to man are inhalation and ingestion. Absorption through intact skin appears to be low and does not contribute significantly to the intake, even though the contamination can remain on the skin for a longer period of time in the environmental situation as compared to occupational exposure.³² Wounds, while a significant source of exposure in occupational situations, do not pose a significant problem in the environmental case because of the low concentrations of the transuranium element that are allowed for other reasons. Thus, the quantity of plutonium that can enter a wound with the soil is small. We will, therefore, confine our attention to the pathways of inhalation and ingestion.

TABLE XI
ALLOWABLE DAILY OR YEARLY
INGESTION OF TRANSURANIUM
NUCLIDES TO LIMIT DOSE RATE
AFTER 70 YEARS TO 0.5 REMS/YEAR

Nuclide	<u>Intake-mCi</u>	
	Daily	Yearly
^{238}Pu	930	340 000
$^{239,240}\text{Pu}$	570	210 000
^{241}Am	120	44 000

a. Inhalation

Intake by inhalation can occur through resuspension of the materials on the ground to produce an aerosol. A discussion of the resuspension process and methods of modeling it is given in Appendix E. Briefly, there are three methods for modeling; the resuspension factor, the resuspension rate and mass loading. The resuspension factor is expressed as the ratio of the air concentration at a reference height to the quantity of the contaminant per unit area on the soil. A majority of past measurements have been expressed in this unit but it is difficult to extrapolate these measurements to other areas because of the lack of detailed information concerning the characteristics of the contaminated area and the meteorological conditions at the time of the measurement. The resuspension rate is expressed as the fraction of the contaminant removed per unit time. Studies of the erosion of agricultural soils³³ and the dust flux from soils^{34,35} have indicated that the resuspension rate is dependent upon a large number of factors including the character of the soil, the relationship between the contaminant particles and the soil particles, the wind speed, surface roughness, the vegetative cover of the area, and others. It does, therefore, tend to be site specific. While progress in measuring resuspension rates (or dust fluxes) has been rapid in the past few years, the relationships between the variables noted above and the resuspension are still tenuous so that it is difficult to assign resuspension rates to a given area. The mass loading approach relates the concentration of the contaminant on the dust in the air to the concentration of the contaminant in the soil.^{36,37} Thus, if the

concentration in the soil and the quantity of dust in the air are known, the concentration of the contaminant in the air can be derived. The examination of these modeling techniques in Appendix E leads to the conclusion that the mass loading method is appropriate for a generic study although the resuspension rate method may be more appropriate for a study of a specific site. We will, therefore, use the mass loading model, recognizing that the actual numerical application is not, and can not be, appropriate for all sections of the country.

One problem with this, as with any other modeling technique is predicting the soil fraction that is of importance in resuspension and obtaining the concentration of the contaminant in this fraction. Thus, depending upon the source of the contamination, the contaminant may be in highest concentration in a particular size range of the particles in the soil. If this is in the size range found as dust in the air, then it has been postulated that the air concentration will be higher than that predicted by use of the total soil concentration. Johnson et al.³⁸ have measured the concentration of plutonium in the soil fraction of less than 5 μm , after breaking the aggregates, and have found that the concentration was considerably higher than in the total soil sample. This approach ignores the fact that submicrometer particles exist in only very small fractions in the actual soil because they aggregate with each other and with larger particles. Chepil³⁹ has stated that particles less than 5 μm do not exist as such in ordinary soils and has demonstrated the large increase in smaller particles caused by disaggregation of the soil.²³ Gillette et al.⁴⁰ noted that less than 0.1% of the mass of a soil containing 13.7% clay (<1 μm in size) was in particle sizes between 0.3 and 1 μm again indicating the importance of aggregation in the real world. The EPA¹⁷ has proposed an enrichment factor that uses the concentration in size ranges in the soil and the mass in the same size ranges in the air to describe the effect of changing concentrations in particle size fractions. In examining contaminated soils from several areas, the maximum value of this enrichment factor was found to be 1.5. Tamura⁴¹ has proposed a "soil plutonium index," based upon particles less than 100 μm , that accounts for the fraction of the activity in a given mass fraction, the lung deposition factor for each size and the fraction of the total activity in the resuspendible fraction.

These approaches are based upon the assumption that the dust in the air at a given point is representative of the soils at that point. In fact, Gillette³⁴ has shown that the dust flux arises from the "sandblasting" of the particles in

the soil causing aggregates to shed multiple particles. Once in the air the particles will disperse downwind with the distance a function of the wind velocity, turbulence, and the particle size. The finer particles will travel for very long distances. Thus, the particle size distribution in the atmosphere is a function of the source of the dust, the distance (or distances) of its origin upwind and the deposition during travel to the point of interest. In particular, the smaller particle size distribution will be distorted by the "sorting" of the particles by differential deposition so that one expects the smaller particles accumulated from noncontaminated areas upwind to play a major role in the frequency distribution of contamination on various particle sizes. In other words, most contaminated areas are of limited size so that all of the dust in the air does not originate in the contaminated area. This effect will be most pronounced, when wind resuspension is involved, in the smaller particle sizes of interest in inhalation. It appears probable, although detailed studies have not been done, that such an effect could be of greater importance for mechanical disturbances because of the potentially higher mass loading.

Anspaugh et al.⁴² have compared calculations of air concentrations, using a mass loading of $100 \mu\text{g}/\text{m}^3$, with measured concentrations from areas contaminated with plutonium and for natural nuclides. (Table E-IV, Appendix E). In general, the agreement was good with the exception of one measurement that can be explained by the distribution of the contaminant around the point of measurement. The measured values were ambient air and did not include any contribution from a local dust cloud, caused by actions in the soil, in the vicinity of an individual.

In view of the potential influence of dust from uncontaminated areas, particularly in the smaller particle sizes, and the fact that the proposed corrections have not been demonstrated to be appropriate, we will not make correction for distribution of contamination on particle size fractions in the soil. This is further justified by the small magnitude of the corrections proposed. These are on the order of 1.5 to 2; factors that are small compared to other potential uncertainties.

The actual mass loading to use in calculations requires some consideration. The $100 \mu\text{g}/\text{m}^3$ value used by Anspaugh et al.⁴², is a conservative value as judged by the information available from the National Air Surveillance Network. However, this network measures only ambient air, which may also contain particulates from industrial activity, and does not include any allowance for

the higher concentrations near an individual causing a mechanical disturbance of the soil. The agreement found by Anspaugh et al.⁴² between the calculated concentration, using $100 \mu\text{g}/\text{m}^3$, and the measured ambient air raises a question as to the applicability of this value in describing total inhalation from all causes. A further complication arises from the fact that people spend a significant part of their time indoors. Benson et al.⁴³ concluded that indoor atmospheric particulate concentrations appear to be generally lower than outdoor concentrations, especially at high outdoor concentrations, and the particles indoors contain more organic material than those outdoors. The latter finding would appear to indicate that a significant fraction of the indoor dust may originate from sources other than the soil.

In using these observations to derive a soil limit, we will consider the mass loading of $100 \mu\text{g}/\text{m}^3$ to be appropriate for ambient outdoor air, with 50% of this value, or $50 \mu\text{g}/\text{m}^3$ for the concentration of dust indoors derived from the outdoor soil. We will consider our maximum exposed individual to work outdoors with the average concentration of $400 \mu\text{g}/\text{m}^3$ throughout a working year consisting of 50 weeks, 5 days per week and 8 hours per day. Note that this is an average value including periods of much higher concentration as well as lower concentrations. We then assume that this individual spends 10 hours per day indoors and the remainder of the time outdoors. In order to account for the higher intake of air during work, we assume that he inhales 10 m^3 of air during the eight hours of work and 10 m^3 of air during the remainder of the time. From these assumptions, the average concentration throughout the full year is $200 \mu\text{g}/\text{m}^3$.

We have previously calculated the concentration to limit the organ dose rate to 500 mrem/y at any time in a lifetime of 70 years (Table IX). This was done for three particle sizes from 0.1 μm to 10 μm , and the question of the appropriate size to use for calculations arises. Some guidance is available from measurements made in contaminated areas. Thus, Volchok et al.⁴⁴ estimated that about 25% of the plutonium in the air at Rocky Flats was respirable (i.e. would be deposited in the pulmonary region) while Sehmel⁴⁵ measured the respirable fraction at a height of 2 m as 20%. Since the ICRP lung model predicts a deposition of 25% in the pulmonary region for a 1 μm particle, this size would appear to be appropriate for these conditions. Anspaugh and Phelps³⁷ measured the size distribution of airborne particles at a contaminated area in

the Nevada Test Site and found values of 3 μm AMAD for the plutonium and 1.6 μm for the associated soil.

From these studies of real situations, it appears that a particle size of 1 μm will be reasonable to use in predictions. Assuming an inhalation rate of 20 m^3/day , the total dust inhaled per day will be 4 000 μg . From the inhalation limits designed to limit the dose to 500 mrem/year (Table IX), the permissible soil concentration can be obtained by the ratio of the limiting concentration to the estimated soil mass loading. These values are given in Table XII along with the estimated maximum dose rate to the lung.

b. Ingestion

The ingestion of the transuranium elements in soils can occur by several pathways, the most important of which appears to be food. Healy²⁸ has reviewed the question of casual ingestion of soil and pica in children (the eating of non-food substances, in this case soil) using data accumulated from studies with lead and concludes that it could be a potential problem for a few years but that control of the soils for food production would minimize this problem.

TABLE XII

SOIL LIMITS BASED UPON RESUSPENSION WITH A MAXIMUM
DOSE RATE TO BONE OF 500 mrem/yr

<u>Nuclide</u>	<u>Soil Limit ($\mu\text{Ci}/\text{g}$)</u>	<u>Lung Dose Rate (mrem/y)</u>
$^{238}\text{Pu}^a$	500	380
$^{239}\text{Pu}^a$	300	230
$^{241}\text{Am}^b$	100	8

^aICRP Class Y

^bICRP Class W

Transuranium elements can be transferred to the foodstuffs by the uptake from the soil or by contamination of the surface from direct transfer to the surface from the surrounding soil. Some of the transuranium element from the latter source will be removed by washing, peeling or shelling of the plants before eating.

Studies of the plant uptake of transuranics have indicated that there is a marked degree of discrimination against the movement of the transuranics into plants. However, this discrimination can be reduced drastically by the incorporation of agents, such as chelating agents, into the soil, although Ballou et al.⁴⁶ have shown that the chelate is stable and the additional plutonium taken up by the plant is not well retained in the body upon ingestion of the plant. There is a variation in the uptakes measured with different plants and different soils so that there is some difficulty in assessing a proper uptake factor for calculations. This is compounded by the fact that measurements are not available on all food types. Schulz⁴⁷ notes that measured plutonium concentration ratios (the ratio between the concentration in the plant and the concentration in the soil) have varied from 10^{-10} to 10^{-3} while americium ratios varied from 10^{-7} to 10^1 . The particle size and composition of the transuranium elements were estimated to account for a factor of 10^3 in these ratios while the plant part and plant species can account for another factor of 10^3 . Soil factors, such as pH and organic matter, can account for a factor of 100, and chelation for at least a factor of 100. Another source of variation is the difference in experimental conditions, leading to several orders of magnitude difference in the result.

The above values are for root uptake because the plants were grown in controlled conditions where foliage contamination was negligible. In the field, foliar contamination from resuspended materials depositing on the plant is of importance. Cataldo and Vaughan⁴⁸ note that early work on worldwide fallout and more recent work on plants grown on contaminated soils indicate that foliar retention and absorption may be as important, and in some cases more important, as root uptake. They also note that the leaching or removal of the contaminant is a function of the particle size of the contaminant. Thus, reports of rapid removal from vegetation during fallout studies were undoubtedly due to the large sizes of particles encountered. In more recent work, the plutonium was much more difficult to dislodge by rain or weak acid leaching. This, of course, raises questions as to the fraction removed by washing.

Data are available on the fallout plutonium content of New York foods.⁴⁹ One cannot claim that the plutonium arises completely from the soils or resuspended contaminant on the surface of the foods because plutonium still exists in the atmosphere and a part of the food content may well be due to direct deposition on the plants. The use of concentration ratios derived from these data is attractive, however, because they include a wide variety of foodstuffs, including meat and milk, and any direct deposition from current fallout will result in a conservative estimate of the concentration ratio.

In the derivation of a concentration ratio one must also have a value for the plutonium in the soil at the place where the food originated. The deposition of plutonium over the country is nonuniform with values ranging from 0.7 mCi/km² at Los Angeles to 2.5 mCi/km² in S. Dakota.⁵⁰ New York has a high value of 2.4 mCi/km². Since the food eaten in New York comes from a large part of the country, it is not clear exactly what value should be used. An average of the 19 locations across the country reported in reference 50 is 2 mCi/km². In view of the widespread source of the foods in New York, this value was chosen.

In order to obtain a soil concentration so that concentration ratios could be calculated, it was assumed that this plutonium is distributed through a 20-cm depth of soil by cultivation and the concentration ratios for the various foodstuffs were calculated. The measured plutonium concentrations along with the estimated concentration ratios and the per capita annual intake of that foodstuff are given in Table XIII.

The uptake of americium from the soil is less well known than that of plutonium. In a Workshop on Environmental Research for transuranium elements,⁵¹ held in Seattle Washington in 1975, a plant uptake panel, after review of the then available data on studies of uptake in plants grown in pots, concluded that americium is taken up by plants 100-1000 times more readily than is plutonium. However, Shultz⁴⁷ noted that a previous experiment with barley and soil from the Nevada Test Site indicated a ratio of only 8 between the uptake of americium and plutonium. Schreckhise and Cline⁵² grew cheatgrass, peas, barley and alfalfa in small outdoor lysimeters and noted that the uptake of both americium and curium was 10-20 times that of plutonium. Bennett⁵³ has been studying the ²⁴¹Am and ²³⁹Pu content of the New York City diet. While this work is incomplete, preliminary indications are that there is little or no preferential uptake of the americium over the plutonium originating from fallout.

TABLE XIII

PLUTONIUM IN NEW YORK FOODS - 1972

<u>Food Item^a</u>	<u>Plutonium</u>	<u>Conc.</u>	<u>Per Capita Annual Intake</u>	
	<u>Concentration</u>	<u>Factor</u>	<u>Food</u>	<u>Plutonium</u>
	(10 ⁻¹² Ci/kg)		(kg/yr)	(10 ⁻¹² Ci/yr)
Plant-Derived				
Bakery Products	0.0085	1.3x10 ⁻³	44	0.37
Whole Grain Products	0.0060	9.1x10 ⁻⁴	11	0.066
Fresh Fruit	0.0051	7.7x10 ⁻⁴	59	0.30
Dry Beans	0.0049	7.4x10 ⁻⁴	3	0.015
Fresh Vegetables	0.0043	6.5x10 ⁻⁴	48	0.21
Root Vegetables	0.0035	5.3x10 ⁻⁴	10	0.035
Flour	0.0028	4.2x10 ⁻⁴	34	0.095
Rice	0.0015	2.3x10 ⁻⁴	3	0.004
Potatoes	0.0013	2.0x10 ⁻⁴	38	0.048
Macaroni	0.0012	1.8x10 ⁻⁴	3	0.004
Canned Vegetables	0.0009	1.4x10 ⁻⁴	22	0.019
Fruit Juice	<0.0003	<5x10 ⁻⁵	28	<0.007
Canned Fruit	<0.0002	<5x10 ⁻⁵	<u>11</u>	<u><0.002</u>
Total Plant-Derived Diet	0.0037 ^b	5.6x10 ⁻⁴	314	1.17 - 1.18
Animal-Derived				
Poultry	0.0033	5x10 ⁻⁴	20	0.066
Meat	0.0026	3.9x10 ⁻⁴	79	0.20
Eggs	0.0012	1.8x10 ⁻⁴	15	0.019
Milk	<0.0003	<5x10 ⁻⁴	<u>200</u>	<u>0.064</u>
Total Animal-Derived Diet	0.0010 ^b	1.5x10 ⁻⁴	314	0.29 - 0.35
Total Diet				1.5

^aFood categories and quantities are based on Department of Agriculture food purchase surveys.

^bValues are calculated from total per capita annual intake of food and of plutonium.

These brief summaries indicate the wide variability in the data presently available. Perhaps the detailed analysis of the diet being performed at the Environmental Measurements Laboratory will shed further light. At the moment, however, the evidence seems to indicate that the estimate of the plant panel is high in that estimates of uptake of 100-1000 times that of plutonium do not seem to be confirmed by other studies. We tend to place more confidence in the estimates made from experiments in which plutonium and americium are both measured in the same experiment. This would indicate a factor on the order of ten as the upper value, with the data of Bennett⁵³ indicating no difference. In this study, we will use concentration ratios for ²⁴¹Am in plant materials ten times those given for plutonium in Table XIII. This uptake factor is different than those in previous documents because of the use of data accumulated in recent years.

There is little information on uptake in animal products upon which to base a judgement. Normally, one would state that the animal concentration ratios should be increased by the ratio of the uptake from the GI Tract of these two nuclides or a factor of five. However, the review given in Appendix D indicates that the ICRP value for uptake from the GI Tract may be somewhat high and Bennett's observation⁵³ on the ²⁴¹Am in the New York diet indicates that the increase of ²⁴¹Am over ²³⁹Pu of a factor of ten may be somewhat high for actual foods grown and prepared under real conditions. The factor of ten increase for ²⁴¹Am in concentration factor used for plant foods will, therefore, also be used for the animal foods.

From these concentration ratios one can estimate a soil concentration corresponding to the daily or yearly ingestion limits given in Table XI by

$$I(\text{Conc. Ratio} \times \text{Soil Conc.} \times \text{intake}) = \text{pCi/day}$$

or

(2)

$$\text{Soil Conc.} = \frac{\text{Allowable pCi/day}}{(\text{Conc. ratio} \times \text{daily intake})}.$$

The appropriate foods will differ depending upon the foods that are considered to be grown by the maximum exposed individual. At the present time, the bulk of the foods eaten by most people are grown elsewhere. There is a sizeable number of people, however, who maintain home gardens for vegetables and

a much smaller number who grow the bulk of their own food, including meat. Soil limits to provide a limiting dose rate of 500 mrem/y to the bone at any time in a 70 year lifetime for several assumed patterns of food growing in an area are given in Table XI. The inhalation limit from Table XIV is included for comparison.

As can be seen from Table XIV, ingestion can be a significant addition to the dose for plutonium if a major portion of the food is grown on the contaminated area. For ^{241}Am the ingestion dominates the dose under the assumptions used in these calculations.

The total dose to any individual will be contributed by both pathways so that a weighted summation of the inhalation and ingestion limits are needed. In view of the uncertain practices to be carried on in the future, we have considered the ingestion limit based upon the total diet to be appropriate for our hypothetical maximum exposed individual. This produces overall soil limits of 350 pCi/g for ^{238}Pu and 200 pCi/g for $^{239,240}\text{Pu}$. For ^{241}Am the inhalation component is small but the weighted limit is 14 pCi/g. These limits, along with the associated lung dose rates are given in Table IV.

TABLE XIV
SOIL LIMITS BASED ON SEVERAL PATTERNS OF GROWING FOODS

Foods Grown	Soil Limits (pCi/g)		
	^{238}Pu	$^{239,240}\text{Pu}$	^{241}Am
Fresh and Root Vegetables	8 600	5 300	110
Above + Fruits	4 000	2 500	51
Above + Poultry and Eggs	3 600	2 200	46
Above + Meat, Milk, Potatoes and Dried Beans	1 400	890	19
Total diet	1 200	750	16
Vegetarian ^a	970	600	13
Inhalation Limits ^b	500	300	100

^aCalculated as a 628 kg/y total intake with a concentration ratio of 5.6×10^{-4} for plutonium and 5.6×10^{-3} for ^{241}Am .

^bFrom Table VII.

TABLE XV
DERIVED SOIL LIMITS AND ASSOCIATED LUNG DOSE RATES

Nuclide	Soil Limit (pCi/g)	Lung Dose Rate (mrem/y)
^{238}Pu	350	270
$^{239,240}\text{Pu}$	200	150
^{241}Am	14	1

g. Daughter Products

The immediate daughter products of the nuclides considered in this study are given in Table IV of section II while the complete chains and buildup curves for the daughters are given in Appendix B. All of the plutonium isotopes decay to activities of daughters that are on the order of 10^{-4} to 10^{-5} times those of the parent (with the exception of ^{241}Pu - ^{241}Am which are treated explicitly). In general, from the studies available this decrease in quantity will outweigh the increased mobility of the daughters so that the dose rate from these daughters will be lower than the parents.⁵⁴⁻⁵⁶

There is one daughter nuclide, however, that may have potential for increasing the dose over that of the parent. This is ^{237}Np , with a half-life of 2.14×10^6 years, arising from the decay of ^{241}Am . Unfortunately, data are not available to permit a realistic estimate of the potential hazard. Indications are given in the few experiments discussed in Appendix D on the GI uptake of the nitrate. Data abstracted from the tables in this Appendix on the retention in the skeleton of ^{237}Np are given in Table XVI. As can be seen, the skeletal content is 5×10^{-3} to 10^{-2} of the intake as compared to the values used in this report of about 5×10^{-5} for plutonium or 2×10^{-4} for americium. (Corrected for the 45% deposition in the skeleton). In addition, Schreckhise and Cline⁵² have noted plant uptakes ranging from 2 200 to 45 000 times that of plutonium in cheatgrass, peas, barley and alfalfa. The product of these two factors indicates that the ingestion pathway for ^{237}Np could be 2×10^5 to 9×10^6 more important than for plutonium or 4400 to 2×10^5 more important than for ^{241}Am assuming equal dose per unit of intake. Since the quantity of ^{237}Np is 1/5000th

TABLE XVI
SKELETAL CONTENT OF ^{237}Np FOLLOWING
INTRAGASTRIC ADMINISTRATION OF THE
NITRATE TO ADULT RATS

Number of Animals	Skeletal Content (% Administered Dose)
11	^a
15	0.63 ^b
7	0.54 ^b

^aReference 57.

^bReference 58.

of the ^{241}Am , these data would indicate that a net hazard greater than that of ^{241}Am could possibly result when the decay of ^{241}Am is completed.

There are, however, experiments that indicate that biologically incorporated ^{237}Np is absorbed by factors of 10-20 more poorly from the GI Tract than the nitrate.⁵⁸ In addition, the greater mobility of the ^{237}Np in the environment will result in more rapid dispersion so that concentrations at any given location may well be lower than those of the parent.

These data are not adequate to permit detailed accounting for this nuclide and it does appear that the doses will be somewhat less than those for ^{241}Am . However, the doses may extend over periods of millions of years rather than the thousands of years from ^{241}Am .

B. Pathway Estimates for Shallow Earth Burial

As a prelude to the discussion of potential exposures from both shallow earth burial in this section and deeper earth burial in the next section it is well to consider the objective to be served by such estimates. It is clear that the scenarios used in the calculations are devised and may well represent situations that will not develop at any given site. Even if they do develop, the conditions at the time may be so greatly different than we envision, that the exposures may be considerably lower or slightly higher than we estimate.

However, we have tried to pick critical coefficients in a conservative manner so that the probability is that the doses will be lower. Our goal was to provide reasonable limitations on the transuranics placed in the ground so that future harm, if any, from uses of this ground will be minimal and the area could be used in a reasonable fashion after the control period. It is recognized that there are many uncertainties, including some that we may not be aware of, in both the future and in our present understanding of the behavior of the transuranium elements in the environment. However, there have been many studies that tend to bolster our confidence in the latter factor and we believe that the approach of using the maximum exposed individual and examination of a number of potential pathways to obtain the limiting one (or ones), provides reasonable assurance, that the limits so-derived will meet their goal even though one cannot assure that the limiting dose used in the calculations will be met in every conceivable case.

The question arises of site-specific limits based upon the conditions at a given site or, at least, limits more specific to a given region such as the arid steppes or deserts of the west as compared to the high rainfall areas of the Eastern United States. Since we are dealing, in many of the cases, with very long-lived radionuclides, it is not prudent to believe that current conditions will last through the effective life-time of these materials. Appendix F is a brief description of the conditions that have occurred in the United States over the past 250 000 years. Here it is seen that climatic changes accompanied the ice ages with changes in river courses and even the coast lines were somewhat different than at present. Also, it is noted from the previous calculations of dose rates from plutonium in soils, that the exposures to people may not be as greatly different in the arid and wet areas as is generally believed because of the contribution from uptake in foods and in ingestion. For these reasons we believe that a generic limit for all areas is useful, although it will be noted in the pathway studies to follow that there may be wide differences from one site to the next for a particular pathway.

Implicit in the scenarios used and the calculations of dose is the assumption that a "good" burial site is used in all areas. It is beyond the scope of this study to provide criteria for a good site but it is a premise of the study that such criteria will be developed and used for the DOE locations.

The scenarios for estimation of dose to man and limitation on quantities buried, cover several different mechanisms of escape of the transuranium

nuclides or exposure of individuals. For shallow burial, they include erosion to uncover the wastes with transport to a stream and exposure from use of the stream; erosion to uncover the wastes with people then taking up residence on the area; movement of the transuranium elements to an aquifer and then to a stream; and intrusion by individuals into the waste mass.

It should be noted that the pathway calculations in this section do not take into account any dilution by less contaminated wastes or soil in those cases where the hazard depends largely on the average concentration. This was done to simplify the presentation in this section. Appropriate values will be included before the final recommendations are made. Similarly, those nuclides which are to be limited by the ingrowth of longer-lived nuclides, such as ^{241}Pu or the curium isotopes, will not be discussed during the calculations but will be included in the recommendations.

1. Erosion with Transport to Stream

In this scenario, we will consider the erosive effects of wind or water on the surface of the burial ground with eventual elimination of the soil covering of the wastes. Continued erosion, once the wastes are uncovered, will result in transport of the degraded wastes to surface streams where potential exposure results from drinking of the water, use of the water for irrigation and eating of fish or other aquatic life raised in the water. The alternate possibility, that the wastes will be buried even deeper, exists but this appears to be unlikely for most of the DOE sites because they are located in areas of net erosion.

a. Erosion Rates

Rates of erosion vary widely depending upon the rainfall, type of soil, slope of the ground and, in particular, on human activities. For croplands in midcontinent, soil loss with runoff averages 2-6 tons per acre.⁵⁹ Some examples of erosion rates and runoff under different conditions are given for agricultural fields in Table XVII.⁶⁰ This table indicates a variation in erosion rate by several orders of magnitude from the clean tilled field to the field with dense cover. The U.S. Department of Agriculture⁶¹ has provided an equation for the estimation of soil losses from agricultural fields which provides some indication of the influence of various factors. Rainfall is obviously a factor and the erosion loss includes the cumulative effects of the

TABLE XVII

ANNUAL SOIL AND WATER LOSSES PER ACRE FROM FIVE WIDELY SEPARATED
TYPES OF LAND UNDER CONDITIONS OF CLEAN TILLAGE AND DENSE COVER OF VEGETATION^a

Soil, location and years of measurements	Average Annual precipitation	Slope	Clean-tilled Cron		Dense cover- thick-growing Cron		Approximate number of years to remove 18 in. of soil	
			Annual Soil loss (tons)	Annual water runoff ^b ($\frac{1}{2}$)	Annual soil loss (tons)	Annual water runoff ^b ($\frac{1}{2}$)	Clean Tillage	Dense Cover
	(in)	(%)						
Shelby silt loam, Bethany, MO, 1931-35.	88	8.0	68.78	28.31	0.29	9.30	16	3 900
Kirvin fine sandy loam, Tyler, TX, 1931-36.	104	8.75	27.95	20.92	0.124	1.15	49	11 100
Vernon fine sandy loam, Guthrie, OK, 1930-35.	84	7.7	24.29	14.22	0.032	1.23	50	38 200
Marshall silt loam, Clarinda, IA, 1933-35.	68	9.0	18.82	8.64	0.06	0.97	48	15 200
Cecil Clay loam, States- ville, NC, 1931-35.	115	10.0	22.58	10.21	0.012	0.33	51	95 800

^aMeasurements at the soil and water conservation experiment stations of the Soil Conservation Service.

^bOf total precipitation.

many moderate-size storms as well as the effects of the occasional very severe ones. The rainfall-erosion index is the product of the total kinetic energy of the storm times the maximum 30-minute intensity. An iso-erodent map for the Eastern 37 states indicates this factor to vary from 50 to 600. The maximums occur along the Louisiana, Mississippi and Georgia coasts. It is noted that in the mountainous states, west of the 104th meridian, the sporadic rainfall pattern of the mountains makes generalization difficult. In the Pacific Northwest snowmelt carries a large part of the field erosion so that the practical value of the rainfall-erosion equations has not been established for this area.

A second factor is the soil erodibility which accounts for the fact that some soils erode more readily than others even with all other factors the same. Soil properties that influence erodibility by water are those that affect the infiltration rate, permeability and total water capacity and those that resist the dispersion, splashing, abrasion and transporting forces of the rainfall and runoff. The soil-erodability factor varies from 0.69 for a Dunkirk silt loam to 0.03 for an Albia gravelly loam.

The erosion rate also depends strongly on the length of the slope of the ground and the gradient of the slope. For a length of 60 m (200 feet) the soil-loss ratio varies from about 0.3 at a 2% gradient to about 6 at a slope of 20%.

The effect of crops on the land is complicated and must take into account the coincidence between erosive rains and the stage of the crop at the time, as well as other crop management techniques. However, with established meadows of grass, alfalfa or clover, the soil loss rates are 0.4 to 2% of that from fallow land.

"Tolerance" values for soil loss have also been set by judgment as to effects of such losses on the long-term productivity of the soils. The U.S. Department of Agriculture⁶¹ indicates, that for the soils in the U.S., this rate ranges from 1-5 tons per acre per year depending upon soil properties, soil depth, topography and prior erosion. A deep, medium textured, moderately permeable soil that has subsoil properties suitable for plant growth has a tolerance of 5 tons per acre per year. Soils with a shallow root-zone or with a high percentage of shale at the surface, have low tolerances. Smith and Stamey⁶² have given a range of 0.5 tons to 6 tons per acre per year depending upon whether the soil is shallow or deep.

These data are all based upon agricultural soils where erosion is increased by cropping procedures. In undisturbed areas, rates of regional erosion have been estimated from the volume of deposits in closed systems. These calculations⁶² show that for the Appalachian region, the past rate for 125 million years averaged 6.2 cm per 1 000 yr (0.74 tons/acre annually). For the entire Mississippi drainage basin the past rate was 4.6 cm per 1 000 yrs (while the present rate is 4.2 cm/1 000 yrs). Based on the production of ⁴⁰Ar in the atmosphere from ⁴⁰K in the crust, the weathering of igneous rock is estimated to have been proceeding at a rate of 0.68 cm/1 000 yrs (0.982 tons of igneous rock per acre annually).

Judson⁶³ has noted that a maximum rate of erosion is reached in areas of limited rainfall (>25 cm/yr) with decreased rates in more humid as well as in more arid climates. He also stated that the activities of man can increase the regional erosion rates by large factors.

While these erosion rates are low under natural conditions, the question of the effect of later use of the area for living arises. Urbanization and the creation of roadcuts ~~has been found~~ to increase erosion to the extent of 75-200 ~~cm per year~~ per year, with depths of erosion on roadcuts of 3-6 cm.⁶⁴ However, the transition from relatively stable rural terrain to a steady-state urban environment may take only from 2 yrs to 10 yrs, depending on the size of the drainage area, the intensity of home building, and sequence of placement of streets, water and sewage systems, and other utilities.⁶⁵ Once construction has ceased there follows a steady decline in erosion rate to levels often below the preconstruction rate.⁶⁵ Thus, the overall impact of urbanization of a burial site may be difficult to assess: it could result in more rapid exposure of buried waste under one set of circumstances, and considerably delayed exposure in others. These several erosion rates are summarized in Table XVIII.

As can be seen from this discussion, erosion rates are highly variable with a strong influence being man's activities. One would expect gully formation in disturbed earth of the burial ground to be increased in the years following the emplacement of the wastes. However, during this period, the burial ground will be monitored and actions can be taken to correct the situation. We feel that erosion control should be a major factor in choosing a burial ground and that specific steps to provide a permanent type of erosion control should be taken.

Under these conditions, the choice of a specific erosion value is difficult. However, values of 1 to 6 tons per year would seem to be

TABLE XVIII
EROSION RATES UNDER VARYING CONDITIONS

Soil or Rock Description	Use	Erosion Rate (ton/acre per year)
Igneous rock	Geologic past	0.08
Appalachian Mountains	Geologic past	0.7
Midcontinent farmland	Typical farming (other than clean tilled)	0.5-6
	Clean tilled	10-60
Urban or suburban	During construction	70-200

appropriately conservative. If we assume a bulk density for soil of 1.5 g/cm^3 , this would require from 2 000 to 13 000 years to remove the two meters of cover from the waste pits with erosion continuing for 4 000 to 25 000 years through the waste mass.

b. Sediment Transport

Once the cover has disappeared, the sediment from the waste pits will wash down-slope to the nearest stream. It is anticipated that the transuranium elements will be attached to the soil particles and will be distributed on the surface between the burial grounds and the streams along the erosion paths. The dose to people from this intermediate area will be included later in the pathway of people living on the area. Here we are concerned with the dose resulting from transport in the stream.

Once in the stream, the contaminated sediments will mingle with the sediments already present and will be moved downstream by sediment transport. Such a mechanism of movement is different from the dilution in the water and transport downstream with the water but is more realistic when the absorptive capacity of the sediments is taken into account.

A generalized model for sediment transport is developed in Appendix G, using data primarily from the transport of radioactive materials in the Columbia, Tennessee and Miami River systems and several Atlantic coast estuaries. The sediment is transported as individual particles, spending part of the time being carried in water currents (suspended load) and part of the time

in the sediments on the bed of the stream (bed load). The entire mass of the sediment can be treated as a single entity moving with its characteristic velocity and dispersion. The results of the study indicate an exponential decrease in concentration as one progresses downstream from the source. However, the rate of decrease is greater for the Columbia River than for the Tennessee River with the Tennessee river also showing an even slower rate of decrease than that present initially beyond about 30 miles, presumably due to the extensive system of dams in the lower reaches. The model predicts the sediment concentrations 30 miles down-river from the source to be about 1% of that in the source area for the Columbia River and about 20% for the Tennessee River, indicating the influence of river velocity and channel.

A second phenomenon occurs in sediment transport -- sorting of the sediments by particle size. The smaller sediment particles are more easily carried in suspension than the larger particles. When these particles reach an area of low flow (such as the upper reaches of a dam) they will deposit in this area. Since such smaller particles have a much larger surface area than the larger ones, they have a higher exchange coefficient and can carry larger amounts of a contaminant per unit weight. Thus, a mechanism for reconcentrating the contaminant on a weight basis exists in such pools. In the modeling, the ratio of the total sediment-bound concentration of the contaminant in areas of high deposition to the total annual release of contaminant was used as a parameter. A value of 2 pCi/g per Ci/yr released was considered to be appropriate for calculations of the dose at the maximum point in the stream (remembering that the sediment concentrations then falls off with distance so that doses further downstream will be lower).

a. Drinking the Water

While the great majority of the transuranics are in the sediments, there is some interchange between the water and the sediment. This may not be a true solubility but may actually be due to small particles. The net result could be similar, because such small particles can be dissolved in body fluids and can be absorbed. We will use two primary environmental sources of data for this relation for plutonium. Rogers,⁶⁶ at Mound Laboratory, measured the water and sediment in the river, ponds and abandoned canals near the Mound Laboratory. The contaminant in this case was heat-source plutonium that was mostly ²³⁸Pu by

activity. His measured values for the ratios between activity per cm^3 of water and activity per gram of sediment are given in Table XIX.

Hakonson, et al.⁶⁷ measured the sediment concentration and water concentration during a runoff event in one of the canyons that has been used for disposal of effluent from a liquid waste treatment plant at Los Alamos. His data are given in Table XX. They also noted that about 1.5% of the plutonium was in the water phase with the remainder carried by the sediment. In this case, the flow was very turbulent with good mixing between the sediment and the water although the time of the entire event was relatively small.

From these data, we will consider a value of $5 \times 10^{-5} \text{ g/cm}^3$ to be representative of this ratio. This is lower than that measured by Hakonson in very turbulent conditions, using the sediment being transported as the reference, but is somewhat higher than Roger's value in relatively stagnant conditions using bottom sediments.

TABLE XIX
RATIO OF WATER TO SEDIMENT CONCENTRATIONS
NEAR MOUND LABORATORY

Location	Ratio - Water to Sediment (g/ml)	
	Sediment 0-2 cm	Sediment Surface
South Pond	2×10^{-5}	$<2 \times 10^{-5}$
North Pond	2×10^{-5}	$<4 \times 10^{-5}$
Great Miami River	8×10^{-6}	-
North Canal	7×10^{-6} - 1×10^{-5}	2×10^{-5}
South Canal	-	4×10^{-5}
Overflow Creek	-	1×10^{-5}

TABLE XX
RATIO OF WATER TO SEDIMENT CONCENTRATIONS
IN LOS ALAMOS CANYON RUNOFF

Time After Runoff Started Min	Ratio - Water to Sediment (g/ml)	
	Water Surface	Bottom of Water
5	7×10^{-5}	5×10^{-4}
160	8×10^{-5}	1×10^{-4}
260	2×10^{-4}	-

Similar data are not available for americium although it is, in general, considered to be more mobile in the environment than plutonium. Martell,⁶⁸ for example, postulates that this increased mobility in soils may result in more rapid movement from the root zone, thereby alleviating, somewhat, its greater uptake in plants. This would imply lower adsorption on the soils and higher concentrations in the water. We will consider the americium to have a ratio of water to soil ten times that of plutonium or 5×10^{-4} g/cm³.

From this ratio, in the region where the sediments contain 2 pCi/g, we would expect that the water would contain about 1×10^{-4} pCi/cm³ of plutonium or 1×10^{-3} pCi/cm³ of americium. From the earlier study on ingestion, the daily intake over 70 years required to eventually deliver 0.5 rems per year is 570 pCi for ^{239,240}Pu and 120 pCi for ²⁴¹Am. (Note that ²³⁸Pu will have disappeared by radioactive decay before this pathway becomes of importance.) The ICRP Reference Man⁶⁹ provides estimates of the water intake for adult men and women and a 10 year old child. Their estimate of the consumption of tap water ranges from 100 to 200 cm³/day. However, a larger value occurs under the heading "other", which means such items as soft drinks or beer. The sum of the tap water and "other" is 950 cm³/day for the child, 1 200 cm³/day for the adult woman, and 1 650 cm³/day for the adult man. We will consider an intake, as water, or drink derived directly from the water, of 1 650 cm³/day to be appropriate. This translates to an allowable concentration of 0.35 pCi/cm³ for plutonium and 0.073 pCi/cm³ for americium if the water is used exclusively and continuously for 70 years.

It is to be expected that some of the transuranium elements would be removed in water treatment processes before the water is used to any extent by

humans. Detailed data on the degree of removal are not available but it would be expected that some fraction would be removed to the sludge by the current flocculation and clarification procedures. However, it is impossible to predict future processes that may be used and to predict the degree of transuranium element removal. Thus, such removal will not be considered.

These concentrations in the water would, from the ratios given above, result from 7000 pCi/g for $^{239,240}\text{Pu}$ and 150 pCi/g for ^{241}Am in the sediments. Since each curie per year released results in 2 pCi/g at an impoundment, the allowable release to the river is 3500 Ci/yr of $^{239,240}\text{Pu}$ and 75 Ci/yr of ^{241}Am .

d. Use of Water for Irrigation

In a second pathway, the use of water for irrigation will disperse the sediments and dissolved contaminant over agricultural land with eventual buildup in the soils until an equilibrium is reached between the overall rate of loss from erosion and the rate of application. The fact that the land is irrigated implies that it is used for agriculture with periodic cultivation and mixing of the soil. We will assume a mixing through 12", or about 30 cm of soil. The use of two feet (60 cm) of water per year, with an assumed sediment content of 1 g/L containing 2 pCi/g would add 0.06 g of sediment or 0.12 pCi per year to each cm^2 or about 0.0027 pCi/g of soil. For americium, the activity added may be associated with the water to a greater degree than for plutonium but, upon reaching the soil, it will be readsorbed on the soil to behave the same as the sediment. We will consider the erosion rate in this field to be the same as for the burial area or 6 tons per acre per year or about 0.09 cm/yr. This is about 0.3% of the mixed layer per year. At equilibrium, then, the soil would contain $q/(0.003 + \lambda)$ pCi/g where q is the annual rate of addition and λ is the decay constant in years^{-1} of the isotope. Thus, for the long-lived alpha emitting plutonium isotopes, the equilibrium concentration in the soil becomes 0.9 pCi/g per Ci per year released. For ^{241}Am , the equilibrium soil concentration is 0.59 pCi/g.

The above calculation of the soil content was based upon a release of 1 Ci/yr to result in 2 pCi/g in the sediments at the maximum location. Earlier in this section of the report, we derived limits for soil, for all uses, of 200 pCi/g for $^{239,240}\text{Pu}$ and 14 pCi/g for ^{241}Am . While these values included living on the area, we cannot preclude a farm in the midst of an irrigated area and we will use these values for calculation of the discharge rate. By direct ratio of

the allowable concentration and the concentration calculated to exist, we arrive at permissible discharge ratio to the stream of 220 Ci/yr for $^{239,240}\text{Pu}$ and 24 Ci/yr for ^{241}Am .

2. Eating of Fish

A third pathway of importance to those living in the vicinity of the river is the ingestion of fish and possibly other products obtained from the stream. Appendix H provides a review of current data on the uptake of transuranics by the biota in an aquatic medium. Large concentration ratios (ratio of concentration in organism to concentration in water) are measured for algae and plankton with a general decrease in concentration ratio as one goes up the trophic chain. However, even for fish the concentration ratio can vary depending upon the type of fish. Bottom feeders, such as carp or suckers, are estimated to have concentration ratios of 250 for plutonium and 2 500 for americium. While not considered as an edible fish by many people, there are those who do eat them. At the other end of the range, those fish that live by eating other fish have ratios estimated at 5 for plutonium and 50 for americium, or about 50 times lower than the bottom feeders. For these estimates we will use a ratio of 25 for plutonium and 250 for americium, recognizing that there may be variability. These are taken as concentration ratios for the parts as eaten, although the data in Appendix H are not that clear on this point.

Once again, the intakes to result in 500 $\mu\text{rem/yr}$ to the bone are 570 pCi/day for $^{239,240}\text{Pu}$ and 120 pCi/day for ^{241}Am . The data presented in ICRP-23⁶⁹ indicate that people in the U.S. ate 22 g of fish and seafood per day in 1955. In other regions of the world the quantities eaten per day range from 12 g in the Near East to 38 g in Europe. This includes both seafood and fish with the values for other regions of the world based upon "landed" weight, thus ignoring the waste. The quantity actually eaten by an individual will undoubtedly depend upon opportunity as well as taste. In regions where fish are available it is possible that the sportsman and/or the family on a subsistence diet who can catch their own fish would eat more. We will consider that our maximum individual will eat 100 grams per day. From the drinking water calculation, a release of 1 Ci/yr resulting in 2 pCi/g of sediment would produce 1×10^{-4} pCi/cm³ of plutonium or 1×10^{-3} pCi/cm³ of americium in the water. This would result in concentrations of 2.5×10^{-3} pCi/g for $^{239,240}\text{Pu}$ or 0.25 pCi/g for ^{241}Am in the fish. The ratio of the permissible daily intake of the nuclides to

these values, multiplied by the daily fish intake of 100 g/day provides the allowable releases of each nuclide. These are 2300 Ci/yr for $^{239,240}\text{Pu}$ and 5 Ci/yr for ^{241}Am .

A situation can be postulated that does not exist at the present time. This is the growth of algae or similar materials for use as a food source. Here concentration factors are estimated to be 5 000 for plutonium and 50 000 for americium or some 200 times the value used for fish. Thus, ingestion of 100 g/day of such material would result in greater doses at the discharge rates estimated. Since we have no way of knowing whether this is a real possibility or of estimating the amounts that will be ingested by individuals, this pathway will not be taken into account explicitly.

The estuary into which the river eventually empties will pose a similar situation with fish to that of the river except that it now involves seawater. From Appendix H, the data are not adequate to differentiate between the uptake in seawater and fresh water so that the concentration factors are taken as the same as for the river. There is some concentration of the sediments in coves or still areas but there is also some decrease in concentration with distance down the river. It may be noted that Danster⁷⁰ described the use of an edible seaweed in areas along the coast in the vicinity of discharge from the Windscale plant. From a survey in the region, consumption ranges up to 172 g/day. Obviously, if this were postulated to occur in the cove or estuary, a situation would be presented where the greater uptake from the water by the seaweed could lead to high individual potential doses although the decrease in sediment concentration down river would alleviate this situation if the source were 80 or more miles upstream from the estuary.

f. Conversion to Waste Concentration

The foregoing pathway discussions estimated the rates of release to a river system with the transuranics carried on sediments. These release rates can be approximately converted to concentration in the burial ground by use of an assumed erosion rate of six tons per acre per year as used in the derivation of the sediment model. The model burial ground has an eventual area of 10^6 m^2 or about 250 acres. The rate of erosion of 6 tons per acre per year produces a total of 1500 tons of sediment per year. This process will require several thousands of years for the cover material to be eroded and the wastes to be reached. In 1000 years the ^{241}Am will be reduced to 22% of its initial activity

and in 2000 years to 5%. Thus, at the time that actual release by erosion is postulated to start, the ^{241}Am will be reduced to 5% of the initial quantity.

The limiting quantity released to the river per year can be related to the quantity in the burial ground by assuming that the release is associated with the 1 500 tons per year erosion rate. This value is given in conventional U.S. tons which amounts to 1 360 metric tons. The average concentration in the burial ground from this weight and the release rate in curies per year calculated earlier is given in Table XXI.

2. Movement in Local Waters

There are two main types of movement to local waters considered as important in these studies. The first is the possibility of leaching of the transuranium elements from the wastes and movement of the leachate through any intervening sediments to an aquifer underlying the burial ground, where the contaminant will move down-gradient to areas of possible use. The second, then, arises from the possible movement of water in this aquifer to a surface stream resulting in the introduction of contaminants to this stream. The latter movement is similar in result to the introduction by direct erosion into the stream but is included to assure that it may not be of greater importance.

TABLE XXI
LIMITING CONCENTRATIONS OF $^{239,240}\text{Pu}$ AND ^{241}Am IN THE BURIAL
GROUND BASED ON EROSION TO STREAMS

Pathway	$^{239,240}\text{Pu}$		^{241}Am	
	Allowable Release (Ci/y)	Burial Ground Conc. (nCi/g)	Allowable Release (Ci/y)	Burial Ground Conc. (nCi/g)
Drinking	3 500	2 600	75 ^a	1 100
Irrigation	220	160	24 ^a	180
Fish	2 300	1 700	5 ^a	74

^aThese values are increased by a factor of 20 to allow for decay in the 2000 years before erosion of the wastes starts.

A third method of movement is to local seeps or springs if the wastes are buried in fractured, impermeable formations in a region with high rainfall where the leachate can accumulate in the waste mass and then percolate through fractures. This is discussed but not included in the limits because it is assumed that future burial criteria will eliminate such undesirable conditions.

a. Movement to and in Aquifer

The ground water in many areas of the country is an important resource that is used for domestic, industrial, and agricultural purposes. A vital aspect of the management of wastes from all sources, including nuclear, is the prevention of pollution that will degrade this resource.

For buried wastes, any contaminant must find its way through the earth materials between the burial site and the aquifer underlying the area. For many DOE sites this is several hundreds of meters, while for others the bottom of the burial pit is within a few meters of the aquifer (Appendix B). The usual driving force to move the wastes into the aquifer is percolation of water from the surface through the wastes and into the ground. In arid regions this driving force is low since only a small fraction, if any, of the rainfall penetrates as far as a few meters into the ground with the majority of the water removed by evaporation. However, in these areas movement may occur by unsaturated flow, a phenomenon that is little understood. Such movement will be considerably slower than that in saturated media and, because of the lack of adequate information and slow movement by this mechanism, it will not be considered further. However, the following analyses, based upon the bottom of the pit being close to the top of the aquifer, bypasses the unsaturated flow question in a very conservative manner.

The types of materials intervening between the bottom of the waste pit and the aquifer will vary widely from site to site. In some areas, such as Hanford, the wastes are buried in sediments of a relatively uniform nature, although some stratification of permeability occurs in the layers of sediment. Between the pit and the water table, however, the water flow through soil-like materials. In other areas, the intervening materials are solid, with percolation occurring along fissures. The burial grounds at Los Alamos, in volcanic tuff, and at Oak Ridge, in shale, are of this nature, although the distance to the water table is vastly different in the two cases. In areas where the distance is large, particularly if the intervening sediments have a high exchange capacity, these

materials will delay any leachate from reaching the ground water for many hundreds or even thousands of years. However, this factor is not taken into account in the following analysis.

In order to arrive at an estimate of a generic limit we have used our simplified model of the burial ground, consisting of a total area of 1000 m x 1000 m, with the bottom of the waste pits at the top of the aquifer. The aquifer consists of a uniform bed of sediments 10 m thick and effectively infinite in extent in the horizontal dimensions. As a basis for the calculations we consider that the leachate is released uniformly to the water table over the area of the burial ground. This contaminant is, then, assumed to mix through the full depth of the aquifer. Such mixing, of course, will not occur until some distance downstream, but equivalent dilution will result when the water is pumped from the ground through a well perforated along the full depth of the aquifer. The movement of water in the aquifer is assumed to be at a rate of 30 cm/day perpendicular to one side of the burial grounds, a value reasonably representative of the slower movement at the ERDA sites. (See Appendix B.) Dispersion coefficients are not introduced and the water concentration is calculated at the downflow edge of the burial ground.

Other more realistic models derived from transport theory are available and could be used in this basic configuration. (See Appendix I.) However, the lack of adequate input data for most sites and the conservatism of the simpler model make it attractive for this generic calculation.

An important reaction in the interaction between a contaminant in water and the soils or sediments in the aquifer is that of ion exchange. This results in removing the contaminant from the solution until the surface sites on the soils are saturated at the resulting concentration. Thus, the contaminant moves through the ground at a slower rate than the water and also lowers the concentration at the front. The adsorption parameter is normally expressed as K_d , which is expressed as

$$K_d = \frac{\text{Quantity of contaminant in the soil/gram of soil}}{\text{Quantity of contaminant in water/cm}^3 \text{ of water}} \quad (3)$$

A distribution coefficient (K_d) is a measured empirical sorption coefficient used to express the sorption relationship of a radionuclide between the aqueous and solid phases of a soil suspension. It is usually expressed as the activity per gram of soil to the equilibrium solution activity per cm^3 of solution, thus, the units are cm^3/g . The purpose of defining K_d was to assign a numerical value to the sorptive characteristics of a soil for a particular radionuclide. Thus, K_d values have been used extensively in modeling the movement of radionuclides through soils and porous media. Unfortunately, K_d values between individual soils and radionuclides are not constant. In fact, they are highly variable depending on the pH, competing cations, and method of measurement. The following is not the result of an extensive search for K_d values for Pu and Am, but represents some data published from 1958 to 1977.

Prout⁷¹ measured the K_d of plutonium with a South Carolina subsoil containing approximately 20% clay and 80% sand. He noted the differences in K_d at various pH values and oxidation states. For Pu(III), K_d 's were greater than 10^3 at pH values between 4 and 11. However, K_d 's were measured as low as 10 for Pu(III) at a pH less than 2. For Pu(IV), the dominant oxidation state, the K_d 's were greater than 10^2 for pH values ranging from 1 to 11. At pH values greater than 5, the K_d 's were greater than 10^3 . For Pu(VI), the lowest K_d values were observed at pH values less than 7; these K_d 's were generally less than 10^2 . At pH's greater than 7, K_d 's increased to a maximum at pH 7.5 and decreased to approximately 200 at pH 10. Rogers⁶⁶ observed a similar pH relationship between ²³⁸Pu and soil. However, the maximum K_d of 10^5 was observed between pH 5 and 6 rather than between 7 and 8. Bondietti et al.⁷² observed K_d values of approximately 10^5 using Pu(IV) with a Miami silt loam, pH 6.5 containing 5mM of calcium in the aqueous phase. On montmorillonite clay, Pu(IV) had a K_d of 10^4 while Pu(VI) was lower (10^2). Approximately 200 hours were required for equilibrium of Pu-238 with three soils.⁷³ The K_d 's after 200 hr were on the order of 10^4 for neutral base-saturated soils from Washington and Illinois but about 400 for the acid base-saturated sandy soil from South Carolina. Francis,⁷⁴ using the same soil from South Carolina and Pu-237, found the K_d in water-soil suspension to range from 3 to 121 over a pH range of 3.3 to 7.8. The low K_d observed in this soil is due to the solubilization of soil organic matter at pH's greater than seven. In the presence of 0.01 M Ca, the K_d 's were greater than 10^3 at pH values between five and seven. One should appreciate the fact that K_d values cannot be indiscriminately assigned to soils, but rather

should be qualified as to environmental conditions, such as pH, base-saturation, soil-solution ratios, contact time, and oxidation states of plutonium.

There is a limited amount of data on the K_d of Am with soils. Probably some of the best work is that of Houtson et al.⁷⁵ For a base-saturated Washington subsoil, K_d values for Am were greater than 1200 and independent of sodium or calcium concentrations in the soil solution. However, the K_d 's for an acid South Carolina subsoil were quite low, ranged from 280 to 1, and were functions of sodium and calcium concentrations in the soil solutions. Generally speaking, for neutral base-saturated soils, K_d 's for plutonium and americium are usually greater than 10^3 ; however, for acid sandy soils the values can often be as low as 10, depending on the pH and soil solution composition. We will consider a value of 100 for all transuranics, as representative of a reasonable worst case at the pH's and composition of water expected in most aquifers.

The calculation of the ground water concentration can be simplified by noting that at equilibrium between the rate of addition of contaminant and movement downgradient, the concentration at the edge of the burial ground will be equal to that which would occur if the entire seepage occurred as a line source along the down-gradient edge of the burial ground. Thus, in our case, we assume an addition of 1 Ci/yr or 2.74×10^{-3} Ci/day as a line source along a strip 1000 m wide or 2.74×10^{-8} Ci/day per cm length of the source. When diluted in the full 10 m depth of the aquifer, this is 2.74×10^{-11} Ci/day per cm^2 front of the total aquifer. While the initial concentration in the water phase will be greatly reduced due to adsorption on the soil, continued addition of further increments will result in buildup on the soil until an equilibrium is reached between concentration left in the water and that on the soil. This condition will be reached when the concentration in the water is equal to the concentration in the added increment. In essence, the soil is now saturated for the exchange at this concentration and any movement of the contaminant from water to soil is exactly balanced by the reverse movement from soil to water. Thus, after a long time the concentration in the water will not depend on the value of K_d but only on the rate of addition of transuranic, in our reference case 2.74×10^{-3} Ci/day; the movement, 30 cm/day; and the porosity of the aquifer. The time required to build to equilibrium, however, will be greater for higher K_d 's and for lower values of the porosity.

At equilibrium, then, the concentration in the ground water will be equal to the quantity added per day per square centimeter of aquifer depth divided by

the volume of water passing through the square centimeter per day or the water velocity multiplied by the porosity. These concentrations are given for our model case in Table XXII.

At water velocities in the aquifer higher than the 30 cm/day chosen, the concentrations would be correspondingly reduced.

The movement of the contaminant front is also controlled by the adsorption on the aquifer medium with the front moving at a lower rate than the water in the aquifer. The velocity of the front, v , is given by:⁷¹

$$v = \frac{R}{(K_d \rho + c)}, \quad (4)$$

where R is the percolation rate, or cm^3 of water passing through each cm^2 per day, ρ is the soil density, and c is the soil porosity. Since the area porosity is equal to the volume porosity,⁷⁶ the velocity of the water, v_w , in the ground is given by R/c . The distance, x , traveled by the front in a given time, t , is then:

$$x = \frac{t v_w c}{K_d \rho + c}. \quad (5)$$

TABLE XXII

WATER CONCENTRATIONS IN MODEL CASE
1 Ci/day Addition

<u>Porosity</u>	<u>Concentration</u> (pCi/ml)
0.1	9
0.2	5
0.3	3

The influence of K_d and porosity on the movement is illustrated in the calculations presented in Table XXIII.

These results indicate that for K_d of 100 or greater the distance of travel in our model aquifer is modest even for very long times. Radioactive decay has not been factored into the travel given. Longer distances could be covered in areas where the flow of water is between impermeable layers or fractures. For example, at the Idaho burial ground the flow is through sediments between basalt layers with the highest permeability at the basalt interfaces.⁴ Even here, however, the concentration will be limited by ion exchange and the total flow of the aquifer under the burial ground.

The quantity penetrating to the water table represents that which is leached from the wastes and penetrates the formations underlying the burial pit to the water table. Data to estimate the rate of leaching from actual wastes are scarce. To obtain an approximation we will use the data from Maxey Flats.^{77,78} Here the wastes were buried in an impenetrable formation and the

TABLE XXIII
DISTANCE OF TRAVEL (mm) OF CONTAMINATION FRONT
WITH WATER VELOCITY OF 30 CM/DAY

Time Years	K_d				
	0	10	100	1 000	10 000
$c = 0.1$					
10	1	0.007	7×10^{-4}	7×10^{-5}	7×10^{-6}
100	10	0.07	7×10^{-3}	7×10^{-4}	7×10^{-5}
1 000	100	0.7	7×10^{-2}	7×10^{-3}	7×10^{-4}
10 000	1 000	7	0.7	7×10^{-2}	7×10^{-3}
$c = 0.3$					
10	1	0.02	2×10^{-3}	2×10^{-4}	2×10^{-5}
100	10	2	2×10^{-2}	2×10^{-3}	2×10^{-4}
1 000	100	20	0.2	2×10^{-2}	2×10^{-3}
10 000	1 000	200	2	0.2	2×10^{-2}

pits filled with water from the rain. The leachate from the waste pits was collected in tanks and analyzed. These leachates contained an average of 217 dis/min per cm³ of ²³⁸Pu and 10.2 dis/min per cm³ of ²³⁹Pu with maximum values of 3290 dis/min per cm³ and 163 dis/min per cm³ respectively. The inventory in the wastes in these pits can only be obtained approximately but is estimated as 810 g or 14 000 Ci of ²³⁸Pu and 24 kg or 1470 Ci of ²³⁹Pu. Over 10⁷ liters of leachate have been obtained since pumping started in about 1972 to remove water that had infiltrated the trench. This occurred over a period of about three years although the initial pumping undoubtedly removed leachate that had accumulated in the trenches prior to this time. From the average values, about 1 Ci of ²³⁸Pu and 0.05 Ci of ²³⁹Pu were accumulated in the leachate or 0.007% of the ²³⁸Pu inventory and 0.003% of the ²³⁹Pu inventory. Taking into account the possibility of a longer leach period than the pumping period, this would indicate a leach rate of 10⁻⁶ to 10⁻⁵ per year although the value is uncertain. This leach rate also represents the effect of a longer term of contact between the wastes and the water than would occur in the passing through of infiltrating water and, as such, would represent a high value. We will accept a value of 10⁻⁶ per year on the basis that the Maxey Flats conditions allowed much more leaching than should occur if proper drainage is provided.

There is no real reason to differentiate between the leaching of plutonium and the leaching of americium in such a situation, because the leaching undoubtedly depends more upon the stray constituents in the waste, such as complexing agents and oxidants, than upon the strength of the adsorption of the transuranics on the waste material. We will, therefore, use the same value for each.

The potentially contaminated water is considered to be used for two purposes: domestic water and irrigation. This requires drilling of a well or wells in the area where the water is contaminated and pumping of the water to the surface. The probability of such an occurrence must be considered as low because the area involved will be small. In a water table flowing in sediments, for example, Table XXIII indicates that the area will be up to several kilometers in the direction of flow and one kilometer wide after 10 000 years. However, the possibility of movement to a point of use cannot be excluded.

b. Domestic Use

The primary use of the domestic water that is of importance to transuranium

element uptake is for drinking. In this case we will include ^{238}Pu although it has decayed to only 20% of its original activity in the 200 year control period. The probability of exposure to this isotope is even smaller than for the others because the area of contaminated water will be limited by the radioactive decay in the years following the control period.

In Table XXII we established water concentrations for different porosities of soil for the addition of 1 Ci/day to the aquifer. The allowable concentration is obtained by dividing the limiting intake from Table XI by the daily intake of fluids, $1650 \text{ cm}^3/\text{day}$. The ratio of the allowable concentration to the concentration resulting from a release of 1 Ci/yr provides the allowable release to the aquifer. The total quantity in the burial ground is then obtained from the leach rate of $10^{-6}/\text{yr}$. These calculations are given in Table XXIV.

a. Irrigation

For use of the water for irrigation, we will consider the application rate to the soil and the erosion rate to be the same as was discussed in the pathway

TABLE XXIV

ALLOWABLE QUANTITIES IN THE BURIAL GROUND FOR DOMESTIC USE OF THE AQUIFER

Nuclide	Allowable ^a Conc. (pCi/ml)	Allowable Release to Aquifer ^b (Ci/yr)	Allowable Quantity in Burial Ground (Ci)
^{238}Pu	0.56	0.052	260 000 ^c
$^{239,240}\text{Pu}$	0.35	0.039	39 000
^{241}Am	0.073	0.0081	8 100

^aFrom Table XI and intake of 1650 ml/day .

^bFrom Table XXII and column 2 using a porosity of 0.1.

^cCorrected by a factor of five for decay during the control period.

of erosion to the stream, resulting in an erosion elimination rate from the soils of 0.003 yr^{-1} . Additions, for the 60 cm of water per year and 1 Ci/yr release from the burial ground would be 60×9 (Table XXII for a porosity of 0.1) or 540 pCi per cm^2 to result in 12 pCi/g per year added to the 45 g of soil beneath a 1 cm^2 area to the mixing depth of 30 cm. It is assumed here that the nuclide in solution in the soil will adsorb rapidly and will be mixed by agricultural practices through the soil. The calculation of the allowable quantity in the burial ground is given in Table XIV.

d. Movement from Aquifer to Stream

In some cases an aquifer could flow into surface streams that are close enough to the disposal site so that the contamination front will reach the stream before radioactive decay reduces the concentration to insignificant levels. While there may be retardation of the flow due to adsorption on soils, when the front does reach the stream, the input to the stream will be the same as the input to the aquifer if allowance is made for radioactive decay. This assumes that the entire aquifer enters the stream.

TABLE XIV

ALLOWABLE QUANTITIES OF TRANSURANUM ELEMENTS BASED UPON TRANSFER TO AQUIFER WITH WATER USED FOR IRRIGATION

Nuclide	Allowable ^a Limit For Soil (pCi/g)	Equilibrium Quantity In Soil (pCi/g)	Allowable Release To Aquifer (Ci/yr)	Allowable Quantity In Burial Ground (Ci)
^{238}Pu	350	1 100	0.32	1 600 000 ^b
$^{239,240}\text{Pu}$	200	4 000	0.05	50 000
^{241}Am	14	2 600	0.0054	5 400

^aFrom Table XV.

^bCorrected by factor of five for decay during control period.

Once the contamination enters the stream, it will be adsorbed on sediments with behavior similar to that described for transport during erosion. However, since we do not have a defined distance from the burial ground to the aquifer for this case, it is more difficult to assess any decay corrections. From Table XXIII it would appear that any stream further than 0.5 to 2 km from the burial ground would receive no contaminant for about 10 000 years. However, this table was derived for hypothetical conditions that do not take into account the possibility of non-homogeneities causing channeling of the flow and a faster movement of the water in the water table. We will assume, therefore, that the quantities entering in the sediment study apply with only the decay correction for ^{238}Pu occasioned by the 200 year control period. The quantities released to the stream as calculated in the erosion model and the corresponding average quantities in the burial ground are given in Table XXVI.

TABLE XXVI
ALLOWABLE QUANTITIES IN BURIAL GROUND BASED ON AQUIFER EMPTYING
INTO SURFACE STREAM

Nuclide	Pathway					
	Drinking		Irrigation		Fish	
	Allowable ^a Release (Ci/y)	Allowable ^d Quantity (Ci)	Allowable ^b Release (Ci/y)	Allowable ^d Quantity (Ci)	Allowable ^c Release (Ci/yr)	Allowable ^d Quantity (Ci)
^{238}Pu	5 700 ^e	2.9×10^{10g}	380 ^f	1.9×10^{9g}	3700 ^e	1.9×10^{10g}
$^{239,240}\text{Pu}$	3 500	3.5×10^9	220	2.2×10^8	2300	2.3×10^9
^{241}Am	75	7.5×10^7	24	2.4×10^7	5	5×10^6

^aFrom Section VB1c - Release to Stream.

^bFrom Section VB1d - Release to Stream.

^cFrom Section VB1e - Release to Stream.

^dIn burial Ground.

^eEstimated by ratio of ingestion limit for ^{238}Pu to ingestion limit for $^{239,240}\text{Pu}$.

^fEstimated by ratio of soil limit for ^{238}Pu to soil limit for $^{239,240}\text{Pu}$.

^gIncreased by factor of five to account for decay during control period.

a. Conversion to Average Concentration in Burial Ground

All of the limits derived for the leaching to the aquifer depend upon the total quantity of the transuranium element present in the burial ground. For comparison with the other limitations based upon concentration and for operational use in deciding which batch of waste should not go to the burial ground, we will convert these total quantities to average concentrations in the burial ground by using the volume in our model burial ground.

The model burial ground has an area of 1000×1000 m, or 10^6 m², with burial in pits 4 m deep. This leads to a volume of 4×10^6 m³. Assuming an overall bulk density of the mass of 1.5 g/cm³, this provides an overall weight of 6×10^{12} g for dilution. Averaging over this volume is appropriate because small volumes of higher concentrations will not perturb the overall pattern significantly.

Results of averaging the previously estimated allowable quantities in the burial ground over this volume are given in Table XXVII.

TABLE XXVII
ALLOWABLE AVERAGE CONCENTRATION IN BURIAL GROUNDS
FROM MOVEMENT TO AQUIFER

Pathway	²³⁸ Pu		^{239,240} Pu		²⁴¹ Am	
	Total Allowed (Ci)	Average Conc. (nCi/g)	Total Allowed (Ci)	Average Conc. (nCi/g)	Total Allowed (Ci)	Average Conc. (nCi/g)
Aquifer						
Drinking ^a	260 000	43	39 000	6.5	8 100	1.4
Irrigation ^b	600 000	270	50 000	8.3	5 400	0.9
Stream						
Drinking ^c	2.9×10^{10}	4.8×10^6	3.5×10^9	5.8×10^5	7.5×10^7	1.3×10^4
Irrigation ^c	1.9×10^9	3.2×10^5	2.2×10^8	3.7×10^4	2.4×10^7	4×10^3
Fish ^c	1.9×10^{10}	3.2×10^6	2.3×10^9	3.8×10^5	5×10^6	830

^aFrom Table XXIV.

^bFrom Table XXV.

^cFrom Table XXVI.

2. Seeps

The experience at Maxey Flats⁷⁷, as well as at other locations, has indicated that burial of the wastes in a relatively impermeable formation in an area with high rainfall can lead to accumulation of water in the waste pits with consequent leaching of the transuranics into this water. If, then, the formation contains cracks through which this water can flow to a lower elevation, seeps and springs of the contaminated water can occur. A similar phenomenon could occur in soils or sediments if the liquids moving downward out of the waste pit encounter a layer of impermeable materials resulting in a perched water table. If the impermeable layer intersects the ground surface at a lower elevation, a spring or seep could be formed. As the ion exchange sites, particularly in flow-through fissures, become saturated, the concentration of transuranics will become essentially equal to that in the leachate.

The water supply in such a case is limited by the area of the burial ground and the rainfall. In Maxey Flats⁷⁷ the 10^7 liters pumped in three years represent about the amount from a household faucet running one-third of the time. This quantity would doubtless occur in multiple springs or seeps so that any one would be a small fraction of the total. It, therefore, appears doubtful that this water would be used extensively by any population group for domestic or irrigation purposes.

The primary potential problem would appear to be with an occasional individual drinking the water or livestock grazing in the region or drinking the water. It is noted, however, that there will undoubtedly be many other chemicals present in the water, at least in the early phases of leaching, and these may well affect the potability.

We have not pursued this pathway further since it is felt that such conditions of waste burial in wet areas should not be permitted.

3. Intrusion into Waste Mass

We have considered two types of intrusion possible. The first, described in this scenario, is the actual intrusion into the waste mass by digging into the area either accidentally or on purpose. The second, to be described in the next scenario, is use of the area of the burial grounds at some time distance in the future, when erosion has removed the cover, for living or farming. The first form of intrusion will be described in the following scenario.

The question of intrusion, particularly people digging into the area, has raised some heated debate in the Task Group meetings. On the one hand, the feeling has been strongly expressed that our obligation is to provide warning that hazardous material exists under the ground. Anyone digging, who does not heed this warning, is responsible for any ill-effects that may result. Notification would be by above-ground monuments clearly marked to indicate danger or by any other scheme deemed feasible, such as covering the wastes with red concrete. Inclusion of restrictions on land use and the reasons for these restrictions in any legal description of the property would also assist because such documents usually have a life longer than governments. In essence, this approach would say that we would not include this intrusion pathway because the responsibility should be placed upon the individual who is inclined to intrude into the mass.

On the other hand, there was the feeling that one cannot assure that such markers would last the lifetime of some of the nuclides, particularly with man's known penchant for vandalism. In addition, if the burial of solid objects of interest, such as machines or laboratory equipment, continues, the burial grounds could be construed as an "attractive nuisance" because there will always be someone to take the risk to gain an object he desires. Thus, this reasoning would say that we should consider intrusion as a possibility. This approach essentially states that our obligation is not to bury "body traps" even if we warn that they are there.

Since the differences between these approaches could not be resolved, it was decided to include intrusion into the waste mass in the calculations with, however, the proviso that those more acquainted with legal and sociological principles should examine this question and provide a reasonable policy answer. Thus, we will examine the case to determine its importance relative to others and provide recommendations so that either course can be chosen by policy makers.

For the intrusion case, one must consider the motive for the intrusion as well as the method. One type of intrusion would be accidental, such as unknowingly drilling a well through the mass or digging a foundation for a house. A different type of intrusion would be the individual who finds a useful or interesting artifact and proceeds to excavate the entire area, retrieving items and, perhaps, distributing them to the general public. We will consider the accidental intruder in a separate category from the scavenger because of the

different methods of potential exposure and the potentially larger number of people exposed.

The waste matrices consist of almost any conceivable material, ranging from process room trash, through construction items, to various types of equipment. They vary widely, not only in material but also in size and ratio of surface to volume. The latter factor has raised questions as to the usefulness of activity per unit weight, or volume, as a basis for a limit, since an item made of high density material with a low surface to volume ratio could have extremely high surface activity and still meet the concentration limit. We will treat the two categories of low surface to volume and high surface to volume ratio separately.

The use of plastic bags to control the contamination during handling and emplacement of the wastes also raises a question as to the degree of preservation that the plastic containment will provide for the contents. Horton,⁷⁹ at Savannah River, reports on the exhumation of wastes after burial for 14 years. The wastes encased in plastic were well preserved with, for example, the writing on a piece of paper still clearly legible through the bag. He concluded that such items would be preserved "indefinitely." The Environmental Protection Agency reports on a study of the biodegradability of packaging plastics by the Union Carbide Corporation.⁸⁰ The results of this study verify the common belief that the current high-volume, high molecular weight, packaging plastics are not biodegradable at practical rates. The only synthetic, high-molecular-weight polymers found to be biodegradable were the aliphatic polyesters and their derivatives, with structural modification of polyethylene and polystyrene by random copolymerization with other monomers not leading to biodegradability. While these were relatively short-term studies, the techniques used, growth of test microorganisms on plaques, should indicate the utilization of the carbon in the plastics to provide degradation. If the growth does not occur, then presumably, the plastic could be stable, in the absence of sunlight, for long periods of time. It was therefore, concluded that items contained in the present non-degradable plastics may be preserved for relatively long times. This is not to say that all such items will remain since some, and possibly many, will be broken during the burial operation and others will be breached by rodents or other burrowing animals. However, if the possibility exists of some of the packages with the higher waste levels remaining intact, then concern must be felt for the individual who may recover the package and consider the contents as valuable artifacts. For this reason,

we feel that wastes contained in non-degradable plastic should be limited in transuranic content to a level lower than the waste which will degrade in some reasonable time.

We fully realize the importance of the plastic containment for wastes in protecting the workers who transport and bury the waste as well as the reduction that the use of these materials makes in release to the environs during such operations. However, we cannot ignore the potential problem that could exist in the future if curious archeologists or scavengers were to regard these materials as relics of our civilization or as curios.

a. Low Surface to Volume Ratio - Degradable or Contamination Incorporated

These types of waste will consist of papers, cleanup rags, or other biodegradable materials as well as wastes with the transuranium elements incorporated in the matrix (not on the surface), such as sludges from waste treatment. The degradation rate of the biodegradable waste is expected to vary from site to site depending upon the moisture available to the wastes and the oxygen content of the soils at the burial depth. However, for these estimates, we will assume that the waste, at the time of intrusion, consists of a soil-like mixture with the contamination distributed through the material. "Hot Spots" will still be present because of the non-uniform contamination levels in the original wastes, but these will be averaged if any extensive excavation is done.

A number of different scenarios for intrusion into the waste mass are possible, ranging from drilling or digging a well, through digging foundations for houses, to extensive excavation in the waste mass for artifacts (see next section). It has been pointed out that most of these activities appear to require some form of official sanction if it is assumed that some form of government exists.⁸¹ Presumably records would exist as to the existence of the wastes and the work would be stopped. However, there have been structures built on old landfills in the U.S. in the recent past with methane explosions resulting in death and injury.⁸² The present situation in Niagara Falls, with a number of houses built on an old chemical waste dump, illustrates also the present inability of government to control such situations. It must be admitted that radioactive waste burial grounds are better delineated and controlled than has been the practice with other hazardous wastes in the past, so that some reason exists to believe that governmental controls will be useful, at least over the first millennium and, perhaps, even longer.

It would appear that the amateur artifact digger could be the maximum case of intrusion because he does not consider possible hazards of the waste and would not take any precautions. One cannot provide precise calculations of the conditions and intakes because the methods of digging and handling the soils, the pattern of digging and the time worked in the area are unknown. For this estimate we will assume that an individual spends one-half of his week-ends for ten years working in the area and his activities produce dust concentrations averaged over all of the working hours of 1 mg/m^3 .

Anspaugh and Phelps³⁷ have examined the data on pneumoconiosis in mines and the British data that indicate that dust levels in excess of 1 mg/m^3 would result in a large public health problem. While the time of exposure of our hypothetical artifact hunter is limited, Anspaugh's data would indicate the 1 mg/m^3 to be a reasonable upper bound. Such an exposure would result in inhalation of 3.25 g of dust over the 10 year period or an average of 0.00089 g/day. The dose rates from the inhalation of 1 pCi/day at the end of a 10 year period of inhalation, were calculated using the constants from Table VI, again assuming one μm particles with plutonium to be class Y and americium class W. The plutonium dose rates to the bone were increased by 20% to allow for the plutonium remaining in the lung that will transfer to the bone. From these calculations, the allowable "average" concentration in the average waste was estimated. These values are given in Table XIVIII.

This scenario represents a situation where the individual receives a body burden in a relatively short time, with the bone dose rates continuing through the remainder of his life-time. For the plutonium isotopes, the short time of accumulation means that the lung is limiting and the bone dose rate is about $1/4$ to $1/5$ th of the limit. For ^{241}Am , however, the use of Class W means that the bone is limiting and the dose rate of 500 $\mu\text{rem/yr}$ will continue essentially throughout the rest of the lifetime. One could use the dose commitment concept and allow only exposure each year that would provide a dose over the remainder of the life-time equal to 0.5 rem/yr . However, the dose commitment concept has an anomaly in that the 0.5 rem/yr is allowed every year for short-lived nuclides or external radiation, while with nuclides such as ^{239}Pu it results in low dose rates at early times finally building to 0.5 rem/yr at the end of life. This results in a life-time dose for the short-lived nuclides of about twice that of ^{239}Pu . While one can argue that in occupational exposure the probability of continued repetition of the dose year after year is low, the same cannot be said

TABLE XVIII

SOIL CONCENTRATIONS TO LIMIT ORGAN DOSE
OF ARTIFACT HUNTER

Nuclide	Organ Dose Rates for 1 pCi/day inhaled		Average Soil Limits	
	Lung (rem/yr)	Bone (rem/yr)	Lung pCi/g	Bone pCi/g
^{238}Pu	0.19	0.050	15 000	65 000
$^{239,240}\text{Pu}$	0.19	0.049	3 030	14 000
^{241}Am	0.019	0.17	30 000	3 200

for environmental exposure where the individual has no control over the source. We, therefore, believe that the application of the criterion of 0.5 rem/yr used here is appropriate.

b. High Surface to Volume Ratio Items

Deliberate scavenging for articles of possible use or interest is one possibility that poses great difficulty in analysis, since such objects can be widely distributed and used in a variety of ways that could result in exposure to many different people. The potential problem will be dependent upon the time of intrusion following burial and the corrosion stability of the material of which the object is made in the burial environment. If the materials are degradable and the conditions in burial are such that complete corrosion or biodegradation occurs, then the contamination will be dispersed throughout the medium and the previous analysis will apply.

However, under many conditions, such corrosion may take long periods of time, particularly for resistant materials, and it must be assumed that such items may be exhumed and put to use in a variety of ways. It is concluded from this, that limits should be applied to such items so that later use will not result in an unacceptable hazard.

As a definition of the type of items that may be of concern, we would consider those that may interest people, either for the use for which they were designed or for the many other applications that people can dream up for such items. For example, an old glove box may become a planter for flowers and all

sorts of scrap metals seem to have a second life as part of an artistic endeavor. Small tools have an intrinsic value of their own, and many pieces of discarded equipment show up in home workshops or even in school shops.

The quantitative hazard from use of such materials is extremely difficult to define because of the different uses that objects will undergo and the lack of adequate data on transfers from the object to man. The problem is similar to that of providing limits for contamination on objects to be disposed of to the public from a nuclear plant.

Limits for surface contamination on objects to be released to the public have been derived by the American National Standards Institute (ANSI).⁸³ These values are based upon experience rather than analysis of hazard so that it is not possible to associate a dose value with them. In fact, present methodology for providing such estimates is very inexact, particularly in accounting for doses resulting from transfer of contamination from the object to other places where doses can result.

The ANSI limit for plutonium (or other transuranics) is 100 dis/min per 100 cm², which corresponds to an NCRP value used earlier.⁸⁴ Other values have been recommended by individuals using various approaches. Dunster,⁸⁵ for "inactive" areas of a plant, recommends a value of 2000 dis/min per 100 cm². An "inactive area" is one where radioactive materials are not handled and non-radiation workers are present. He also indicates that averaging can be done over 300 cm² for inanimate surfaces in general and 1000 cm² for surfaces such as floors or walls. Blatz and Eisenbud⁸⁶ studied the workers in radium plants and concluded that a reasonable limit for this nuclide was 10 000 dis/min per 100 cm². When the greater solubility of radium as compared to plutonium is considered, this would appear to be a conservative value for plutonium. This limit did, however, apply to workers. They also included a review of current contamination limits used throughout the world and concluded that they had safety factors of 3 to 30.

We have looked at other methods of deriving the limiting surface contamination. If we assume that our artifact seeker spends as much time working with the artifact as he did finding them, then, over a ten year period, he will spend 2600 hours. The total quantity inhaled to limit doses to our criterion of 500 mrem/yr can be estimated from Table XXIX by multiplying the soil concentration by the number of hours, the amount inhaled per hour (1.25 m³) and the assumed average air concentration of 1 µg/m³. This gives 9750 pCi for the plutonium isotopes or 10 400 pCi for americium. Thus, the allowable

concentration is 3 pCi/m^3 for plutonium or 3.2 pCi/m^3 for americium during the hours of working on the artifact. We will further assume that he does this work inside in a room with two air changes per hour and a volume of 500 m^3 . Healy⁸⁷ has pointed out that, in an enclosed area, the concentrations are probably more a function of the total contamination present rather than the amount on any single area and has presented an equation to estimate the concentration:

$$x = \frac{f q A}{V n}, \quad (6)$$

where f is the fractional resuspension rate (hr^{-1}), A is the area of the contaminated area (m^2), q is the contamination per m^2 on the contaminated area (pCi/m^2) V is the volume of the room (m^3) and n is the number of air change per hour. This equation assumes instantaneous mixing of the contaminant with the air of the room.

The value of f has been examined by Healy⁸⁷ who concludes that a value of $5 \times 10^{-3} \text{ hr}^{-1}$ is appropriate for vigorous activity. We will, therefore, assume that the artifact seeker works on items with a vigor to produce this rate of resuspension with one m^2 effective at any one time. This resuspension rate will be the average over the full time of working and will include periods when the contamination has been removed as well as periods when work has started when the resuspension may be somewhat higher.

The limiting concentration, then, can be obtained by substituting the allowable concentrations in equation (7). This provides limits of 600 000 pCi/m^2 or 60 pCi/cm^2 . For ^{238}Pu , the decay during the control period of 200 years would permit increasing the limit to 300 pCi/cm^2 .

The items that are postulated to be recovered from the burial ground will have been in contact with the soil for periods of hundreds and, perhaps, thousands of years. For most, if not all, major corrosion of the surface will occur in this period, releasing some of the contamination to the soil. Even for the non-corrodible materials, some surface reactions will occur that will result in loss of contaminant. The numbers of items recovered, their distribution in the public, and their possible uses are unknown. However, it is anticipated that few will go to the same area so that exposure will not be for extended time periods comparable to the life-span or working life of an individual.

The situation where an object is uniformly contaminated to a limit is not the usual one. Rather, the contamination will be spotty with the limit reached in one area. Thus, for objects with large surface area, the total contamination will be less than the product of the area and the limit. For these reasons we will arbitrarily recommend a limit of 60 pCi/cm^2 averaged over areas of 1000 cm^2 .

This value is not as conservative as the one applied to day to day operations, but when the relatively low probability of intrusion and the time for corrosion or other surface changes is considered, it probably represents a risk lower than calculated for the uniform mixture, although this cannot be proven for reasons given above.

Ideally, in defining the wastes to which either limit applies, one should be able to relate the surface contamination value to the mass concentration values with a point in the comparative curve indicating where each limit should apply. Here, however, a problem arises since the surface contamination is more limiting than the mass concentration down to small thicknesses of material. To illustrate this problem we have calculated the surface concentration from geometrical shapes for given values of the mass concentration. These values are given in Table XXIX.

As can be seen, any limit of 60 pCi/cm^2 will be violated for thickness of 0.01 cm or greater for low density materials unless the mass limit is set much lower than the present 10 nCi/g . At 1 nCi/g , for example, the surface limit would be met reasonably well for low density materials, and with some margin, if the limiting thickness were set at 0.1 cm or 1 mm .

In lieu of attempting to derive an interchange point, in view of the uncertainties in the surface level and/or the required low mass concentration for other wastes, we have concluded that the separation should be based upon type of waste. The general trash from the operations (combustibles), the waste treatment sludges, incinerator ash, and other waste types where the contamination is related to the general volume and not the surface should be limited on a mass basis. Equipment, sheets of metal, construction materials, and similar items where the contamination is chiefly on the surface will be controlled as based on the surface contamination.

a. Wastes in Non-Degradable Containers

The rationale for considering wastes in plastic wrapping as potential

TABLE XXIX
SURFACE CONTAMINATION (pCi/cm²)
FOR VARIOUS VALUES OF MASS CONCENTRATION

Radius or Thickness (cm)	density 1 g/cm ³			density 5 g/cm ³		
	Sphere	Cylinder ^a	Plate ^a	Sphere	Cylinder ^a	Plate ^a
Mass concentration = 10 nCi/g						
0.01	30	50	100	6	10	20
0.1	300	500	1 000	60	100	200
1.3	3 000	5 000	10 000	600	1 000	2 000
Mass concentration = 100 nCi/g						
0.01	300	500	1 000	60	100	200
0.1	3 000	5 000	10 000	600	1 000	2 000
1.	30 000	50 000	100 000	6 000	10 000	20 000

^aEnd or edge areas not included.

artifacts was given earlier. The evidence of survival of these materials is not good but the evidence that they will not survive is also poor. A way to avoid such a problem is to develop containment materials that are adequate for the handling procedures but which are designed to degrade in the burial ground. This indicates that the goal of providing improved packages to slow the migration of transuranics from the burial ground may be undesirable. The analyses that we have done indicate that the problem of movement from the area by leaching or erosion is much lower than the problem of artifacts remaining. This has led to the conclusion that degradable packages are of lower potential hazard than packages designed for long-time containment.

An obvious question is why such wastes should be considered as artifacts. In general, the items they contain are worth little by today's standards. There is, of course, no assurance that they will not be simply recovered and reburied. However, the trend in our civilization appears to be one of appreciating old and unique things, not for their inherent value, but simply because they reflect the past. Thus the collection of old beer cans, invoices, or documents of any sort is a popular pastime. We cannot assure that certain of the items buried will

not eventually be similarly prized. For, example, in a package of waste there could be notes concerning the work or even personal matters. Even if there is not, the thought that such items may be there could be adequate stimulus for exploration.

We have not attempted to estimate a separate exposure limit for handling of such materials because of the unknown, and unknowable, use that might occur and the air concentration that could result. Instead, we have applied the limit derived previously for intrusion into the degraded material except that dilution factors from uncontaminated wastes and the soil in the burial ground will not apply.

4. Exposure of Wastes

A postulated pathway to man is the eventual uncovering of the wastes with people then living in the area, farming in the area or carrying out other activities. The burial areas will be large so that room will be available for such uses. Once again, the question of marking the area and depending upon the future race to enforce these restrictions occurs. Here we are concerned with time scales of thousands of years so that the probability of monuments or other markers enduring is not as great as for our intrusion case. We will, however, approach this limit on the same basis as the intrusion limit. That is, the information will be presented with its acceptance subject to opinions of those skilled in legal or sociological fields.

One method for removing the cover and leaving the wastes exposed has been discussed in the erosion pathway. Here, the surface cover is removed in 2000 to 13 000 years leaving the exposed wastes over the next period of 4000 to 25 000 years. There are other ways that might be considered. Floods that greatly enhance erosion could not only uncover the wastes but also spread them out over a larger area, although with significant dilution. Glaciers could accomplish a similar scouring of the cover, although it appears more likely that the effect here would be to scoop out the entire waste area and deposit it over a larger area. In the shorter term, the wastes could be unearthed in limited areas by people digging a trench or foundations for buildings.

The probability of the eventual release and uncovering of the wastes by erosion or other occurrence is high at most of the DOE sites since they are in areas of net erosion, not buildup. It is difficult to estimate the probability of any unearthing by man because of changes in population and distribution of

population that will occur. Most DOE sites are located in areas that were isolated until 1940-1950 because they were not considered as prime sites for other uses under the population patterns then prevalent. Whether this would continue in the future in the same pattern would probably depend on any climatic changes or drastic population shifts. Thus, we cannot state that the probability of this pathway occurring is high; neither can we say that it is insignificant.

The consequence of such unearthing could be to produce an area with soil concentrations approaching that of the waste mass, depending upon the amount of dilution from outside soil that occurs. The area could range from a limited one around a house, where the excavated material is spread out in the vicinity, to the entire burial area, or a larger area than the burial area, although at lower concentration, from spread down-gradient by erosion.

In the worst case of no dilution of the resulting waste mass and assuming that people live on the area, the soil limits derived previously would apply to the average mass. However, in the majority of the scenarios leading to exposure of the waste, major dilution of the material would be encountered. Even in the erosion case, mixing with uncontaminated sediments from the remainder of the watershed could occur. We will, therefore, consider an arbitrary dilution factor of two by uncontaminated soils to be applicable in estimating the final concentration.

The ^{241}Pu will have disappeared and the ^{241}Am will have decayed to 0.04 of its original value in the 2000 years required to uncover the wastes in our erosion scenario. However, the possibility of moving the wastes to the surface early by human action exists, although there will be dilution and limitation of areas disturbed. As a conservative measure appropriate to the unknown future we will consider only one half-life for ^{241}Am and five half-lives for ^{238}Pu in deriving the limits. These limits are given in Table XIX.

5. Other Pathways

a. Uptake by Vegetation

A potential chronic release pathway is the uptake of transuranium elements by plant roots, and transport to the surface where the transuranics will accumulate in the litter and humus over the years. The discrimination of plants against most transuranium elements means that this will be a slow process but

TABLE XXX
LIMITS FOR AVERAGE CONCENTRATION
BASED UPON EXPOSURE OF WASTES

<u>Nuclide</u>	<u>Limit</u> (pCi/g)
^{238}Pu	22 000
$^{239,240}\text{Pu}$	400
^{241}Am	60

the continued "pumping" to the surface could, conceivably, be of importance over long times.

A biomass model, using the parameters of the ecosystem that follow the generation and transfer of biomass, and incorporating the uptake of plutonium, is under development at the Los Alamos Scientific Laboratory.^{88,89} The model assumes that 0.2% of the root mass of a mature tree is below 1.5 m with the uptake linearly proportional to this fraction. The uptake coefficient is defined as the fraction of the radioactivity removed from soil due to respiration of the soil water by the leaf biomass. A value of $4 \times 10^{-9} (\text{ml/m}^2)^{-1} (\text{g(leaf)/g(soil)})^{-1}$ was determined from experiments by Romney et al.⁹⁰ with clover over a period of five years. Once again, the uptake is linear with this coefficient.

An evaluation of the uptake was made for a pinon-juniper region typical of Northern New Mexico assuming wastes containing 10 nCi/g were buried 1.5 m below the surface. After 5000 years, the simulated soil concentration resulting from transfer of plutonium to the surface was about 8 fCi/g.⁸⁸ Thus, the results indicate that the net result of transfer over this period was to add an amount of plutonium about equal to that present from natural fallout. This run did not include erosion of the surface so that a higher result could be predicted if it is assumed that erosion reduces the cover and a higher fraction of the roots reach the waste mass. Results with other types of vegetative cover could be somewhat higher but not significantly, within the errors of such a prediction.⁹¹

From this, we conclude that no significant problem exists with plutonium. If the plant uptake of americium is assumed to be higher than that for plutonium, the transfer rate would be higher, however, the relatively short-half life of

²⁴¹As as compared to ²³⁹Pu would limit the time for transfer to the surface. It is also noted, that this mechanism is unlikely to produce surface concentrations exceeding the original waste concentrations so that the limits covered under the pathway of exposure of wastes will limit exposures from plant movement to the surface.

b. Burrowing Animals

Another mechanism for bringing waste to the surface (or introducing water into the waste in normally arid regions) is the action of burrowing animals or insects. The depths of burrows or tunnels for several such animals are given in Table XXXI.^{92,93}

As can be seen, the probability of other than ants reaching the wastes with the two meter cover is low. As the cover erodes, one can expect transfer of the waste material by this mechanism. However, once again, the surface concentrations will be lower than the wastes (initially) and the doses will be controlled by the pathway of people living on the area after the wastes are exposed by erosion.

TABLE XXXI
BURROW DEPTHS^a

<u>Species</u>	<u>Maximum Typical Burrow and Tunnel Depth (m)</u>
Harvester ant	3
Moles	1.2
Pocket gopher	0.6
Pocket mouse	1.6
Deer mouse	0.6
Field mouse	0.6
Earthworms	0.5

^aCompiled from References 92 and 93.

2. Severe Events

One general concern at facilities handling hazardous materials is the possibility of disruption of protective barriers by a severe act of nature, or accident, well beyond the design parameters of the facility, with a postulated subsequent release of the hazardous materials to the environs. The potential problems at shallow earth burial sites are limited because of the natural stability of the earth's surface against many of these forces.

Severe winds such as hurricanes or tornadoes passing over the emplaced waste will not disturb the material below the surface. In the long term, after the burial ground surface has eroded to expose the wastes, there will be some wind resuspension and dispersal. This, however, is one of the pathways considered in deriving the limits. It is to be noted that winds will be of importance during the operational phase when wastes are exposed before burial and precautions against dispersal at this time must be taken.

Earthquakes should produce little vertical displacements or gross movement of the emplaced materials unless the burial ground is on a fault. For this reason, care should be taken to assure that a fault does not run through the area of burial. Volcanic action may be a remote possibility in some areas. However, the probability of a fissure developing in the burial ground is low and considerable dilution and fixation of the transuranics would result if it did occur. Volcanic action at a nearby location would, if it affected the burial ground at all, result in further surface cover through lava flows or ash fallout.

Falling objects must be massive to penetrate the surface cover. The probability of an aircraft accident at a burial ground penetrating to the waste is low. A sufficiently large meteor could penetrate the surface cover. At Los Alamos it was estimated that to penetrate a minimum of one meter depth, the meteor would have to weigh at least 0.6 kg, and the probability of striking pits with an area of 0.1 km^2 is estimated at about 10^{-7} per year.⁸⁹ Thus, the probability of striking our 1 km^2 burial ground is 10^{-6} /yr. Such an accident, if it occurred, would expose only a small portion of the wastes.

In the event of a new glacial advance, a reasonable possibility within the next 10 000 to 100 000 years, the wastes could be exposed either by glacial action or enhanced erosion from the melting snow and ice. At such a time, the transuranics could be considered to be only a minor problem compared to those resulting from the ice age itself. In addition, the movement and mixing of

soils would tend to dilute the transuranic by mixing them with large volumes of sediment. Such a condition would not occur for many years and is one of the imponderables addressed earlier.

Floods could pose a serious threat by eroding the surface cover and transporting all or a portion of the material downstream. An alternative pathway is to provide water for seepage through the wastes in a normally dry area thereby contaminating the ground water. Floods could occur and cause movement of the material within the assumed time of governmental control. Thus a necessary criterion in the location of a repository is natural protection against flooding. We will assume in the following that such a criterion is used.

C. Deep Earth Burial

While the Task Group was commissioned to provide limits for shallow earth burial, it became apparent early in the study that many of the most significant pathways could be eliminated if the wastes were placed deep enough that such forces as erosion, intrusion by man, plant uptake, burrowing animals or falling objects could not uncover the wastes within the time that the concentrations were significant. The original concept was the use of abandoned dry mines or caves at a depth sufficient to provide reasonable assurance that these forces would not penetrate, but not deep enough that water would penetrate to the wastes. A cavern dug into the side of a stable hill or mountain would also suffice. This has been extended to the concept of simply digging deeper pits. This will limit the places where "deep" burial can be used to areas with sufficient depth to the water table so that the wastes are not emplaced in an aquifer. Safety problems also result from such deep excavations and the cost may be higher than is warranted.

If these pathways are eliminated, then the primary concern is transport in the aquifer, with the water used for drinking or irrigation or adding contamination to a surface stream. Limits for the average concentration for this case have been considered for shallow earth burial and are given in Table XIVII.

We have not attempted a firm definition of deep burial because it will depend upon the specifics of the site. One would feel that a depth of about 10 m (30 ft) would be sufficient to discourage all but the most determined intrusion and would essentially eliminate the falling object, except for a very

large meteor which would be highly improbable and would cause major problems in itself. However, our standard case for erosion (6 tons per acre per year) would uncover the wastes in only 10 000 years. This assumes that the wastes are buried in unconsolidated sediments with no erosion protection. The ^{239}Pu , if placed in such an area at the limits for the aquifer case would not reach the limits for the waste exposure case for about 100 000 years. Thus, to qualify for deep burial, the site must have reasonable assurance that erosion rates are such that the actual wastes will be intact for this time period. This would seem to require a site that is relatively stable, or extensive erosion resistance must be provided.

If this option is to be used, the individual site should be documented as to erosion resistance by examination of past geological behavior.

D. Summary of Limits and Conversion to Operational Limits

The limits derived in the previous pathway analyses have referred to the average concentration in the burial ground because this is the quantity generally related to the hazard. For example, the soil limit is averaged over an area dependent upon that used to raise food or upon that from which dust is raised for inhalation. The erosion process, in moving sediments to streams, not only dilutes the contaminated sediment with clean sediments from upstream and downstream of the area but also averages over the particular layer of the burial ground being eroded. In movement to ground water, the averaging takes place both vertically by the random layering of wastes in the pits as well as horizontally by traverse movement of the water as it seeps through the wastes. In intrusion, the averaging occurs by digging through various portions of the waste.

However, any operational limit placed on wastes will refer to the maximum concentration that will be allowed to go to shallow (or deep) earth burial. That is, measurements or estimates will be made of the concentration in batches of the waste and if the waste is lower in concentration than the limit it will qualify for burial. Since wastes lower in concentration than the maximum will be present, the average concentration in the waste will be lower than the maximum used for operational decisions. In addition, the burial ground consists of a series of pits with undisturbed soil between them and the wastes are layered with soil between each layer so that an additional dilution factor exists. Prior to providing a listing of operational limits, we will derive a

dilution factor for each of these sources of dilution so that we can relate the average limit calculated earlier to an operational limit applying to the maximum limit.

1. Dilution by Soils

During burial, dilution occurs from the incomplete use of the burial area. Thus, wastes are placed in the pits or trenches with layers of soil alternating with the layers of waste, and the trenches or pits are located with an undisturbed area between them to allow room so that accidental excavation into a filled trench will not occur when a new trench is dug. A recent survey⁷ of the five major DOE sites where wastes are buried (Hanford, LASL, INEL, SRP, and ORNL) indicated a weighted average for TRU waste buried prior to the establishment of the 10 nCi/gram criterion of about 3500 m³ of waste buried per acre of land. A weighted value based on all low-level waste buried was about 3700 m³ of waste per acre of land.

From Table III the depth of the trenches ranges from 3 - 4.5 m at Oak Ridge to 8 m at Los Alamos. For a 4 m depth of the trenches, the 3500 m³ per acre gives a volumetric dilution factor of 4.6 while an 8 m depth translates to a volumetric dilution factor of 9.2. On a mass basis the dilution factors will be greater for materials with bulk densities less than soil and smaller for materials with bulk density greater than soil. However, the wastes with bulk densities lower than soil generally consist of compactable materials and can be degraded in the ground over long periods of time. In a recent study of wastes exhumed at Savannah River after burial for 14 years,⁷⁹ it was noted that soil around the wastes was dark-colored as if stained by decaying organic matter. Most of this was attributed to the decomposition of the cardboard boxes. However, in general, the wastes were well preserved, particularly those encased in plastic bags, with writing still legible on the paper in the plastic bag. This slow decomposition was attributed to a deficiency of oxygen or nitrogen for the biological processes. They concluded that complete decomposition of wood, cotton cloth, and paper will require several decades while metals and materials encapsulated in plastic will survive indefinitely.

This provides a picture of the burial ground, using present burial practices, of a mass of soil interspersed with remnants of the initial waste in varying degrees of preservation depending upon the time since burial and the conditions at the burial ground. An overall mass dilution factor of 5-10

appears reasonable with small local areas in the burial ground lower than this value. We will use a value of 6 for the overall mass dilution by soils in the burial ground.

The use of this factor of six could be questioned for the scavenger case because he would not spend time digging in the non-productive areas of the spaces between the pits. However, this is not the only source of the soil dilution factor because a part of it is due to the soil covering each layer. In addition, the scavenger does not know which are the productive and non-productive volumes and considerable time will be spent in spaces between the pits looking for the artifacts. We, therefore, believe that the factor chosen is appropriate for this case.

2. Dilution by Less Contaminated Wastes

It is the practice in all DOE facilities to consider any material brought into a process or laboratory area as contaminated when it leaves as waste, whether it has contacted radioactive material or not. This is because of the difficulty and expense of measuring each piece of paper, cloth, rubber, etc. to a level that will assure that contamination levels are minimal and acceptable for uncontrolled release. This results in a dilution of the contaminated wastes with this clean material. Some additional dilution arises from the fact that most of the boxes will have lower concentrations than those at the maximum limit set for burial.

In order to estimate the degree of dilution from this source, data are needed on the distribution of contamination in the individual packages of waste. However, such data are scarce because of the difficulty of making such measurements. Dieckhoner⁷ has surveyed the five major DOE sites and indicates that greater than 97% of the material buried at these sites is only very slightly radioactive or is suspected of being radioactive because of the place where it is generated. These five sites account for 86% of the total DOE waste volume and greater than 99.9% of the activity. If it were assumed that the 3% of the waste that is contaminated is at a maximum limit and the 97% of low radioactivity or suspected contamination is clean, then a dilution factor on the order of 30 would occur.

Data on one type of waste, room trash, are available from the Plutonium Research and Development Facility at Los Alamos. Here, the individual cardboard boxes containing the waste are measured so that they can be routed to either

retrievable storage or burial. The data from nine months of operation are given in Table XXXII.

The data in the highest two categories were strongly affected by operation over one month during this period when 90% of the total number of boxes in these two categories were measured.

The cumulative frequencies in Table XXXII, when plotted against the logarithm of the concentration on probability paper, approximate a straight line indicating that the values are distributed in an approximately log-normal fashion. The dilution factors, considering each of the listed values to be a maximum limit, were calculated assuming the average concentration in each interval to be at the mean of the logarithmic interval. It is highly probable that this procedure underestimates the dilution factor in the range of 10 to 100 nCi/g because of the known disposal of large quantities of trash with no contamination. This would lower the average concentration of 0.32 nCi/g for the 0-1 nCi/g category that makes up the largest volume of the waste. To illustrate the potential change, the dilution factors were also calculated assuming the 0-1 nCi/g category to contain no transuramics. The true answer is probably between these values. Both sets of calculated values are given in Table XXXIII.

TABLE XXXII
PLUTONIUM CONTENT OF ROOM TRASH
AT THE LOS ALAMOS FACILITY

<u>Contamination Levels</u> (nCi/g)	<u># Boxes at Level Given</u>	<u>% of Boxes at Level Given</u>
<1	1 625	90.7
1-10	97	5.4
10-100	39	2.2
100-5000	31	1.7

TABLE XXXIII

DILUTION FACTORS CALCULATED FROM LASL ROOM TRASH

<u>Limit</u> (nCi/g)	<u>Dilution Factors</u>	
	<u>≤ 1 All Contaminated</u>	<u>≤ 1 UnContaminated</u>
10	20	60
100	85	110
5 000	380	390

These data illustrate the expected trend of an increasing dilution factor (or ratio of the maximum to the average) as the maximum permitted increases. Thus, for a limit of 10 nCi/g a dilution factor of 20-60 is expected for these wastes and at a 100 nCi/g limit the dilution factor is increased to 85 to 110.

This is only one type of waste from one facility, with other types of wastes such as sludges or oils probably more uniform than the trash. The use of incineration will tend to increase the uniformity of the transuranium content of individual packages, and the sludges from treatment of wastes have a similar characteristic of relatively constant concentrations. In addition, a single limit on the maximum concentration can result in mixing of wastes or batches of ash or sludge so that the dilution factor predicated above is not appropriate. We believe, therefore, that the dilution factor attributed to the lower contaminated trash is not appropriate for these types of waste.

3. Total Dilution Factor

For the routine trash or decommissioning types of waste, the two dilution factors of soil and lower contaminated waste are independent so that the total dilution factor is 120.

However, for the ash from oxidized combustibles or sludges from water treatment, only the soil dilution factor is appropriate. These dilution factors are not appropriate for the artifacts, either solid items with surface contamination or for trash types of waste contained in nondegradable plastic containers.

3. Conversion to Operational Limits and Summary

The limits derived earlier are described in various sections of this report and are scattered in various tables. In this section we will bring these limits for the individual pathways together so that they can be compared with one another and at the same time we will apply the dilution factors derived in the previous sections, as appropriate, to obtain limits for the various classes of wastes. At the same time we will include the relatively short-lived parent of the transuranium elements ^{241}Pu , ^{242}Cm and ^{244}Cm using the ratios of curie of daughter per curie of parent from Table IV.

Several classes of waste have been described in the derivation of the limits. The first is characterized by the non-uniformity of the contamination levels so that a dilution by less active waste occurs. This consists primarily of degradable trash and random materials in which the contamination is distributed through the matrix rather than being on the surface. The maximum limits for this type of waste, including the dilution by soil and less contaminated waste, are given for each pathway in Table XXXIV. The second class of wastes is characterized by a relative uniformity in concentration, so that the dilution by less active wastes does not occur and the maximum limit differs from the average in the burial ground only by the dilution by the soil. Incinerator ash or sludges from the treatment of liquid waste fall into this category because of the averaging over large quantities of trash for the incinerator ash or the nature of the process for the sludge. Limits for this class, including the soil dilution factor, are given in Table XXXV.

Two sub-classes, based upon the intrusion and artifact hunting scenarios were also defined. These include a surface contamination limit for solid objects and an arbitrary limit on degradable trash packed in non-degradable plastics. It will be remembered that this scenario was included for completeness although no decision could be made in the Task Group as to whether intrusion should be considered if efforts are made to warn. These limits, for the intrusion pathway only, are given in Table XXXVI.

V. RECOMMENDATIONS

A. Shallow Burial

The results of the pathway calculations summarized in tables XXXIV through XXXVI indicate that the exposure of wastes and recovery of artifacts are clearly the controlling pathways and limits based upon these pathways will result in

TABLE XXXIV

MAXIMUM LIMITS FOR DEGRADABLE WASTES FROM TRASH
OR DECONTAMINATION (DILUTION FACTOR = 120)

Pathway	Maximum Conc. to Burial (pCi/g)					
	<u>^{238}Pu</u>	<u>$^{239,240}\text{Pu}$</u>	<u>^{241}Am</u>	<u>^{241}Pu</u>	<u>^{242}Cm</u>	<u>^{244}Cm</u>
<u>Erosion to Stream</u>						
Drinking of Water	-	3.1×10^5	1.3×10^5	4.3×10^7	-	1.1×10^8
Irrigation	-	1.9×10^4	2.2×10^4	7.3×10^5	-	7.0×10^6
Eating Fish	-	2.0×10^5	8.9×10^3	3.0×10^5	-	7.4×10^7
<u>Leaching to Aquifer - use from well</u>						
Drinking	5.2×10^3	780	170	5.7×10^3	1.0×10^6	2.9×10^5
Irrigation	3.2×10^4	1×10^3	110	3.7×10^3	4.4×10^6	3.7×10^5
<u>Leaching to Aquifer, then to stream</u>						
Drinking	6.6×10^8	7.0×10^7	1.6×10^6	5.3×10^7	1.4×10^{11}	2.6×10^{10}
Irrigation	3.8×10^7	4.4×10^6	4.8×10^5	1.6×10^7	7.6×10^9	1.6×10^9
Eating Fish	3.8×10^8	4.6×10^9	9.0×10^4	3.0×10^6	7.6×10^{10}	1.7×10^{10}
<u>Intrusion</u>						
Scavenging	360	360	380	1.3×10^4	7.2×10^4	1.3×10^5
<u>Erosion to Wastes</u>						
Living on Area	2.6×10^3	48	7.2	240	5.2×10^5	1.6×10^4

TABLE XXXV
MAXIMUM LIMITS FOR OXIDATION PRODUCTS OF DEGRADABLE
WASTE OR SLUDGES FROM TREATMENT PROCESSES
(DILUTION FACTOR = 6)

Pathway	Maximum Concentration to Burial (pCi/g)					
	^{238}Pu	$^{239,240}\text{Pu}$	^{241}Am	^{241}Pu	^{242}Cm	^{244}Cm
<u>Erosion to Stream</u>						
Drinking	-	1.6×10^4	6.6×10^3	2.2×10^5	-	5.9×10^6
Irrigation	-	960	1.1×10^3	3.7×10^4	-	3.6×10^5
Eating Fish	-	1.0×10^4	440	1.5×10^4	-	3.7×10^6
<u>Leaching to Aquifer - use from well</u>						
Drinking	260	39	8.4	280	5.2×10^4	1.4×10^4
Irrigation	1 600	50	5.4	180	3.2×10^5	1.9×10^4
<u>Leaching to Aquifer - then to stream</u>						
Drinking	2.9×10^7	3.5×10^6	7.8×10^4	2.6×10^6	5.8×10^9	1.3×10^9
Irrigation	1.9×10^6	6.2×10^3	2.4×10^4	8.0×10^5	3.8×10^8	2.3×10^6
Eating Fish	1.9×10^7	2.3×10^6	5.0×10^3	1.7×10^5	3.8×10^9	8.5×10^8
<u>Intrusion</u>						
Soavenging	90	18	19	630	1.8×10^4	6.7×10^3
<u>Erosion</u>						
Living on Area	130	2.4	0.36	12	2.6×10^4	890

TABLE XXXVI

LIMITS FOR THE MAXIMUM CONTENT OF TRANSURANIC
WASTE BASED UPON INTRUSION PATHWAY

Nuclide	Type of Waste	
	Solid Items with Surface Contamination	Degradable Waste in Non-Degradable packing
	pCi/cm ²	nCi/g
²³⁸ Pu	300	15
^{239,240} Pu	360	3
²⁴¹ Am	60	3
²⁴¹ Pu	2 000	110
²⁴² Cm	12 000	3 000
²⁴⁴ Cm	22 000	1 100

minimal doses from the other. Both of these pathways can be considered as intrusion, which some people feel can be controlled by adequate warnings. However, exposure of the wastes can occur by natural phenomena such as erosion, and at least partial exposure can occur from plants moving the material to the surface, burrowing animals mixing the material, scouring floods or other unforeseen actions. Most of these actions will be slow so that the exposure (and possible spread over areas adjacent to the burial ground) may not occur for thousands of years with some consequent probability that markers or other warning devices will be gone. Thus, we believe that the exposure of the wastes is a possibility and that this pathway is appropriate for limitation of the concentrations buried. On this basis, we would recommend the general limits given in Table XXXVII.

The key difference between these classes of waste is the uniformity or non-uniformity of the contamination levels in the wastes so that the dilution in the burial ground and averaging of exposure is appropriate. Such items as routine trash or decommissioning wastes would qualify for this limit. Uniformly contaminated material would be primarily incinerator ash (or other ashes from the oxidation of combustibles) or the sludges from the treatment of liquid wastes. Of particular concern here would be the use of incineration for all

TABLE XXVII

GENERAL LIMITATIONS FOR SHALLOW BURIAL OF SOLID WASTES CONTAMINATED WITH TRANSURANIC ELEMENTS^a

Nuclide	Waste Type ^b	
	Randomly Contaminated Materials (nCi/g)	Uniformly Contaminated Materials (nCi/g)
²³⁸ Pu	3 000	100
^{239,240} Pu	90	2
²⁴¹ Am	5	0.3
²⁴¹ Pu	200	10
²⁴² Cm	500 000	30 000
²⁴⁴ Cm	20 000	900

^aIf dose rate other than 500 mrem/g is chosen, the values will scale proportionally.

^bValues rounded to one significant figure.

combustible wastes placed into the burial ground with blending by the operator to obtain a final disposal mass that was uniform at or near the limit. Under these conditions, the non-contaminated waste available would be small and the average in the burial ground would approach the maximum limit. In the event that the sludge or ash is "stabilized" by mixing with a solidifying agent, such as concrete, the limit given above should apply to the stabilized mass.

As in past practice with the 10 nCi/g limit, these limits should apply to the average contents of the individual primary package in which the waste is packed. It is not appropriate to average over a number of packages in a secondary container, such as a Dumpster. This procedure is, we believe, adequate to protect against a small portion of the waste containing the bulk of the activity in the package because of the averaging mechanisms of exposure described in the pathway sections.

A second set of limits was derived for which no firm recommendation can be made because of the difference of opinion in the Task Group. These limits are to protect individuals who may intrude into the burial ground, after the 200 year period of control by the government, in search of artifacts. The Task Group, composed of scientific and technical people, believed that the question

of whether warnings are sufficient and adequate should be reviewed by a group with legal, ethical and sociological skills before a decision is made to implement the limits.

In the event that the decision-makers believe that such limits, to protect intruders and artifact hunters, are appropriate, they are listed in Table XXXVI.

B. Deep Burial

The importance of the exposure of the wastes and the possible importance of intrusion led the Task Group to consider the possibility of elimination of these pathways by placing the wastes deeper into the ground where these pathways become impractical.

A firm definition of deep burial is not possible because of wide differences in sites. Instead, it is recommended that each site for which these limits are to be applied must be reviewed carefully for erosion rates and other mechanisms that could expose the wastes. Caverns or abandoned dry mines could be considered as a possibility.

For deep burial, the controlling pathway is possible leaching of the wastes and movement to an aquifer. Limiting concentrations in this case result from domestic uses of the water or irrigation. These limits are given in Table XXXVIII.

TABLE XXXVIII

MAXIMUM LIMITS FOR DEEP BURIAL OF WASTES CONTAMINATED WITH TRANSURANIUM ELEMENTS

Nuclide	Waste Type ^a	
	Randomly Contaminated Materials (mCi/g)	Uniformly Contaminated Materials (mCi/g)
²³⁸ Pu	5 000	300
^{239,240} Pu	800	40
²⁴¹ Am	100	5
²⁴¹ Pu	4 000	200
²⁴² Cm	1 000 000	50 000
²⁴⁴ Cm	300 000	10 000

^aValues rounded to one significant figure.

The same applications to individual packages or treatment described for shall earth burial apply to these limits.

As with any newly proposed limitation on an action, such as burial of waste that has gone on in the past, the question arises as to how these new limits would apply to burial grounds that have been used in the past. We recommend that these limits apply only to future burials with no implication that past burials are unsafe. This is because these limits were derived on a generic basis without reference to individual site characteristics which may be far better than were assumed here. In addition, the exhumation of wastes poses potential risks to those carrying out the operation. We, therefore, recommend that such past burial sites be evaluated on their own merits before any proposal to exhume is considered.

VI. DISCUSSION

In the body of the report we have attempted to explain the choices of scenarios and to discuss the origin of the parameters used and the data available for such choices. There are, however, several matters that should be discussed at greater length so that the implications and consequences of the recommended limits are clear. These items are included in this section.

A. Separate Limits for Nuclides

In the previous draft, there were comments about the problems in operation that would be caused by use of separate limits for individual nuclides rather than recommending a single limit for all transuranics, such as the value of 10 nCi/g.

This approach, of using single nuclides, was taken because each of these nuclides have their specific radiological characteristics as well as characteristic behavior in the environment. It would be possible to recommend a single limit based upon the most restrictive nuclide (^{241}Am in this case), but this was believed to cause other operational difficulties in limiting some of the more important nuclides to artificially low levels. With this approach of single nuclides, emphasis is given to measurement techniques that will provide the information needed. (See next section on measurements) However, even without measurements of individual packages, we believe that operational methods based upon characterization of the materials used in an area and assuming that the same ratio applies in the waste would be successful. Care should be used in

the application of such a procedure, particularly when processes in the plant or laboratory could result in preferential separation of one or more of the nuclides in the waste, but, with reasonable care, the results should be no worse than the uncertainties of the pathway study.

It should also be stressed that the limits for the individual nuclides are not independent of the presence of the other nuclides. That is, one should not permit the full complement of ^{238}Pu and ^{239}Pu or ^{241}Am and ^{239}Pu because the resulting pathway doses will be additive. Instead, the total mixture present in the waste should meet the relation that

$$\sum \frac{C_i}{L_i} \leq 1,$$

(7)

where C_i is the concentration of the i th nuclide and L_i is the limit for that nuclide. Under these conditions the total dose from the pathway will be limited to the reference dose to the maximum exposed individual of 900 mrem/yr.

3. Implementation

In the application of any limit, measurements to determine whether a given item is above or below the limit is vital to assure that actions are taken to comply with the limitation. In past years, it has been customary to assume that any item that had been in a contaminated area was contaminated and was sent to the controlled burial ground for disposal. With the advent of the 10 nCi/g limit, which decided whether the waste should go to retrievable storage rather than to burial, a second decision point was provided that could not be judged upon as simple a basis as was used earlier. Initially, waste was classified on the basis of origin. That is all wastes from one location (any glove boxes) was treated as above retrievable limits, while wastes from another location (any room trash) was treated as suitable for burial. There is little doubt that occasional boxes that should have been stored retrievably went to burial while others that could have been buried were placed in retrievable storage. The first path would cause only an incremental increase in any hazard in the burial ground because of the averaging effects in the pathways and the vastly greater volume of wastes with low contamination. The second pathway caused possible

unnecessary use of retrievable storage with consequent future implications for costs and risks at the time the wastes are retrieved.

The need for methods of differentiating the transuranic content of various packages of waste was recognized shortly after the 10 $\mu\text{Ci/g}$ limit was instituted and research was started to provide instrumentation to be applied to the waste handling operations. The first measurement system was applied to 57 liter, cardboard boxes of low density wastes, such as are generated in plutonium handling facilities. This resulted in MEUAS I, in which the box is rotated and measurements made of the 17 KeV X-rays with a sodium iodide crystal. This system has been used routinely at the Los Alamos Scientific Laboratory for several years with sensitivities on the order of 1 $\mu\text{Ci/g}$. Recently a MEUAS II, which incorporates a hyper-pure, germanium, photon detector and a ^3He neutron detector, has been placed in operation. This unit provides enhanced capability of detecting transuranium elements in higher density matrices and with higher backgrounds.

While these units have operated well for the types of waste for which they are intended (those with low fission-product or activation product content) there is still need for the investigation of methods of measuring other wastes with high density matrices or high content of other radionuclides that will interfere in the measurement. Work is being done with an active interrogation system using a 30 MeV electron accelerator with monitoring of prompt and delayed neutrons, as well as photons, as signatures of the transuranium elements. However, considerable work is required before a satisfactory, operational system can be built.

A more detailed account of the measurement techniques now in existence and the possibilities for further development is given in Appendix J.

C. Population Doses

As was discussed in the criteria section, a basis for calculation was derived that limited the dose to the organ receiving the highest dose of the individual most exposed to the transuranium elements once these materials are released from confinement. Additional doses, however, will be received by other people so that the population dose would include these doses as well. We have not attempted to include an estimate of the population dose for the postulated scenarios because this would mean a detailed study of the environment of each site and an extrapolation of both the physical and cultural environment to the

times that the exposure scenarios take place (100 to several thousands of years). We believe that the uncertainty involved in such extrapolations and calculations would be so great as to negate the usefulness of any calculations.

However, there is assurance that the population doses will be lower than the limiting doses used in these estimates. The exact amount lower depends upon other imponderables in the estimates - particularly the customs and habits of the individuals who make up the population, since these will define not only the possibility of exposure but also the degree of exposure. Qualitative estimates of the relationship between the dose to the maximum exposed individual and the average population dose for several important pathways will, however, provide some indication of the magnitude of dose expected.

Soil Pathway The soil pathway occurs in the scenarios involving irrigation and exposure of wastes. Normally one would consider the number of people living full time on either area to be relatively small, and intermittent exposures from workers in the area or visitors would be well below those for which the limits were based - continual residence. Under these conditions the population dose would be to only a few people. However, the possibility exists of having the irrigated area or the area of waste exposure used as housing area - such as a subdivision. Under these conditions a population on the order of 10 000 people could be exposed. However, the limit is based upon an individual growing his own food and inhaling an average of $200 \mu\text{g}/\text{m}^3$ of dust derived from the soil. The actual exposure of members of a population will vary from about this for individuals who work outdoors in a dusty environment down to, perhaps, 5-10% of this for individuals who work elsewhere and spend most of their time at home indoors. The ingestion pathway depends strongly on the amount of food grown at home. For the limiting case we have considered the individual who grows most of his food at home. This is, of course, more appropriate to use of the land by individual farmers than a well-knit community. However, there could be numerous home gardens.

In order to estimate the population dose, data to provide the frequency of the various degrees of exposure must be available for the particular community. These frequencies will vary depending upon the type of community (i.e. suburban, urban or rural) and the climate, which will control the amount of time outdoors and the type and degree of gardening. (Note that the exposure will also vary depending upon the amount of rain or snow cover affecting resuspension).

Detailed data of this nature are not available for different types of communities in different parts of the country. However, it is clear that the average doses and dose rates to a heterogeneous population will be well below those received by the maximum individual. The population dose from americium will be particularly low because a large part of this dose arises from ingestion of foods.

Stream Contamination Contamination of streams with transuranium elements will occur in the erosion and leaching to aquifer with discharge to a stream scenarios. Here the dose arises from drinking of the water and eating of fish with the eating of fish the controlling pathway for the maximum exposed individual. In these scenarios total population doses will depend upon the populations downstream who depend upon the water for drinking, the fraction of the population that uses fish from the stream, and the characteristics of the stream. If this is a large population a larger population dose could accrue from drinking of the water. However, the decrease in concentration downstream, to 1% in a stream such as the Columbia River or to 20% in a stream such as the Tennessee River, will tend to limit the total dose and will make the total dose highly dependent upon the actual locations of population centers downstream from the discharge from the burial ground to the stream.

We believe that this pathway could result in the highest population dose.

Intrusion The vast majority of the dose from intrusion would be to the individuals intruding in the area so that the population dose could be estimated by the product of the dose to the intruder and the assumed number of individuals involved. There would be an additional contribution from the contamination on the intruder's clothes or person and from any artifacts removed but these doses would be highly dependent upon the specifics of the case. It is also noted that the data required to provide estimates of the dose, even in defined cases, are extremely meager so that any estimate would be poor.

D. Comparison With Natural Radioactivity

The burial and possible release to the environment of transuranium elements poses no new hazards in kind to the human race because he has evolved in the presence of natural radiation and radioactivity. Because the present limit of 10 nCi/g for shallow-earth burial of solid wastes contaminated with transuranium

elements was derived by comparison with levels of natural alpha emitters in high radioactivity regions of the earth's crust, a comparison of the present limits with levels of natural radioactivity is in order. A more complete discussion of natural radioactivity is given in Appendix K.

Alpha emitters in uranium and thorium chains and their daughter products are ubiquitous in the earth's crust with average levels of 5-10 pCi/g. There are regions in Brazil and India with sizeable populations where the rocks and soils contain an average of 5-20 nCi/g of alpha emitters.

The effects on human health from these natural deposits have been investigated, although not in the detail desired, with no indication of a gross effect that would make people not live in these areas. There have been changes, such as chromosome aberrations, detected that are attributed to the radon isotopes, but any correlation between these effects and human health has not been established. Thus, it would appear that levels of natural activity on the order of nCi's per gram produce no striking incidence of disease.

There are major differences between these natural deposits and the transuranium containing wastes. The area used for transuranic burial is small compared to areas underlain by ore deposits, so that exposure to transuranium elements will be to a smaller population than for natural alpha emitters and occupancy times will be lower. The natural materials have migrated to their present positions over very long times and are now well stabilized, while the transuranics may be in many chemical states with their stabilization yet to occur. The carcinogenic properties of all alpha emitters are grossly similar, with differences between them due to differences in site of deposition in the body. The mobility of many of the natural isotopes in the environment is greater than that of the transuranics so that a larger uptake by man can be predicted. Radium, a chemical analog of calcium, is an example with the best known movement the uptake in Brazil nuts. A different type of mobility is exhibited by radon. This material is a noble gas with alpha-emitting daughters. Since it is not chemically bound, it is released from the soil and causes exposure of people in the neighborhood through inhalation. An important component of the radiation from these natural alpha emitters is the gamma radiation produced in some of the decays. In areas of high concentration in the soil this can produce levels between 0.1 and 1 R/yr. For the transuranics, little gamma radiation is emitted until the isotopes encountered in nature are

reached in the decay chain, and by this time the quantities are reduced by factors of 10^{-3} or more.

Thus, the natural activities will produce alpha radiation doses that will be similar to the transuranics. The differences in dose caused by exposure to natural deposits of alpha emitters and the transuranics in soil depend upon many of the physical and chemical characteristics that have not been thoroughly evaluated. From the above, however, it appears that natural deposits are at least as hazardous at the same level as the transuranics.

This gives a clue as to the order of magnitude of effects that may be observed, however, in a much smaller population, from final levels in the soil of transuranics. With this comparison and the overall dilution of 120 between the limit on the maximum concentration and the average in the emplaced waste, the waste limit would be 100-1000 nCi/g to produce the low, but still unknown, levels of harm that may occur in these natural deposits.

A more quantitative comparison can be made by consideration of the dose to the bone from natural emitters as compared to the limiting bone dose of 500 mrem/yr used in this study. Measurements have been made of the quantity of certain key nuclides in total bone, in the total body, and in bone ash. These nuclides include ^{226}Ra , ^{210}Po , ^{238}U , ^{232}Th , ^{226}Th and ^{226}Ra . These nuclides all produce alpha doses to the bone, either through their individual emission or from alpha-emitting daughters.

The data available in the literature on the bone or body content of these natural nuclides varies in extent and in the number of people measured, with ^{226}Ra receiving the greatest attention and ^{228}Ra and ^{228}Th the least. We have not attempted a complete literature review for nuclides such as ^{226}Ra and ^{210}Po but have selected several papers in which data from investigators other than those making the study reported are listed. We did, however, check to see that the values used are comparable to others reported in the literature.

In general, we have used values for bone ash rather than whole body measurements or estimates because these values are closest to the quantity desired - concentration per gram of mineral bone. To convert to mineral bone we noted, that Moltmann⁹⁴ determined that the average ratio of calcium content, expressed as $\text{Ca}_3(\text{PO}_4)_2$, to the bone ash content is 1.001 ± 0.012 , indicating that the ash composition can be taken as $\text{Ca}_3(\text{PO}_4)_2$ for our purposes. This led to a calcium content in the bone ash of about 39%. The ICRP, in their reference man study,⁶⁹ indicate that the calcium content of bones without marrow for

adults were 19.7% for the skull, 17% for the rib and 14.6% for the ilium. For the following calculations we assumed a value of 15%. Thus, the ratio of bone ash to mineral bone was taken as the ratio of these two or about 40%.

Dose calculations were done using the same methods as for the transuranics with the exceptions that the dose distribution factor was taken as unity and the actual energies of the nuclide and/or the appropriate daughters were used.

Dose rates for ^{232}Th and natural uranium were estimated from the data given by Lucas et al.⁹⁵ for ^{232}Th and by Hamilton⁹⁶ for uranium. The uranium dose rates were only a small fraction of one mrem/yr and were not considered further. The ^{232}Th dose rate was only about 1 mrem/yr for contents close to the upper end of the quantities found and was, again, ignored in the final tabulation.

The ^{226}Ra and $^{210}\text{Pb} - ^{210}\text{Po}$ doses were estimated from a table given by Hunt et al.⁹⁷ that included data on 546 cases for ^{226}Ra and 136 cases for ^{210}Pb originating in the northeast and midwest U.S.A. The average bone contents (corrected from bone ash) were 0.007 pCi/g for ^{226}Ra and 0.06 pCi/g for ^{210}Pb , which result in doses of 14 mrem for ^{226}Ra and 60 mrem for the $^{210}\text{Pb} - ^{210}\text{Po}$.

The data for ^{228}Ra and ^{228}Th are much less extensive with six measurements given by Stehney.⁹⁸ Because ^{228}Th is a daughter of ^{226}Ra and is the parent of a chain of relatively short-lived nuclides, we utilized the data for ^{228}Th for dose calculations. The longest-lived daughter in the chain following ^{228}Th is ^{228}Ra with a half-life of 3.64 days. We, therefore, assumed the total alpha energy of 31 MeV liberated in the chain to be effective in producing dose. The average quantity of ^{228}Th in the bone was 0.0053 pCi/g which leads to a dose of 31 mrem/year.

Thus, the average bone dose from natural, alpha-emitting radionuclides is on the order of 100 mrem/yr. However, the same sources of data also include the range of individual values so that we can estimate a maximum bone dose in the sample sizes available. These values are 160 mrem/yr for ^{226}Ra , 200 mrem/yr for ^{210}Pb and 90 mrem/yr for ^{228}Th . The ^{210}Pb doses were estimated with two high values, leading to 1800 mrem/yr and 530 mrem/yr eliminated. We believe the value of the maximum for ^{228}Th to be possibly low because of the very small sample size. Because these measurements were made on different individuals, the possibility of all occurring in the same individual is unknown. However, Stahlkofen⁹⁹ indicates that the elevated activities of ^{228}Th were found in bones that contained high levels of ^{226}Ra indicating the possibility that the maximum for both natural activity series would coincide in the same person. On this

basis we would estimate that the maximum in the series measured to date could have bone dose rates from alpha emitters on the order of 500 mrem/yr.

E. Estimates of Risk

A quantitative estimate of risk to people exposed to the pathways postulated in this study can be made by assuming a linear, no-threshold relationship between cancer and dose, using observations of human populations at much higher doses and dose-rates than are used here, to provide the coefficients. Such estimates are crude because of the dosimetry in the populations studied and the conditions under which the people were exposed - usually high dose and dose-rate. One cannot exclude a zero effect at the low-doses and dose-rates in this study by any scientifically acceptable procedure, but we will, in the following, assume the linear, no-threshold hypothesis to apply.

The most exhaustive study and application of human data to risk estimates has been that of the National Academy of Sciences, Biological Effects of Ionizing Radiation (BEIR) Committee in 1972.²² This report is now being reviewed and revised but the new conclusions are not available. The BEIR report postulates four different models for calculation: 1) an absolute risk model with a given absolute risk per rem, and "plateau" (time over which the risk is expressed) less than a lifetime; 2) an absolute risk with a plateau extending over the remainder of life; 3) a relative risk in which the risk is increased by a percentage of the existing risk and a plateau less than a lifetime; and 4) a relative risk with a lifetime plateau. Coefficients, latent periods and plateaus were obtained by study of human data. The doses were expressed in rems, which causes a technical problem because the rem uses a Quality Factor defined for radiation protection purposes rather than a relative biological effectiveness for a particular type of cancer, but this makes little practical difference in view of the many other uncertainties.

The coefficients used in these calculations were taken as much as possible from the BEIR study with resort to other references only when the particular value could not be found in the BEIR report. Both the absolute and relative risk model were used with latent periods and plateau lengths as recommended in the BEIR report. These coefficients are given in Table XXXIX.

As can be seen from the table, the coefficients used came from a variety of sources. This was made necessary by the fact that the BEIR report did not give

TABLE XXXIX
PARAMETERS USED IN ESTIMATING RISKS

Organ	Latent Period (years)	^a Plateau Length (years)	Absolute Risk (Cancers/year)	Relative Risk (\$ increase/year)
Lung	15	15	1.3×10^{-6}	0.3% ^b
Bone	10	30	2×10^{-7}	1.1% ^c
Liver	15	30	8×10^{-8d}	1.1% ^e

^aFor model with effectiveness less than lifetime.

^bFrom BEIR Report²² pp. 150.

^cFrom BEIR Report²² pp. 132.

^dFrom Ref. 99.

^eFrom Ref. 17.

values for the liver, and the summary tables provided only the relative risk for all cancers other than leukemia. We used a latent period for bone cancer somewhat shorter than the BEIR report would indicate because their table (pp. 129) in which the risk estimates were derived indicated a relatively short latent period.

The absolute risk coefficients are lower than those used by the EPA in their proposed guidelines for transuranium elements in the environment.¹⁷ The lung and bone risk coefficients are both lower by about a factor of two than the EPA values, while the liver risk coefficient is lower by about a factor of five. It was impossible to compare the relative risk coefficients because they were not explicitly given in the EPA document. However, it is noted that the lung coefficient is based largely on the uranium miners data and a dose to the presumed most sensitive tissue, the bronchial epithelium, rather than to the total lung.

The risk per year was calculated for each of the four models and converted to the individual risk in age groups by using the population distribution, and, for the relative risk case, the incidence of bone, lung and liver cancers from the 1970 Vital Statistics.¹⁰⁰ These data are shown in figures 2 and 3 as

Fig. 2. Results of Risk Calculation for ^{239}Pu for intake resulting in 500 mrem/yr to bone after 70 years.

——— Limited Plateau
 ——— Lifetime Plateau
 - - - - - Relative Risk
 ——— Absolute Risk

compared to the total cancer risk for each age group from the 1970 Vital Statistics. To aid in presentation we have shown the total cancer risk as a line across the age group while the estimated risks from exposure to ^{239}Pu and ^{241}Am are drawn as a smooth curve through the center of the age distribution.

The data are presented as individual risk, rather than risk per 100,000, because the basis of the calculation was the inhalation or ingestion of sufficient nuclide to result in a dose of 500 mrem/yr to the bone at the end of 70 years. This is the description of the maximum exposed individual so that populations will not be exposed to these levels.

Fig. 3. Results of Risk Calculation for ^{241}Am for intake resulting in 500 $\mu\text{rem/yr}$ to bone after 70 years.

— Limited Plateau
 - - - Lifetime Plateau
 . . . Absolute Risk
 -o- Relative Risk

An alternate method of presenting these data is the total lifetime risk to the maximum exposed individual. These data are given in Table XL, again as the risk to this individual. The ^{239}Pu soil limit corresponds to the 200 pCi/g which results from a contribution of 2/3 inhalation and 1/3 ingestion. The ^{241}Am limit is controlled by the ingestion under the assumption used in these calculations.

An average lifetime risk can be estimated from these figures by dividing by the assumed lifetime of 70 years. For the ^{239}Pu soil limit this ranges from about 6×10^{-7} to somewhat over 1×10^{-6} per year. For ingestion of ^{239}Pu (as in

TABLE XL

LIFETIME RISK FROM INHALATION AND INGESTION EXPOSURE
RESULTING IN 500 MRAD/YR TO BONE AFTER 70 YEARS

Nuclide	Pathway	Type Model	Plateau	Lifetime Plateau
^{239}Pu	Inhalation	Absolute	5×10^{-5}	7×10^{-5}
		Relative	1×10^{-4}	2×10^{-4}
	Ingestion	Absolute	1×10^{-5}	1×10^{-5}
		Relative	2×10^{-5}	2×10^{-5}
^{241}Am	Inhalation	Absolute	1×10^{-5}	1×10^{-5}
		Relative	2×10^{-5}	3×10^{-5}
	Ingestion	Absolute	1×10^{-5}	1×10^{-5}
		Relative	2×10^{-5}	2×10^{-5}
^{229}Pu	Soil	Absolute	4×10^{-5}	5×10^{-5}
		Relative	1×10^{-4}	1×10^{-4}

eating of fish or drinking of water) this average risk is 1×10^{-7} to 3×10^{-7} per year. For ^{241}Am the average risk for both soil and ingestion is 1×10^{-7} to 3×10^{-7} per year.

It must be emphasized that these are the estimated risks to the maximum exposed individual. Risks to an average individual would be considerably lower.

We acknowledge the uncertainty in these estimates with possibly higher risks possible if one accepts other risk coefficients. However, we believe the values given above are within an order of magnitude of the maximum estimates.

F. Uncertainties

Models and combinations of models, such as we have used in this study, are simulations of reality and mirror reality only to the extent that our knowledge and assumptions agree with nature. Another factor that is difficult to account for is the variability in the habits of people and in locations that could result in different transport than used in these estimates.

In this section we will review the major parameters used in the study and attempt to assign an uncertainty to each parameter. We have included rough estimates of the ranges of values that could exist in the models used for dose calculation due to differences in size, sex and activities of people in order to provide some measure of the uncertainties engendered from this source.

This examination is confined to only those pathways that result in the limiting recommendations in order to keep the size and complexity of the examination to a reasonable limit. These pathways include exposure of the wastes with people living on the area for shallow burial and leaching to an aquifer with use of the aquifer for domestic water and irrigation.

In Table XLI we indicate, for each of the parameters, the value used in the study, an estimated range of values and the multiplicative effort of this range on the dose assuming that the parameter, as used, leads to a dose of 500 mrem/year to the maximum exposed individual. Each of the parameters is numbered and a discussion of the basis for the estimated range is given below. It should be noted that we did not attempt a statistical treatment in deriving these ranges because the amount of information available for many of them is meager and even the estimate of range is uncertain.

1. For the ICRP reference man⁶⁹ the weight of total lung tissue (parenchyma plus arterial and venous blood) is listed as 1000 g in the male and 800 g in the female. The 500 g value used was approximately that of the lung tissue without blood as a conservative estimate. However, since the blood does absorb alpha energy, the higher values may be more appropriate. The ranges were obtained from data on blood-free lungs from males given in ICRP-23.⁶⁹ These data indicate a factor of about 1.5 between the lowest and highest values or a factor of ± 1.25 from the mean. These ranges were applied to the reference man weights.

2. Data in ICRP 23⁶⁹ indicate that the skeletal weight varies from 9% to 18% of the body weight in males and 5.6% to 15% in females. It was assumed that this skeletal weight includes marrow with the mineralized portion of bone, of interest here, as one-half of the skeletal weight. These fractions were applied to a male body weight range of 120-200 pounds and a female body weight range of 100-180 pounds.

TABLE XII
ESTIMATES OF THE RANGE OF UNCERTAINTY IN KEY PARAMETERS

<u>Parameter</u>	<u>Value Used</u>	<u>Estimated Range</u>	<u>Effect on Dose</u>
<u>Dose Calculations</u>			
1. Lung Weight	500g	300-600 (600-1200)	0.6-1.2 0.4-0.8
2. Bone Weight	5 000g	2000-8000g	0.6-2.5
3. Quality Factor	10	10-20	1-2
4. Bone Distribution Factor	5	5-15	1-3
<u>Inhalation</u>			
5. Particle Size	1 μ m	0.1-10 μ m	0.5-2
6. Inhalation Rate	20 m ³ /day	10-27 m ³ /day	0.5-1.4
7. Class Y - Fraction to Bone	0.057	?	0.5-2.0
8. Class W - Fraction to Bone	0.12	?	0.5-2.0
9. Am - Class	W	W-Y	0.4-1
10. T _{1/2} - Lung - Class Y	500	150-2000	-
<u>Ingestion</u>			
11. Fraction-GI to Bone-Pu	4.5x10 ⁻⁵	9x10 ⁻⁶ -3.5x10 ⁻⁴	0.2-8
12. Fraction-GI to Bone-Am	2.25x10 ⁻⁴	3x10 ⁻⁵ -3x10 ⁻⁴	0.1-1.3
13. Biological Incorporation	0	0.2-13	0.6-8
<u>Soil Limit</u>			
14. Resuspension	200 μ g/m ³	20-200 μ g/m ³	0.1-1
15. Particle Size Correction	1	1-2	1-2
16. Plant uptake - Pu	W.Y. Diet	-	0.2-2
16a. Plant uptake - Am	10xPu	-	0.1-2
17. Diet	628 kg/y	-	0.1
18. Occupancy	1	0.9-1	0.9-1
<u>Living on Area</u>			
19. Dilution by clean soil	0.5	1-0.1	0.2-2
<u>Water Limit - Aquifer (Deep Burial)</u>			
20. Geology - Hydrology	-	-	0-1
21. Leach Rate	10 ⁻⁶ yr ⁻¹	10 ⁻⁷ -10 ⁻⁵ yr ⁻¹	0.1-10
22. Velocity of Aquifer	30 cm/day	-	-
23. Depth of Aquifer	10 m	5-50 m	0.2-2
24. Porosity of Aquifer	0.1	0.1-0.3	0.3-1
25. Water intake	1650 cm ³ /day	365-3000 cm ³ /day	0.2-1.8
26. Erosion Rate of Soil	6 T acre ⁻¹ yr ⁻¹	0.1-20 T acre ⁻¹ yr ⁻¹	0.3-60

TABLE D-III

RETENTION IN VARIOUS ORGANS OF PIGS AND RATS
AFTER A SINGLE INTRAGASTRIC ADMINISTRATION

Organ	<u>% of Administered Dose</u>	
	Pigs	Rats
Skeleton	0.0015	0.0020
Liver	0.00036	0.00093
Kidney	0.000009	-
Spleen	0.000003	-
Heart	0.0000007	6
Other Soft Tissue	0.00033	0.00021
Total Soft Tissue	0.00071	-
Total Animal	0.0022	0.0032

The rats excreted 88% of the dose administered in the feces and 7.7×10^{-4} % in the urine in the 9 days before sacrifice. If one ignores absorption and return to the bowel and includes only the urine data as a measure of absorption, a total value of about 0.004% of that fed is obtained.

In the third part of this experiment, studies were made of the effect of valence state and the use of citrate instead of nitrate on the absorption. In this experiment, solutions containing approximately 0.5 mg/ml were oxidized to the VI valence state with $K_2Cr_2O_7$, and reduction to the III state was accomplished with $FeSO_4$ and urea. The citrate solutions contained a ten-fold excess of citric acid and, in all cases, the desired pH was achieved by dilution and adjustment with NaOH. After preparation, the solutions were allowed to stand for 24 hours before administration and the plutonium valence state was determined at or about the time of administration. A single 1 mL dose was given by stomach tube to 12 young, adult, male Sprague-Dawley rats. Results from sacrifice of half of the group at 4 days and the other half at 60 days are given in Table D-IV.

It is difficult to draw quantitative conclusions from these data because of the lack of knowledge of the valence state in many of the experiments. However, it is apparent that these data show the uptake of both Pu(III) and Pu(IV) to be greater than that of Pu(V).

The deposition of ^{239}Pu in the liver and skeleton of miniature swine was studied by Bustad et al.⁷ Swine were used because their gastrointestinal tract is similar to that of man. Only the liver and skeleton were analyzed because

3. The Quality Factor is a quantity defined for use in radiation protection taking into account the relative biological effectiveness for different types of harm. The QF of 10 for alpha particles has been used by the ICRP and NCRP since the 50's. Recently the ICRP has changed to a QF of 20¹⁰ and this has been used by the EPA¹⁷ in their study of transuranium elements in the environment.

4. The dose distribution factor of 5 for bone has been used for all alpha-emitters except radium since 1950. It was derived from the data available on the effects of radium in man by biological comparison between radium and plutonium.¹⁰² The ICRP has announced their intention of setting the limit based upon calculation of the dose to the endosteal and epithelial cells on the bone surface¹⁰, but have not, as yet, specified a method for calculation. Harley and Pasternack¹⁰³ have calculated the radiation doses from ²³⁹Pu and ²²⁶Ra to cells on trabecular bone surfaces. The estimated 30-year dose to humans, for maximum permissible body burdens of ²²⁶Ra and ²³⁹Pu, were higher for ²³⁹Pu than ²²⁶Ra by a factor of 1.2. They concluded that this difference was not significant. Spiers and Whitwell¹⁰⁴ estimated the radiation dose received by endosteal cells and concluded that ²³⁹Pu is a factor of 4.5 times as damaging as ²²⁶Ra. In both of these cases, 90% of the plutonium in the body was assumed to be in the bone. Dougherty and Mays¹⁰⁵ have estimated the relative damage factor to be 15 from the extended study with ²³⁹Pu and ²²⁶Ra in beagle dogs at Utah.

5. Although resuspension studies show fractional deposition about equivalent to a 1 μ m particle in the ICRP Task Group lung model or measured aerodynamic size in the range of 1-3 μ m, it is conceivable that in specific situations higher or lower sizes could be encountered. A maximum range from 0.1 to 10 μ m seems reasonable.

6. The ICRP reference man⁶⁹ gives values for the adult man of 23 m³/day with 42% of this inhaled at work and 21 m³/day for an adult woman with 43% inhaled at work. The value used in this report of 20 m³/day was used in the ICRP-II²¹ and NCRP¹¹ reports on internal emitters. The Task Group on Lung Dynamics¹⁹ provided calculations for a respiratory frequency of 15 sec⁻¹ and tidal volumes of 750 cm³, 1450 cm³ and 2150 cm³ that correspond to total inhalation of 16, 31 and 46 m³/day. The 750 m³ volume was quoted as being

representative of mild to moderate activity. ICRP²³ reports an inhalation of 7500 ml/min for men at rest and 6 ml/min for women at rest. These correspond to daily intakes of 11 and 9 m³/day respectively. Thus, a minimum rate of inhalation is taken as 10 m³/day while the maximum will be taken as a male resting 8 hours/day and working at moderate to heavy activity with an inhalation rate of about 35 m³/day for the remainder of the time. This is about 27 m³/day.

It should be recognized that there is a systematic variation in these intakes with body size and a random fluctuation with individuals dependent upon their age and state of health so that some values below the minimum and above the maximum are possible.

7. The fraction of that inhaled to blood is taken from the deposition and uptake parameters of the ICRP Task Group on Lung Dynamics¹⁹ and from the ICRP study on metabolism of actinides.²⁰ There are many possible variables in these parameters that would require essentially rederiving the models to estimate them. For example, the deposition depends upon the tidal volume and inhalation rate of the individual which will vary depending upon his activity. We have estimated a variation from about 0.8 to 1.3 for a change in tidal volume from 750 to 2450 cm³ at a respiration rate of 15 sec⁻¹. There is some variation with the particle size distribution. From the curve on page 161 of the Task Group report¹⁹ we estimate this to range from about 0.7 to 1.1 for 1 μ m AMAD particles with σ_g from 1.2 to 4.5 with a tidal volume of 1450 cm³. The influence of deposition during mouth-breathing is not well quantified but this action does bypass the protective features of the nasopharyngeal region and a higher deposition is expected in the tracheobronchial region. The Task Group¹⁹ points out that there is evidence of trapping in the mouth. However, it is difficult to model this because the deposition will be sensitive to the degree to which the mouth is open. They assume that this will be an infrequent problem and, in most cases, will not affect pulmonary deposition.

The fraction transferred to the blood from the individual regions of deposition also contains uncertainties that are difficult to quantify because of the lack of detailed data on each. Similarly the transfer from blood to bone and liver, taken as 0.45 to each, has uncertainties that are difficult to quantify. The data in Appendix D indicate that plutonium absorbed from the GI Tract may result in a skeleton to liver ratio of about 5 rather than one as in

the ICRP model. However, the application of this to inhaled material is questionable.

As a result of these many uncertainties, we have arbitrarily chosen to assign a range of a factor of two to cover all of these uncertainties. The question mark in the "Possible Range" column indicates the arbitrary nature of this choice.

8. The same arguments as were used for Class Y apply also to the Class W and the same factor applies.

9. The choice of ^{241}Am as a Class W material was based upon the fact that it is a +3 valence actinide, a class that is usually more soluble than plutonium. It may be that certain of the compounds are more Class Y than W. For example, Craig et al.¹⁰⁶ present data for $^{241}\text{AmO}_2$ that indicate that the long component in the lung of dogs may have a half-life on the order of 400 days. However, Ballou and Gies¹⁰⁷ present data for $^{241}\text{Am}(\text{NO}_3)_3$ with rats that indicate a half-life of about 50-60 days. Since the exact state of ^{241}Am in the soils is unknown, and, in fact, may vary from one place to another, the possibility that it may behave as a Class Y aerosol in some conditions cannot be discounted.

10. The half-life of 500 days in the ICRP Task Group Model can be exceeded by high-fired plutonium oxide. The Task Group¹⁹ report lists a range of half-lives in the lung from 150 to ≥ 1400 days in mice, dogs and rabbits. This will have little effect on the limits because bone is critical. However, if the half-life in the lung becomes long enough then the lung will become critical. This turnover point is estimated to occur at a 1000-1500 day half-life.

11. The range is the range in measured uptake from nitrate solution as given in Table D-XXXI. The upper value is about twice the next highest but is included for completeness. It is also in the same range as the uptake from 4-5% citrate solutions in Table D-XXXII.

12. The range of values is that from Table D-XXXIII for ^{232}U , ^{233}U and ^{241}Am .

13. The range was obtained from the data on experiments with rats and guinea pigs fed solutions and incorporated plutonium. However, it was assumed that only one-half of the transuranium element was incorporated, with the remainder from surface deposition. The 0 in the "value used" column indicates that no correction was applied while the range indicates the measured ratios between incorporated and control experiments.

It must be remembered that this is a difficult experiment to perform and the results are variable.

14. A value of $200 \mu\text{g}/\text{m}^3$ average was used, composed of exposure indoors at $50 \mu\text{g}/\text{m}^3$, ambient air outdoors at $100 \mu\text{g}/\text{m}^3$ and work exposure that averaged $400 \mu\text{g}/\text{m}^3$ over the year. These are not actual air concentrations but result from a correlation between the measurement of ambient air and calculations using a dust loading of $100 \mu\text{g}/\text{m}^3$. These are high dust loadings that we believe will be difficult to exceed. We have, therefore, chosen the $200 \mu\text{g}/\text{m}^3$ as the maximum. Lower dust loadings would be possible if, for example, a major part of the area were paved or planted in lawns and the people worked mostly in offices - perhaps air-conditioned offices. For this condition an average dust concentration originating from the contaminated area could be in the range of $10\text{--}20 \mu\text{g}/\text{m}^3$. For this study we used $20 \mu\text{g}/\text{m}^3$.

15. Both Tamara⁶⁵ and the EPA⁶⁴ have estimated correction factors to account for increased concentrations in the smaller particles that are more readily deposited in the lung. These correction factors range from 1.4 to 2 for soils investigated to date. We have rejected the much larger increase proposed by Johnson et al.³⁸ in using only the $5 \mu\text{m}$ particles in the soil, after aggregates are broken, because his sample does not correspond to conditions actually found in soils and he has demonstrated no mechanism for calculating air concentrations from this sample.

16. The variability of measurements on plant uptake is discussed in the body of the report with individual measurements for plutonium ranging from 10^{-10} to 10^{-3} and for americium from 10^{-7} to 10^{-1} . We used the New York diet because it represents a sampling of foodstuffs that are actually eaten by people and these foods incorporate the variability that has been found in the individual plant species as well as in experimental conditions. We believe that the values

for plutonium may be high by an unknown factor because of the continued deposition of plutonium from the atmosphere. They may be low, however, for a specific area because the soil properties may result in higher uptake.

The americium was arbitrarily taken as ten times the plutonium uptake after review of the data available. However, it is noted that the measurements at the Environmental Laboratory⁵³ indicate about the same uptake.

For plutonium we will assume the uptake could vary from about 20% of the value used to about twice the value. For americium, the assumed range is 0.1 to two times the coefficients used.

17. The intake of food grown on the contaminated area can be extremely variable depending upon the actual conditions. In the calculation we have assumed that the diet consisted of food grown entirely on the contaminated area - essentially a small farm on the area. Under other conditions, however, none of the food used in the diet would be grown on the area. Thus, the range of 0 to 1 for the factor.

18. In the calculations we assumed continued occupancy of the area for 70 years. In our mobile society, it seems highly unlikely that an individual would live on a 250 acre plot of ground (the size of our model burial ground) for 70 years and never leave to visit relatives, to shop or to take vacations. However, it is possible and the range will be set from 0.9-1. 0.9 amounts to 35 days per year away from the area.

19. The relative amount of clean soil mixed with the waste material at the time that the land is occupied will depend upon the method of exposure of the wastes. We predicated the scenario on exposure by erosion with clean soil brought into the area by the erosion process. In an extreme case, where the burial ground is at the upper end of an erosion area, little or no clean soil would be brought in and the dilution with clean soil would be zero. On the other hand, if exposure occurs by digging a pipeline or foundations of houses, only a portion of the area would be exposed and the effective dilution factor could be as high as 10 parts of clean soil to 1 part of contaminated.

The other factors involved in this scenario have been discussed in the soil limit case.

20. The geology-hydrology of the burial area is a prime factor in determining the potential exposure from movement to, and use of, the aquifer. Table XXIII shows the estimate of the distance traveled in times to 10 000 years. Movement will be slower by an undetermined amount in the leaching from the burial ground to the aquifer since a part of this flow is in unsaturated conditions and the rest is the relatively slow movement by percolating water. This table envisions a relatively uniform soil with high adsorptive capacity. However, in extrapolating, one would predict a movement of only 70-200 meters in 100 000 years if the conditions of the leachate were such that a K_d of 10^5 occurred. All conditions between this and an almost instantaneous discharge to these depths are possible, either by a lower K_d or a flow through sizeable fissures. Thus, if the burial ground is between 70 and 100 meters above the aquifer, the discharge to the aquifer will range from zero to about what we postulated in the model burial ground.

Again, depending upon the character of the aquifer, additional removal and dispersion could occur between the point of entry into the aquifer and the point of use.

21. The leach rate of 10^{-6} per year was derived from relatively uncertain data at Maxey Flats. Other data that are more reliable do not seem to be available, although lysimeter tests at Savannah River could provide improved data within the next 10 years or so. There are many factors that could affect the leach rate including the aridity of the area with consequent effect on soil moisture, the presence of chemicals in the wastes that could enhance leaching, or the production of agents during the decay of the organic material that could enhance leaching. The data from Maxey Flats was from an area with reasonable rainfall and the wastes arose from a number of places so that it should be representative of a reasonably maximum area. However, because of the uncertainties in the data, we will consider our maximum case to be a factor of ten higher than the value used. We believe that a case could be made for a very low leach rate in arid regions with low soil moisture but will consider the lower limit to be a factor of ten below the leach rate used.

22. The movement of water in the aquifer will define the direction in which potential exposures may occur as well as the time of movement to a given spot and the concentration. Here there is a trade-off because speeds lower than

the chosen value will result in slower movement but higher concentrations due to the lack of dilution by incoming water. Because of the wide variations possible at given sites as well as the complicated interactions between concentration and distance traveled, which also affects the probability of use of the aquifer, we will assign no uncertainty.

23. Dilution in the water pumped from a well will depend upon the depth in the aquifer that the casing is perforated. If we assume perforation through the full depth of the aquifer, then any change in aquifer depth will result in a directly proportional change in dilution. At the 10 meter depth chosen a typical well could draw 10-30 gallons per minute.¹⁰⁸ It seems doubtful that a well to be used for domestic and irrigation water could use a much thinner aquifer although this would be more than adequate for domestic use. The total thickness of an aquifer (or a series of aquifers) can exceed that chosen for calculation by an order of magnitude although it is possible that the well may not penetrate the full depth if the maximum flow is not needed. We will consider a range of effective depths from 50 meters to 5 meters as appropriate.

24. The effect of the porosity in the aquifer sediments is to allow more or less room for water and to increase the quantity flowing across a given cross-section of the aquifer. The effect of porosity on the concentration for a range of porosity of 0.1-0.3 is given in Table XXII. We will use the same range in this uncertainty estimate.

25. The daily water intake of people will vary depending upon their size, metabolism and consumption of other fluids. ICRP Report 23⁶⁹ gives the range in consumption of tap water for adults under normal conditions as 45-730 cm³/day. People will vary depending upon their size, metabolism and consumption of other fluids. ICRP Report 23⁶⁹ gives the range in consumption of tap water for adults under normal conditions as 45-730 cm³/day and for water-based drinks, such as cider, soft drinks, beer and wine as well as tea and coffee the intake is 320-1450 cm³/day. The total intake for this group is 1000-2400 cm³/day. For adults in high environmental temperatures the intake is 2840-3410 cm³/day and moderately active adults drink a total of 3700 cm³/day. (The totals include milk, which is included in the food pathway.) From this we will consider a

range of 365-3000 cm^3/day for intake of water derived from the well in all forms.

26. In the irrigation pathway the erosion rate of the irrigated field becomes of importance because it determines the equilibrium level of the nuclide in the soil. The use of 6 tons per acre per year leads to a removal of 0.3% of the nuclide per year. The erosion rate depends strongly on the method of cultivation, the type of crop, and the slope of the field. From Table XVII we will consider 0.1 tons per acre per year as a reasonable minimum and 20 tons per acre per year as a representative maximum rate.

Table XII provides estimates of ranges that are indicative of the major uncertainties in each of the parameters used for calculation. There are three classes of this uncertainty; 1) the either-or class which is exemplified by the choice of Quality Factor, dose distribution factor in the bone or the class of an aerosol to use in the inhalation calculation; 2) the distribution class dependent upon the size or actions of an individual such as the inhalation rate or the dust concentration used for resuspension; and 3) the very uncertain class where data are unavailable to fully evaluate the distribution and assumptions are used, such as the leach rate from the waste mass or the relative deposition in each of the compartments of the pulmonary region with subsequent movement to the bone. To these uncertainties, then, are added the uncertainties, due to the generic nature of the study, in the environmental conditions, now and in the future, of the area in which the burial ground is placed and the unknown possible uses to which the area may be placed by future generations.

It is impossible to estimate an overall influence on the final limits of these uncertainties because of the unknown distribution of the values between the limits given. In some cases there are correlations that must be accounted for, such as between organ weight and inhalation or ingestion rate. An extreme range could be obtained by multiplication of the values but this produces an extremely unusual individual such as one with bone weight of 8000 grams who inhales 10 m^3 per day of air with $20 \text{ } \mu\text{g}/\text{m}^3$ of $10 \text{ } \mu\text{m}$ particles and 9×10^{-6} of the contained plutonium is transferred to the bone. Thus, until adequate data of the distribution of many of these parameters is obtained, a combination into an overall distribution of doses in a population will be in itself uncertain.

G. Possible Developments

During the extensive discussions of the Task Group on this problem, a number of items were brought forth which could be investigated as improvements to the present system or as possible alternatives. These are given in Appendix L.

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APPENDIX A
UNITED STATES
ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION
WASHINGTON, D.C. 20545

SEP 29 1975

J. L. Liverman, Assistant Administrator for Environment and Safety

SPECIAL HANDLING REQUIREMENTS FOR TRANSURANIUM-CONTAMINATED SOLID WASTES

As you know, in 1970, ERDA decided that some special handling technique was warranted for long-term disposition of transuranic-contaminated (TRU) wastes. As our initial step, wastes containing more than 10 nanocuries of transuranium nuclides (primarily plutonium) per gram of packaged solid waste were to be put into retrievable storage pending future decisions regarding their handling and disposition. This arbitrary "cutoff" level was justified on the assumption that wastes below this 10 nanocurie level were "safe" for burial in controlled ERDA burial sites, based on a comparison between the 10 nanocuries per gram and the level of radium-226 in naturally occurring uranium deposits. At that time, it was also felt that TRU wastes at significantly higher contamination levels might also prove acceptable for land burial based on a careful evaluation of both waste and site characteristics.

The NRC subsequently published, in September 1974, a proposed rulemaking requiring that transuranium-contaminated wastes from licensed facilities would need to be sent to ERDA for ultimate disposition, with the commercial sector bearing all related costs. In effect, the rulemaking established 10 nanocuries per gram as the upper limit of wastes that could continue to be shipped to commercial (licensed) burial grounds. This proposed rulemaking had many repercussions. Three of the six states responsible for the long-term control of the commercial burial grounds have unilaterally precluded commercial burial ground operators from receiving any additional TRU wastes above the 10 nanocuries per gram level. Many commercial people voiced strong objections to having this arbitrary level imposed in a way which was not intended by ERDA in its initial decision. This point was stressed by many companies commenting on the Draft Environmental Statement, WASH-1539, "Management of Commercial High-Level and Transuranium-Contaminated Radioactive Wastes."

The enclosed paper by Walton A. Rodger, "Critical Evaluation of the Limit of Transuranic Contamination of Low Level Waste," was presented at the 80th AIChE meeting in Boston, Massachusetts, September 8, 1975. It is indicative of the interest and, I believe, the reaction of the commercial nuclear industry to the 10 nanocurie per gram number.

J. L. Liverman

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While not judging the particular approach or rationale taken by Dr. Rodger, I believe it is timely for NEPA to take a positive step toward quantifying the health hazard related to THU wastes. I therefore request that you establish an official NEPA health and safety viewpoint identifying those THU wastes which should not be disposed of by simple land burial and, therefore, would require long-term control such as afforded by a Federal repository. I realize that this is no simple task, and will require substantial time and effort. Since WASH-1539 has been withdrawn and the replacement Environmental Statement will not be issued for at least one to two years, this seems an appropriate period within which to accomplish this task.

I will be pleased to support whatever steps you feel appropriate in accomplishing this goal, since it impacts on decisions of importance to the nuclear energy program. Please let me know of the time schedule you feel would be required and how the ANE staff can assist in this effort.

Richard H. Roberts
Assistant Administrator
for Nuclear Energy

Enclosure:
Paper by Walton A. Rodger

cc w/encl:
F. P. Baranowski
W. R. Voigt, Jr.

APPENDIX B

DOE WASTES AND BURIAL SITES

J. W. Healy and J. C. Rodgers

I. INTRODUCTION

The sources discussed in the present study of the solid wastes contaminated with transuranium elements, are all DOE contractor operations with the burial sites also associated with DOE facilities. As a consequence, the waste characteristics, the relative abundance of the individual nuclides, and the existing burial site characteristics and burial practices may be somewhat different from their counterparts in the commercial nuclear fuel cycle. In the following sections these aspects of DOE waste and waste management practices will be briefly reviewed.

II. TRU WASTE CHARACTERISTICS

The TRU wastes from DOE facilities are widely varied in shape, form, and material with only one thing in common: they are, or are suspected to be, contaminated with transuranium elements. As with other radioactive, solid waste, they may consist of any item or piece of equipment taken into a building where transuranium elements are handled and, eventually, will include all or a portion of the building itself. As a reminder of the complexity of the items to be handled in any waste system, Table B-I provides a partial listing of some of the items and materials that may be encountered.

Combustible materials occur in the classifications of routine combustible, other operational wastes, and in facility decommissioning, although most attention on possible treatment and volume reduction has been focused on the routine laboratory wastes. It is anticipated that systems for incinerating, or otherwise oxidizing, many of these materials will become available, thereby eliminating these items as identifiable wastes and substituting a more uniform ash. However, until such equipment is available and operational on a routine basis, these combustibles will continue to be buried.

Reduction to ash will aid in another problem found almost entirely with such items as cheesecloth, rags, and similar materials used in a laboratory for cleaning. This problem is the inclusion of stray reagents and decontaminating

TABLE B-1
TYPICAL TYPES OF WASTE MATRICES

Routine Waste

Combustible

Cellulose - paper, cheesecloth, cloth
 Rubber - surgeon's gloves, hood gloves
 Plastic - polyvinyl chloride, polyethylene, polypropylene
 (sheet and reagent bottles)
 Oils and greases - cutting oils solidified
 Graphite - molds; crucibles; cores, scarfings

Non-combustible

Glass - Broken or contaminated laboratory glassware;
 tubing; bottles
 Ceramics - magnesium oxide liners and other refractories
 Insulation - asbestos board; asbestos pipe insulation
 Metals - aluminum foil, steel ("tin") cans, scrap pipe
 (stainless steel, steel, aluminum), small tools,
 tubing, small equipment
 Sludge - iron sludge precipitates from the processing of
 liquid wastes
 Filters - HEPA

Other Operational Wastes

Building repair and revision - wood, partitions, benches, cement
 block, insulation, floor tile, electrical wiring, metal
 dustwork.

Equipment (obsolete or broken) - glove boxes, lathes, drill presses,
 vacuum pumps.

Wastes from Facility Decommissioning

Wood, concrete block, chunks of concrete, insulation, dustwork, glove
 boxes, metal working equipment, wiring, windows, fixtures.

agents used with these items. Decontamination reagents, including complexing agents, are used on equipment and people, with the rags and other items discarded in the wastes. Significant quantities of such materials in the wastes could result in mobilization of the transuranium elements after burial.

The non-routine and decommissioning wastes result largely from discarding of contaminated, broken or obsolete equipment, building renovation or decontamination, or elimination of facilities no longer required. These wastes are frequently very bulky and the practice in the past has been to provide packages adequate to transport to burial or retrieval with no attempt to reduce

volume by compacting to eliminate void space. Examples of this are plenum chambers or air ducts from ventilation systems or obsolete glove boxes. Handling of these items poses a problem because they will be difficult to get into a repository, but attempts to reduce their bulk will pose the risk of excessive exposure to people unless special facilities are available. The void spaces are also undesirable in the burial ground since they may lead to cracking of the ground cover when they collapse, leading to possible increased intrusion of water.

Packaging of wastes for burial differs at the various locations depending primarily upon whether the site has a burial ground or the wastes must be shipped elsewhere. For those sites with burial grounds, the packaging is minimal, usually sufficient to permit transport to the burial grounds without spread of contamination. For routine wastes the packaging may consist of a sealed plastic bag inside of a cardboard container with the seams taped shut. Larger items may be encased in plastic sheeting or a wooden box. For those sites where the wastes must be shipped elsewhere for burial, the shipping packages must meet the Department of Transportation regulations. Normal laboratory wastes are usually shipped in a steel drum, frequently 55 gallons (210 liters). For large items plywood boxes have been used.

An estimate of the total waste volumes for all DOE sites is given as $33.8 \times 10^4 \text{ m}^3$ in retrievable storage and $1.4 \times 10^6 \text{ m}^3$ buried. The total plutonium content of these wastes is estimated as 1 000 kg. In the future, about $3 \times 10^4 \text{ m}^3/\text{yr}$ will be buried and about $8 \times 10^3 \text{ m}^3/\text{yr}$ stored retrievable. The burial and storage will involve some $70 \text{ kg}/\text{yr}^1$ of plutonium. The buried wastes include other than those contaminated with fission products, while the stored wastes are all TRU contaminated.

It is important to note that not all of the wastes are contaminated with transuranic elements. In effect any item taken into an area where transuranics are handled is considered to be contaminated and is discarded in the contaminated waste. Thus, if a maximum limit for burial is established, a major fraction of the materials buried will be considerably lower in contamination with some having little or no contamination. For example, Dieckhoner¹ notes that, on the average, greater than 97% of the material buried at the major DOE sites is only slightly radioactive or is suspected of being radioactive because of where it is generated.

III. DOE BURIAL SITES

Since the present proposed limit on concentration of transuranics in wastes suitable for shallow earth burial is meant to apply to DOE waste disposal sites, it is appropriate to review briefly the major site physical characteristics, and their respective burial practices. Summary information from several sources^{2,3,4} is shown in Table B-II and Table B-III and discussed in more detail in the following sections.

A. Location and Site Descriptions

1. LASL: The Los Alamos Scientific Laboratory (LASL) is located in Los Alamos County, New Mexico. It is situated on the Pajarito Plateau at an elevation of about 7 000 feet and solid waste burial is carried out on the mesas of the plateau. These mesas are covered by a weathered tuff and underlain by 70-180 meters (240 to 590 feet) of volcanic tuff. The tuff is irregularly broken by nearly vertical joints which may be closed or open as much as 5 cm, although they are usually less than 5 mm wide. The upper meter of these joints is usually filled with weathered tuff. The site has a semi-arid, continental, mountain climate with limited precipitation (about 400 mm/yr). There is little or no recharge to the zone of saturation from precipitation on the plateau.

Where the soil cover has been disturbed in the disposal areas, precipitation may infiltrate to depths of three meters, but below this depth there is not enough water to leach the contaminants. The burial site mesas are bounded on the north and south by steep walled canyons cut 15 to 30 meters below the surface of the mesas by intermittent streams. It has been estimated⁵ that downward erosional processes would require 27 000 years to expose wastes at ground level, and 110 000 years for lateral erosion from the canyon walls to reach the wastes. If radioactive contaminants reach the canyons, storm runoff events would be the principal potential transport vector.

2. INEL: The Idaho National Engineering Laboratory (INEL) is located in southeastern Idaho near (20 miles) Idaho Falls, Idaho. The site is on the Columbia Plateau in the eastern part of the Snake River Plain, lying within a closed topographic depression. Soil cover is thin, (1.5 to 6 meters), with surface basaltic rock over large areas. The top of the zone of saturation beneath the solid waste burial ground is at a depth of about 200 meters.

TABLE D-II

DOE BURIAL SITE CHARACTERISTICS

Type of Information	Savannah River Plant S. Carolina	Oak Ridge National Laboratory, Tennessee	Los Alamos Scientific Laboratory, N. Mexico	Idaho Nuclear Engineering Laboratory	Hanford Washington
Mean Annual Precipitation (mm)	1 200	1 300	400	200	200
Surficial Material Type	Clayey sand	Weathered shale and fill	Weathered tuff	Alluvial sand and gravel	Clay, sand, and gravel
Thickness (meters)	300	0-10	0-2	1-10	over 100
Interstitial Perme- ability to water (cm/day) ^a	10-100	very low	moderate (1.0-10.0)	moderate	variable
Bedrock Material Type	Gneiss	Shale	Volcanic tuff	Basalt	Volcanics
Structure	eroded peneplain	Folded	Flat-lying	Flat-lying	Flat-lying
Ground Water					
Depth to Shallowest Saturated Zone (meters)	10-20	0-5	200-300	60-300	100
Depth of Continuous Groundwater Zone (meters)	10-20	2-5	200-400	60-300	100
Depth to Regional Aquifer (meters)	120	None present	200-400	60-300	100
Total thickness of ground- water zone (meters)	10-200	120	>1 000	300-3 000	unconfined 70
Surface Water Proximity	on site	on site	1 kilometer	3 kilometers	10 kilometers
Flow characteristics	Small, ephemeral	small, perennial	small, ephemeral	small, ephemeral	large, perennial (Columbia River)
Cation exchange Capacity of Material Surrounding Burial ^a meq/100 gm	1-15	2-20	0.5-3.0	11	4-20
Principal Flow away from Burial	Pore spaces in clayey sand	Shale fractures and Pores in fill	Fractures and pores in tuff	pores in sand	pores in sand
Horizontal velocity of flow of ground- water (m/d)	<0.3	?	0.3	3	0.01

^a Interstitial permeability and absorptive capacity can be bypassed by flow along fractures or other high permeability zones.

TABLE B-III

BOX WASTE SERIAL GROUND OPERATIONS

Site	Savannah River Plant, SC	Oak Ridge National Laboratory, TN	Los Alamos Scientific Laboratory, NM	Sanford, WI	Idaho National Engineering Laboratory, ID
OPERATION Serial Trench/Pit Size (meters)	6 wide x 6 deep x variable length	3 wide x 15 long x 3-4.5 deep	8-30 wide x 130-180 long x 6 deep	1.5-5 wide (Bottom width) x 4-6 deep x variable length	Trench: 3-5 wide x 275 long x 4 deep Pit: 30 wide x 4 deep variable length
Provisions for Water Collection and Containment	None; trenches have monitoring wells	Trenches sloped to one end; 19 on metal casing as monitoring well	None	None	None
Waste Covering Frequency	Covered after disposal for fire, contamination, radiation control	When trench is filled	Combustibles covered day of delivery; Others as required for contamination control and layering	Daily after deliveries	As trench/pit is filled
Type of Final Cover	Excavated fill to ground surface; Mounded as necessary	Excavated material; to ground surface; Few experimentally sealed 0.5 below surface; recessed.	Excavated buff fill with compaction by heavy earth moving equipment	Excavated fill to surface; Mounding as necessary	Excavated soil fill; recessed
Depth of Final Cover	Minimum 1.2 m cover, or that needed to reduce dose to <1 mSv/hr at surface	Minimum 1 m to ground surface	Minimum 1.5 m total excavated buff cover with mounding to 0.5-1 m above grade	Minimum 2.5 m total, or that needed to reduce dose to <1 mSv/hr at surface	Minimum 1 m to ground surface
Other		Minimum 1.5 m between trenches	Minimum 4.5 between pits at surface; Minimum 15 on crushed, compacted buff in pit bottom prior to waste fill		Minimum 0.5 m soil in pit/trench bottom to underlie waste

Precipitation is limited (200 mm per year), but there is some evidence of precipitation infiltration to depth at the burial grounds because the sediments are at field capacity.⁴

3. THE HANFORD MONITOR: The Atlantic-Richfield Hanford Company site is located in the southeastern corner of the State of Washington, 30 miles east of Yakima. The area is on the Columbia Plateau of the Pacific Basin, bounded on the north and east by the valley of the Columbia River. Unconsolidated silts, sands, and gravels up to 600 meters thick make up the surficial material, and below that are other layers of clays, conglomerates, and sands up to 370 meters thick. The climate is mild and dry; the annual precipitation is only about 200 mm. Ground water in the surficial sediments occurs under unconfined conditions, although locally confined zones do exist. Water in the basalt bedrock occurs mainly under confined conditions. The major source of natural recharge to the unconfined aquifers is precipitation on the uplands to the south and west of the reservation, the effects of precipitation on the lowlands being unmeasurable due to high evaporation losses. The depth from the surface to the water table varies greatly from place to place depending chiefly on local topography ranging from less than one to over 100 meters.

4. ORNL: The Oak Ridge National Laboratory (ORNL) is located in the west central portion of eastern Tennessee (15 miles west of Knoxville, Tennessee). The site is placed in a ridge and valley setting characterized by southwest-northwest oriented ridges of sandstone, shale, and dolomite, separated by valleys of less weather resistant limestone and shale. Annual precipitation is about 1 300 mm. Surface drainage is controlled by topography, with many small streams feeding into the Clinch River. Solid waste burial is being carried out in the Conasauga shale group, consisting of weathered and weakened shale three to six meters deep, overlying shales with siltstone beds and some lenses of grey limestone in the upper few hundred feet of the formation. The elevation of the zone of saturation is usually between 1.5 and 5 meters below the surface. It is assumed that all of the water that moves beneath or infiltrates into the burial ground discharges into a nearby stream valley, but the base of this local flow system has not been defined.⁵ Leachate springs have been found downslope from certain of the burial sites indicating that groundwater and infiltrating surface runoff enter the trenches and then seep

through the refuse to spill over and emerge at the surface, because the permeability of the subsurface materials is not adequate to allow all of this water to move out in the subsurface.

5. SRP: The Savannah River Plant (SRP) is situated in the Atlantic Coastal Plain region of South Carolina about 25 miles southeast of Augusta, Georgia. Almost all of the site is drained by tributaries of the Savannah River and no part is very far from a continuously flowing stream. Average annual rainfall is about 1200 mm. The burial site is located between two tributaries of the Savannah River. No flowing streams cross the area; surface runoff is controlled by drainage ditches. Underlying sediments consist of sandy clays and clay beds, with a depth to bedrock of about 300 meters. The average depth to the water table is 12 meters. Infiltration into the burial trenches moves downward to the zone of saturation and then laterally via the local flow system to the neighboring creeks. The base of this flow system has not been defined, and some infiltration may move into a larger flow system. Transuranium elements have not been detected more than three meters below the bottom of the trenches.

B. Site and Burial Practices

1. LASL: The primary disposal area used at LASL since 1959 is Burial Ground G (there are at least four waste disposal sites no longer used). The burial area contains six large pits, 30 meters wide, 200 meters long, and about 10 meters deep, eight smaller pits of various dimensions, and 80 disposal shafts of various depths (maximum 20 meters).³ As standard procedure, low-level radioactive wastes are placed in 30-liter metal trash cans, lined with plastic or in cardboard boxes lined with plastic prior to disposal. Wastes are buried in two meter thick layers and covered with a maximum of 0.6 meters of tuff. Pits are filled to within one meter of the land surface and then filled with tuff and mounded to enhance surface runoff. The estimated content of transuranium elements in the waste material placed in the disposal pits and shafts of Burial Ground G are: $^{238}\text{Pu} = 15 \text{ Ci}$; $^{239}\text{Pu} = 332 \text{ Ci}$; $^{241}\text{Am} = 2074 \text{ Ci}$.³ There was an estimated 5 kg of ^{239}Pu accumulated in buried wastes prior to the time retrievable storage procedures were instituted. Presently treated liquid streams generate about 150 grams of plutonium per year going to retrievable storage.

2. INEL: About 6 000 m³ of TRU contaminated solid waste are stored annually at INEL from the Rocky Flats operation. This material was formerly buried in pits, but wastes above the 10 nCi/gm level are now stored on asphalt pads under sheet plastic and plywood with a compacted earthen cover. The waste is contained in plastic-lined 210 liter drums or special treated boxes. The accumulated plutonium in the burial site now amounts to about 500 kg (120 kg in retrievable storage). Annual generation of waste plutonium from Rocky Flats is about 20 kg. Test exhumation work is currently being done on certain previously buried TRU wastes. Disposable level wastes are buried in trenches or pits. The trenches are generally 1 to 8 meters in width and 100 to 550 meters long. Pits are 150 by 370 meters and 1.5 to 5 meters deep. The trenches are backfilled and compressed. It is estimated that the primary burial ground has had disposed in it about 17 000 Ci of ²³⁹Pu, and 26 000 Ci of ²⁴¹Am.³

3. HANFORD WORKS: From 1966 to 1974, a program of centralization of the burial grounds in the 200 area was started and completed. There are now seven major burial grounds (three are active). Most wastes have been placed in long trenches: dry wastes in trenches 4 meters deep, 9 meters wide on 15 meter centers; contaminated industrial solid waste has been placed in larger trenches. A minimum of 1.2 meters of earth covers each trench. For the past year or so (1974-1976) TRU waste above 10 nCi/g have been stored for possible retrieval in 210 liter steel drums stacked on asphalt pads constructed in shallow trenches. When a section of a TRU trench is complete, the stack is covered with a large vinyl sheet and covered with several feet of backfill. Estimated total plutonium in each burial ground ranges from 1 to 4 kg.

4. ORNL: Three solid waste storage areas were developed, filled, and closed during the period 1944-1951. Trenches were dug, filled with wastes, and backfilled. There is little information about the volume or character of materials buried. A fourth site was opened in 1951 and closed in 1959. It is in a poorly surface-drained landform. The fill is not only a catchment for precipitation but must receive runoff from the hillslope and drainageways and groundwater from upslope. Most of the landfill is saturated. A fifth site was developed as a result of previous difficulties, with much more care in site selection. Wastes are placed either in trenches 15 to 120 meters in length, 2.5 to 9 meters in width, 2.5 to 4 meters in depth) or in auger holes (0.6 meter

diameter, 4.5 meters deep). Much of the record of the early waste disposal operation was lost in a fire.

5. SER: The original burial ground of 76 acres began to receive waste in 1953 and was filled in 1972. A 119 acre site contiguous to the original area was then opened to receive waste.

Beginning in 1965 TRU waste was segregated into retrievable and non-retrievable waste, according to activity level. Non-retrievable waste was disposed of in trenches approximately 6 meters deep and backfilled with a minimum 1.5 meter soil cover. Between 1965 and 1974 retrievable stored waste was buried in concrete containers or encapsulated in concrete. After 1974 retrievable waste was stored in 210 liter drums on a concrete pad and, according to the alpha activity, the drums may be placed in concrete culverts. The pad with the drums and culverts is protected from rain percolation and mounded with a minimum 4-foot soil cover.

About 11 300 m³ of TRU waste were buried before 1965 and about an equal volume since then. The major TRU's buried or stored include approximately 300 000 Ci of ²³⁸Pu, 2 000 Ci of ²³⁹Pu, and 40 000 Ci of ²⁴¹Cm.

IV. ISOTOPES AND DECAY CHARACTERISTICS

The majority of the TRU wastes from DOE operations result from weapons work and from use of heat source, or high purity, ²³⁸Pu. Some research and development work is done with high burnup plutonium resulting from reactor fuels. In addition, some quantities of individual transplutonic elements, such as ²⁴¹Am, are handled as separated sources.

The plutonium isotopic composition will vary depending upon the exact source. The values given in Table B-IV are typical of these materials.

In contrast to some of the transuranic wastes produced from commercial facilities, there are low levels of beta emitters (either fission products or activation products) and few of the heavier isotopes with the notable exception of ²⁴¹Am, which arises from the decay of ²⁴¹Pu. This is because the wastes, in general, arise from the use of plutonium that is already separated from the many other nuclides produced during the irradiation of the fertile material. The transuranics not recovered during the initial separation are largely in the high level wastes which will go to a repository.

TABLE B-IV
TYPICAL PLUTONIUM COMPOSITIONS

	g by weight		
	Weapons	Heat Source	High Burnup
^{236}Pu	-	10^{-4}	
^{238}Pu	0.012	80.3	2.4
^{239}Pu	93.6	15.9	47.0
^{240}Pu	5.9	3.0	26.0
^{241}Pu	0.4	0.7	15.5
^{242}Pu	0.013	-	9.0
^{244}Pu	0.02		

^aMixed oxide fuels used as representative from reference 2.

The primary transuranic found in the waste is not the only possible potential source of exposure for the long time period, since each is a parent of a decay series eventually ending in a stable isotope of lead or, in one case, bismuth. The decay of each of the parents and quantities of daughters at various times following purification of one curie of the parent are given in Figs. B-1 through B-7. The decay characteristics of the four series involved are given in Tables B-V through B-VIII.⁶

There are several features of these decay chains which may be of importance.

1. Three of the four chains eventually decay to those found in nature. These are the uranium chain (^{238}Pu and ^{242}Pu), the thorium chain (^{244}Pu , ^{240}Pu), and the actinium chain (^{239}Pu). The other chain (^{233}U , ^{241}Pu) has a precursor, ^{237}Np , with a half-life short enough so that any formed at the birth of the universe has decayed.

Fig. B-1. ^{236}Pu and Daughters.

Fig. B-2. ^{238}Pu and Daughters.

TABLE D-XVI
DISTRIBUTION OF ^{238}Pu IN RATS GAVAGED WITH A NITRATE
SOLUTION AT 2 DAYS OF AGE FROM 5 TO 11 RATS

Age at Necropsy, Days	Dose, μCi/kg	Content as Percent of Gavage Dose				
		Skeleton	Liver	Intestine		Total Retained ^a
				Wall	Contents	
3	180	1.1	0.16	58	28	1.2
10	180	1.56	0.21	0.93	1.7	1.8
20	180	1.66	0.24	0.03	0.01	1.7
140	280	0.53	0.027			0.56
140	110	0.65	0.019			0.67

^aEntire carcass, excluding GI tract and content.

The authors indicate that the sum of the skeletal and liver retention is probably the best estimate of total excretion because it is believed, from parenteral administration, that the urine values are high, presumably due to fecal contamination. From these data it is noted that the approximately 1% absorption of neptunium is markedly higher than the other actinides which fall in a relatively narrow range of about 0.005% to 0.05%. The aged curium oxide source appeared to have an absorption about ten times the freshly prepared material which is consistent with other observations of increased mobility of high specific activity oxides.

The distribution for the newborn rats is shown in Table D-XIX. It is noted that absorption in the new born is increased over that in the adult by factors of 50 to 500 except that the increase for neptunium is only about a factor of 6. However, this may be due to the fact that the rats used in this administration were 8 days old. It was also noted that the newborns did not discriminate between the fresh and aged curium oxide suggesting that the mechanism of absorption in the newborn may be different from that of the adult. Of particular interest is the high retention of actinides in the newborn animals in

Fig. B-3. ^{239}Pu and Daughters.

Fig. 4. ^{240}Pu and Daughters.

Fig. B-5. ^{241}Pu and Daughters.

Fig. B-6. ^{242}Pu and Daughters.

Fig. B-7. ^{241}Am and Daughters.

2. Plutonium- ^{241}Pu , a beta emitter, decays to produce two man-made elements, ^{241}Am and ^{237}Np , which are alpha emitters. In the relatively short term ^{241}Am is important, reaching a maximum level of about 0.030 Ci per Ci of parent in about 70 years. In the longer time period, ^{237}Np peaks at about 10^3 - 10^4 years and remains fairly constant at about 5 μCi per Ci of parent to about 10^6 years. In the interval of about 100 to 10,000 years, both isotopes will be present. These two elements are of interest because they do not occur naturally, and the amount of information on their behavior in the environment is small, although what is available indicates they may be taken up in plants to a higher degree than plutonium. The problem is amplified, from the practical standpoint, since normal measurements of plutonium by alpha emissions will not detect the ^{241}Pu parent and the sensitivity of measurement of the low energy beta in the presence of the alpha particles from the other isotopes of plutonium is poor. (The current detection limit of the alpha emitters in environmental samples is on the order of 0.01-0.02 pCi per sample. To measure the eventual ^{241}Am buildup to the same level would require a sensitivity of 0.4-0.8 pCi.)

TABLE B-V

URANIUM CHAIN WITH PLUTONIUM PRECURSORS

Isotopes	$T_{1/2}$	Daughter	Principal Radiations (MeV) ^a		
			Alpha ^b	Beta ^c	Gamma ^c
²⁴² Pu	3.87×10^5 y	²³⁸ U	4.89	-	-
²³⁸ U	4.468×10^9	²³⁴ Th	4.20	-	-
²³⁴ Th	24.10d	²³⁴ Pa	-	0.044	0.008
²³⁴ Pa	1.17m	²³⁴ U	-	0.82	0.011
²³⁸ Pu	87.75y	²³⁴ U	5.49	-	-
²³⁴ U	2.445×10^5 y	²³⁰ Th	4.76	-	-
²³⁰ Th	7.7×10^4 y	²²⁶ Ra	4.66	-	-
²²⁶ Ra	1600y	²²² Rn	4.77	-	0.006
²²² Rn	3.8235d	²¹⁸ Po	5.49	-	-
²¹⁸ Po	3.05m	²¹⁴ Pb	6.00	-	-
²¹⁴ Pb	26.8m	²¹⁴ Bi	-	0.22	0.23
²¹⁴ Bi	19.9m	²¹⁴ Po	-	0.63	2.03
²¹⁴ Po	1.64×10^{-4} s	²¹⁰ Pb	7.69	-	-
²¹⁰ Pb	22.3y	²¹⁰ Bi	-	0.007	0.002
²¹⁰ Bi	5.012d	²¹⁰ Po	-	0.39	-
²¹⁰ Po	138.378d	²⁰⁶ Pb	5.31	-	-
(Stable)					

^aBranches <1% not included.^bAverage energy.^cTotal energy emitted per disintegration.

TABLE B-VI

NEPTUNIUM CHAIN WITH PRECURSORS

Isotopes	$T_{1/2}$	Daughter	Principal Radiations (MeV) ^a		
			Alpha ^b	Beta ^c	Gamma ^c
²⁴¹ Pu	14.4y	²⁴¹ Am	-	0.005	-
²⁴¹ Am	433y	²³⁷ Np	5.48	-	0.022
²³⁷ Np	2.14×10^6 y	²³³ Pa	4.69	-	0.021
²³³ Pa	27.0d	²³³ U	-	0.068	0.155
²³³ U	1.585×10^5 d	²²⁹ Th	4.81	-	-
²²⁹ Th	7.34×10^3 y	²²⁵ Ra	4.87	-	0.040
²²⁵ Ra	14.8d	²²⁵ Ac	-	0.094	0.012
²²⁵ Ac	10.0d	²²¹ Fr	5.75	-	0.012
²²¹ Fr	4.8m	²¹⁷ At	6.35	-	0.028
²¹⁷ At	0.0323s	²¹³ Bi	7.07	-	-
²¹³ Bi	46.65m	²⁰⁹ Tl	5.85(2.16%)	0.019	-
		²¹³ Po	- (97.84%)	0.432	0.132
²⁰⁹ Tl	2.20m	²⁰⁹ Pb	-	0.658	2.10
²¹³ Po	4.2×10^{-6} s	²⁰⁹ Pb	8.38	-	-
²⁰⁹ Pb	3.31h	²⁰⁹ Bi	-	0.198	-
		(Stable)			

^aBranches <1% not included.^bAverage energy.^cTotal energy emitted per disintegration.

mobility of fallout particles cannot be made as yet. Even if plutonium or other TRU leaves a burial trench initially as a soluble species, later interactions with soil materials can result in a particulate form which is as mobile or even more mobile than the soluble one. The most obvious case is sorption on very fine clay particles. But in addition, it has been found^{17,22} that under appropriate conditions, plutonium, on hydrolyzing or polymerizing, behaves as very small discrete particles of an amorphous nature. Upon extensive heating or aging, the polymers may form into small crystallites. The presence of silica apparently affects the distribution of particle size.¹⁸ Particulate TRU transport phenomena has been compared with the environmental transport of natural thorium.¹⁹ If this analogy is correct, one would expect that leaching and transport as a soluble species should be insignificant compared to transport of particulate forms.

Mechanism 2: Mobilization of TRU by complexing agents. The physical and chemical forms of TRU in solid wastes, both initially and over time, can be expected to be extremely varied, ranging from the most insoluble forms, such as PuO_2 , to highly soluble forms resulting from interactions with decontamination chemicals in the wastes and a vast array of biochemicals produced in the composting wastes.¹³ A key consideration in the analysis of the chemical behavior of TRU in the waste pits and in the environment (its biogeochemistry) is oxidation state. It is oxidation state rather than element (Z-number) which governs chemical behavior.²⁰ The most stable oxidation states of the transuranium elements are probably Np(V) , Pu(IV) , Am(III) , and Cm(III) ,²⁰ although in solutions Np , Pu , and Am can exist in at least 4 oxidation states (III, IV, V, and VI). Of these only Np and Pu are likely to exist in more than one state environmentally. The acidity of the solution and the presence of complexing ligands (organic or inorganic), however, significantly affect the relative importance of each of the various oxidation states. For example, complexation and hydrolysis shift the oxidation potential of the $\text{Pu(III)} - \text{Pu(IV)}$ couple towards positive values favoring the oxidation of Pu(III) .¹⁹ Under acidic environmental conditions, the ferric-ferrous system characterizing the environmental chemistry of iron might be a suitable model for the reduction of Pu(IV) to Pu(III) , but the reduction is not likely in alkaline, aerobic environments due to the stability of hydrolyzed or complexed forms of tetravalent plutonium. Like Np(V) and U(VI) , the higher oxidation states of

several possible mechanisms including, but not limited to: a) release of TRU into infiltrating water by solubilization or suspension of TRU which is already in suitable chemical or physical form, b) direct microbial and fungal action on TRU contaminant in biodegradable waste rendering it transportable in leachate; c) chemical interactions between TRU contaminants and organic and inorganic reactive compounds produced during decomposition of organic wastes; or d) radiolysis enhanced dissolution of refractory forms. The geochemical character of the soils in which burial is made is an important boundary condition of these processes. Each of these types of mechanisms will be considered in more detail in the following paragraphs; in reality, of course, probably all or some of these will be simultaneously affecting TRU mobility at a site.

Mechanism 1: Physical transport (particle or colloid). Physical transport of TRU in soil can occur by a variety of mechanisms including, bio-redistribution caused by feeding soil organisms, such as worms or microflora; burrowing by small animals; the cycles of growth and decay of plant roots; and translocation of TRU by soil water as small particles or associated with clay fractions of soil minerals. The case of the penetration of very fine particle size fallout plutonium ($0.04 \mu\text{m}$) into loamy soil has been modeled as an example of this form of transport.¹⁴ The result is a projected mean velocity in the range 0.8 - 1 cm/yr for this process. Thus the migration velocity is two orders of magnitude below the expected percolation velocity of infiltrating precipitation in the same loamy soil (100 cm/yr) in a wet climate. Laboratory studies of the transport of radioactivity attached to small kaolin particles and to bacteria show similar retention and transport properties through packed sand columns.¹⁵ It was found that an increase in strength of ionic species in the infiltrating water can cause a large release of radioactively contaminated bacteria or clay from the retaining sand column. Since the retention mechanisms for particles are dependent in part on size, it would be expected that larger size particle fractions would not be as mobile; this effect might explain the limited mobility observed in the studies of the transport of particulate plutonium in the Hanford Z-9 trench.¹⁶ Particulate plutonium in the size range of 2-25 μm was found to have penetrated not much more than 30 cm in the 7 year life of the trench. However, leaching studies will be required to ascertain the migration rates as a function of present and projected environmental conditions,¹⁶ so that complete comparison with environmental processes affecting

surface elevation and increased infiltration rate, thus raising the groundwater table because of the change in topography and permeability.⁷ The resulting saturated conditions increased the transport of ⁹⁰Sr from the buried waste.

Little is known about the non-radioactive constituents in low-level solid wastes containing transuranium elements going into shallow land burial sites. Only approximate values for the radioactive constituent inputs have been recorded for most DOE and commercial sites.^{8,10} Often only general designations such as "mixed fission products" (MFP), "special nuclear material" (SNM), or "low specific activity" (LSA) have been used.⁸ Characteristically these wastes include, in addition to paper products (constituting as much as 70% of the waste volume), such items as packing materials (wood, plastic, or metal), protective clothing, broken glassware, rubber and plastic sheeting, gloves and tubing, carcasses of experimental animals, contaminated equipment, and contaminated building material. A detailed breakdown of one DOE plutonium process facility TRU solid waste categories is available as an example.⁹ With low-density materials such as these going into the pits, it is understandable that compactable void spaces of 30% to 50% of the trench volume could occur,¹¹ and subsequent settling of trench contents and cracking of cap materials as well.

The results of studies of sanitary landfills in northern Illinois provide some perspective on the long-term aging effects on land burial in humid climates. These studies show that: 1) approximately one-half of the yearly precipitation infiltrates the cap; 2) portions of this water, in the form of leachates, leave the disposal site as subsurface or surface (seep) flow; 3) the greatest decrease in effective porosity of waste (~90%) occurs during the first five years of burial, but may continue for 30 years or more; 4) groundwater mounds often form below trenches as a result of infiltration followed by downward movement of water, and surface seeps can occur as a result of such mounds; and 5) decomposition and settling of the wastes can be expected to occur for 30 or more years.¹² Because it is water (leachate) moving under the influence of gravity and osmotic forces through subsurface soils that is the major natural driving force capable of transporting significant quantities of radionuclides away from a burial site under saturated conditions, considerable research is underway on leaching processes and solute transport by water.¹³

III. FACTORS AFFECTING TRANSURANIUM ELEMENT MOBILITY

Transuranium elements may be mobilized from the waste matrix by any of

APPENDIX C

SUBSURFACE MIGRATION OF TRANSURANIUM ELEMENTS FROM BURIED SOLID WASTE

John C. Rodgers

I. INTRODUCTION

The focus of the following review of current information concerning subsurface migration of transuranium radionuclides (TRU) from shallow land burial sites is on mechanisms whereby a mobile species of TRU could be generated either in buried solid waste or surrounding soil and migrate through intervening soil or rock to become an accessible source of TRU in the environs of a solid waste disposal site. It is the biogeochemistry of near-neutral, dilute solutions of TRU in the environments of the deeper horizons of soil and rock that is of special interest here. Migration of plutonium and other TRU's have been reported in a range of media including volcanic tuff,¹ basalt,^{2,3} alluvium,⁴ and desert soils.⁵ The varied mechanisms of transport reported in these laboratory and field studies remain to be fully explained. Pieces of the puzzle are in place, however. These will be the subjects of the following sections.

II. CHARACTERIZING THE SOURCE

There is considerable diversity in site characteristics (climate, geohydrology, soil characteristics, etc.), trench preparation and filling, and final covering at the DOE low-level shallow land burial sites across the country. But no matter how varied the sites may be, burial universally takes place at shallow depths, in the transition zone between soil materials and deeper subsurface geology. Due to a lack of well developed information about this zone, there are often major uncertainties related to subsurface conditions within the burial ground. In the case of one site underlain by aquitards of low permeability and of a fractured nature, and located in a humid climate, it may not be possible at any reasonable cost to obtain quantitative definition of the hydrogeology of the region.⁶ Even if the hydrogeology of a disposal site were reasonably well understood, engineering operations incidental to the disposal of wastes can significantly alter the conditions of the site such that radionuclide migration can increase. In one case, a burial ground area was used for disposal of uncontaminated fill after burial was completed, resulting in increased

3. Although the eventual products of many of the nuclides are those found in nature, they may enter the decay chain at a different isotope than the natural chain resulting in different ratios than are found in nature. For example ^{238}Pu decays to ^{234}U which is a precursor of the radium series. Normally the quantity of the ^{226}Ra in nature is controlled by the half-life of ^{238}U . However, the ^{234}U half-life is about 5×10^{-5} of that of the ^{238}U so that a given weight of this isotope will result in much higher daughter activities than will the natural material.

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TABLE B-VIII

THORIUM CHAIN WITH PLUTONIUM PRECURSORS

Isotope	$T_{1/2}$	Daughter	Principal Radiations (MeV) ^a		
			Alpha ^b	Beta ^c	Gamma ^c
²⁴⁴ Pu	8.26×10^7 y	²⁴⁰ U	4.58	-	-
²⁴⁰ U	14.1h	²⁴⁰ Np	-	0.102	-
²⁴⁰ Np	7.4m	²⁴⁰ Pu	-	0.631	0.328
²⁴⁰ Pu	6537y	²³⁶ U	5.16	-	-
²³⁶ U	2.3415×10^7 y	²³² Th	4.49	-	-
²³² Th	1.405×10^{10} y	²²⁸ Ra	4.00	-	-
²²⁸ Ra	5.75y	²²⁸ Ac	-	0.010	-
²²⁸ Ac	6.13h	²²⁸ Th	0.376	-	0.915
²²⁸ Th	1.9131y	²²⁴ Ra	5.40	-	0.002
²²⁴ Ra	3.66d	²²⁰ Rn	5.67	-	0.009
²²⁰ Rn	55.6s	²¹⁶ Po	6.29	-	-
²¹⁶ Po	0.15s	²¹² Pb	6.78	-	-
²¹² Pb	10.64h	²¹² Bi	-	0.100	0.117
²¹² Bi	60.55m	²⁰⁸ Tl	6.04(35.93%)	-	0.003
		²¹² Po	- (64.07%)	0.717	0.281
²⁰⁸ Tl	3.07m	²⁰⁸ Pb	-	0.561	2.37
²¹² Po	3.05×10^{-7} s	²⁰⁸ Pb	8.78	-	-
(Stable)					

^aBranches <1% not included.^bAverage energy.^cTotal energy emitted per disintegration.

TABLE B-VII

ACTINIUM CHAIN WITH PLUTONIUM PRECURSORS

Isotopes	$T_{1/2}$	Daughter	Principal Radiations (MeV) ^a		
			Alpha ^b	Beta ^c	Gamma ^c
²³⁹ Pu	2.439×10^4 y	²³⁵ U	5.15	-	-
²³⁵ U	7.1×10^8 y	²³¹ Th	4.28	-	0.140
²³¹ Th	25.52h	²³¹ Pa	-	0.078	0.017
²³¹ Pa	3.248×10^4 y	²²⁷ Ac	4.92	-	0.030
²²⁷ Ac	21.773y	²²³ Fr	4.19(1.38%)	-	-
		²²⁷ Th	- (98.62%)	0.010	-
²²⁷ Th	18.718d	²²³ Ra	5.67	-	0.010
²²³ Fr	21.8m	²²³ Ra	-	0.345	0.051
²²³ Ra	11.434d	²¹⁹ Rn	5.70	-	0.087
²¹⁹ Rn	3.96s	²¹⁵ Po	6.81	-	0.054
²¹⁵ Po	1.780×10^{-3} s	²¹¹ Pb	7.39	-	-
²¹¹ Pb	36.1m	²¹¹ Bi	-	0.453	0.050
²¹¹ Bi	2.13m	²⁰⁷ Tl	5.55	-	0.043
²⁰⁷ Tl	4.77m	²⁰⁷ Pb	-	0.493	0.002
		(Stable)			

^aBranches <1% not included.^bAverage energy.^cTotal energy emitted per disintegration.

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plutonium are expected to be more mobile in the environment than the tetravalent states, particularly in the presence of carbonate or bicarbonate ions.²¹ However, the stability of these higher oxidation states, particularly in soils having an abundance of organics has been questioned,¹⁹ on the ground that at near neutral pH the oxidation potentials of both Pu(V) and Pu(VI) relative to tetravalent plutonium tend to favor reduction to Pu(IV), and to preclude formation from the reduced form.

The formation of stable organic complexes of transuranium elements, such as plutonium for which Pu(IV) complexes are thought to be strongest,²³ has been suggested as a possible mode of mobilizing plutonium. This hypothesis has been advanced partly on the basis of analogy with the known ability of humic compounds to enhance solubilities of metal ions such as aluminum, iron, copper, zinc, and lead in the environment,²⁴ and partly by implication in the case of TRU appearing near a burial ground at Oak Ridge.²⁵ In the latter case, gel filtration chromatography was used to demonstrate that organically chelated ⁶⁰Co mobilized in seep water was due to the action of EDTA present in the waste. The effect of EDTA on ⁶⁰Co mobility is illustrated by the fact that while the K_d of ⁶⁰Co in weathered Conasauga shale at pH 6.7 is approximately 10^4 , in the presence of 10^{-7} M EDTA in the seep water, K_d is reduced to the range 7-70.²⁵ Since all the trivalent rare earths and actinides possess at least as high or higher complexity constants for EDTA as Co^{2+} , and since ²³⁸Pu, ²⁴¹Am, and ²⁴⁴Cm were found in the soil of a seep, the evidence suggests, but does not prove, that EDTA is also contributing to the migration of these TRU's as well.²⁵

Contradictory evidence to the hypothesis of increased mobility due to organic complex formation has emerged from several directions, however. In one study of the differences in the mobility of plutonium in a variety of soils using $\text{Pu}(\text{NO}_3)_4$, as determined by water extraction and subsequent membrane filtration using membranes of different pore size, it was found that the most mobile species was most likely particulate $\text{Pu}(\text{OH})_4$ or hydrated oxides, not organic forms complexed by soil humic compounds.²³ A similar finding resulted from a study of the solubilization of plutonium and americium by fulvic acid - the more soluble humic compound. Fulvic acid solutions were added to samples of contaminated soil at pH values selected to encompass the range of conditions to be expected in groundwaters, and extractions were made repeatedly for periods as long as 357 hours.²⁴ The results indicated very little solubilization occurred if fulvate complexes were formed they were unstable, resulting in slow

Reduction to the tetravalent state occurs both in the solutions containing plant roots (due to root exudates), and in the plutonium assimilated by the plants, appearing in the xylem exudate. Although Pu(VI) in solution was apparently assimilated by plants more rapidly than Pu(IV), it was reduced to the less biologically mobile Pu(IV) state in the plant biochemical transformation processes.

In light of present limited understanding of biologically mediated mobilization of TBU in the environment it is difficult to reach a generalized, bottom line estimate of the potential for greatly increased mobilization of these elements in biological systems over very long periods of time. At present, the available evidence indicates little increased mobility in natural systems. For example, plutonium associated with soil organic matter for 30 years in the environment of Oak Ridge was subjected to extraction by solid chelating resin to determine its descriptive properties. It was found that a fraction of the organically bound Pu was readily desorbed (about 8% of the soil Pu) and the remainder was much more slowly desorbed, approaching 1% of the soil Pu after 14 weeks.²⁰ These studies with resins under conditions favoring exhaustive removal indicate that after 30 years a substantial fraction of Pu in soil does not readily desorb. Another example, which also suggests the low biological availability of even organically modified Pu, is the study of the mobility of Pu introduced to a nonflowing, highly eutrophic pond for over 30 years on the Hanford reservation.²⁶ Sediments play a dominant role in the Pu inventory of this ecosystem. They have the highest concentrations of Pu in the pond and contain greater than 99% of the entire inventory of the system. The highly enriched nutrient supply of the pond supports luxuriant growths of algae and macrophytes which form a sedimentary organic floc. Sedimentation is rapid since the pond has no surface outflow and short retention time (40 hours). Mass balance calculations indicate that the pond has received about 1 Ci of Pu in its 33 yr history, and most of this has been retained by the sediments. Presently the mean inventory of Pu in biota is nearly 3 orders of magnitude smaller than in the sediments, and among these biota, diatoms and pondweed account for most of the biotic inventory. Emergent insects, the most direct biological route of export from the pond, contain $< 1 \times 10^{-1}\%$ of the Pu in biota. It is of interest to note that the largest Pu export route among mammals is man (the researcher), who removed about 200 kg of samples a year containing 4×10^4 nCi of Pu! The evidence here suggests, then, that although Pu is indeed available to biota in

hydrolysis and precipitation. It should be noted here that the soils and water used in these experiments are typical of the Colorado Plains region (calcareous soils, hard water) and not the sandy soil and soft water of the southeast where dissolved, refractory organic compounds are commonplace in surface water. Thus, transport by humics under other environmental circumstances is a possibility not explored in these studies. Even if the plutonium contaminant is present as a valence stable Pu(VI) carbonate complex, both reducing sugars and fulvic acid apparently effectively reduce the plutonyl form to a less soluble tetravalent form, including the case where the organics are present as coatings on clay.²⁰ These studies suggest that organics such as humic substances tend to remove TRU from solution rather than enhance their solubility, at least in the short-term. These organic forms attached to solid particles could, however, become a substrate for microbial action and in this way perhaps be mobilized over the long-term.

Continuing studies of potential microbial transformations of TRU in soil have identified several mechanisms whereby microorganisms could affect the long-term solubility of transuranics: direct transformation such as alkylation and alteration of valence state through microbial oxidation or reduction, indirect mechanisms resulting from TRU interactions with microbial metabolites or changes in Eh or pH, and the effects of incorporation in, and release of TRU from microbial tissues.²³ At the present time it is not possible to determine the relative importance of each of these on the behavior of TRU in soils, but general increased water solubility of plutonium on incubation of Pu amended soils over a period of a month under optimum conditions for microbial activity has been reported.²³ Apparently not all TRU-organic complexes are equally suitable substrates for algal or bacterial growth and uptake of Pu, as was seen in laboratory experiments performed with different molecular weight fractions of natural organics from surface water of the southeast.²⁷ For example, the addition of large molecular weight humic acids reduced Pu(IV) uptake in bacterial cultures (*A. hydrophila*) by 50%, while low molecular weight humic and fulvic acids greatly increased uptake. Thus, TRU mobilization in biological materials in water is not at all a simple function of the total organic carbon in solution. Some fraction of these organics sequesters or otherwise reduces biological availability.

Further, evidence of the reducing tendency in biosystems is seen in laboratory studies with bean plant uptake of Pu(IV) and Pu(VI) from water.²⁴

this organically rich ecosystem, the biologically active fraction represents a very small amount of the total inventory and input. Plutonium is not therefore being rendered highly mobile and translocated to biota in large or increasing amounts, or moving out of the system dissolved in ground water as a result of organic modification.

The search for some perspective on possible limiting conditions on Pu in soil-water-biota relationships has lead some investigators to consider stability field diagrams relating the species of Pu which dominate solid and solution phases under varying Eh-pH conditions in the environment.^{20,28,29} In the absence of complexing species this approach suggests that at pH 8, and Eh in the range +0.6 to -0.3 volts, Pu(V) (PuO_2^+) is the dominant state in the aqueous phase,²⁸ but in the presence of typical environmental inorganic ligands and oxidizing conditions, Pu^{3+} and its complexes predominate; while under reducing conditions, $\text{PuO}_2\text{CO}_3\text{OH}^-$ is the major soluble species at that pH.²⁹ The dominant solid phase under these conditions, according to their analyses, is PuO_2 . These analyses considered Pu(IV) and its complexes to be so insoluble as to be negligible relative to Pu(III). However, another approach is to consider the stepwise hydrolysis of Pu^{4+} under equilibrium conditions with PuO_2 , and evaluate the hydrolytic properties of Pu(IV), the most stable oxidation state, although not necessarily the most soluble.¹⁸ The results indicate that a hypothetical species, $\text{Pu}(\text{OH})_5^-$, dominates the soluble species for pH > 5.5. The equilibrium concentration, charge, and ionic state agree well with White Oak Lake water plutonium field data which characterize the solution phase Pu in this lake as being low molecular weight, in the Pu(III) or Pu(IV) state, and anionic. Indeed, reported concentrations of soluble Pu from a variety of environmental conditions and pH's compare rather well with the indications of this simple model.¹⁸ Although the intent of such a model was not to suggest crystalline PuO_2 controls soluble Pu in natural, trace concentration conditions, it does indicate, along with the previously reviewed studies, that the effects of biological modification (directly or indirectly) on the solubility of Pu are not significantly greater than would be expected from the solubilization of Pu (IV) species from a refractory source.

Mechanism 3: Radiolytic transformations. Several laboratory studies of the dissolution of PuO_2 in near-neutral solutions have found that Pu appears to dissolve faster (in the case of ^{238}Pu) and yield soluble concentrations greater

TABLE D-XV
GASTROINTESTINAL ABSORPTION OF ^{238}Pu AND ^{241}Pu NITRATES BY ADULT RATS,
GUINEA PIGS AND DOGS

Nuclide	Dose (μCi)	Days Until Sacrifice	Species	No. Animals	Percent of Administered Dose				
					Skeleton	Liver	Skeleton + Liver	Urine	Total
Pu^a	5	7	Rat	21	0.010	0.001	0.011	0.018	0.029
Pu^a	7	3	G. Pig	10	0.011	0.008	0.019	0.008	0.027
Pu^b	6	3	G. Pig	5	0.018	0.027	0.045	0.010	0.055
Pu^b	300	3	Dog	2	0.007	0.0006	0.0076	0.028	0.036
Pu^b	300	3	Dog	4	0.002	0.0005	0.0025	0.021	0.032
Am	4	7	Rat	11	0.014	0.003	0.017	0.044	0.061
Am	5	3	G. Pig	9	0.006	0.008	0.014	0.021	0.035
Am	15	3	G. Pig	6	0.004	0.007	0.011	0.006	0.017

a 65% Pu (IV) from TTA Extraction.

b 95% Pu (IV) from TTA Extraction.

contained only 1/3 as much as the muscle. The results of the tissue analysis are given in Table D-IV. The absorption did not differ significantly among any of the species. Absorption may have been somewhat higher in guinea pigs receiving the 95% Pu(IV) as compared to those receiving the 65% Pu(IV) but the opposite effect occurred with dogs. This may indicate that the effect was due to something other than the valence state. The rat absorbed twice as much americium as plutonium but the absorption by the guinea pigs was about the same for the two nuclides.

An interspecies comparison of the behavior of plutonium in the neonatal animal was done by Sullivan²¹ using both rats and swine. Acid solutions of Pu(IV) nitrate were adjusted to pH 2 and administered either by gavage in a 1 mL volume to day old pigs or by gavage in a 0.1 mL volume to 1 or 2 day old rats. Two separate litters of swine were used, one with a high dose of 400 μ Ci/kg and the other with a low dose of 25 μ Ci/kg. The latter were bottle fed. The distribution of ²³⁸Pu in neonatal rats is given in Table D-XVI. At 3 days, 75% of the dose remained in the gut and 1.16% was deposited in the liver and skeleton. At 10 days 2.7% remained in the gut with only a small increase in liver and skeleton.

The data from the swine are shown in Table D-XVII. Distribution of the absorbed ²³⁸Pu indicates that 15 - 25% of the Pu was still in the gut of the high-dose suckled pig at 10 days, while 51% was present in a bottle-fed animal that died at 3 days. About one-half of the ²³⁸Pu absorbed was in the skeleton and about one-fourth in the liver. From these results, it appears that ²³⁸Pu is more highly absorbed from the GI Tract of the swine than from the rats. Closure of the intestine to the passage of macromolecules occurs within a day after birth in swine, as it does in man, while, for the rat, closure takes 3 weeks.

The absorption of a number of nuclides from the GI Tract of the adult and newborn rat was studied by Sullivan and Crosby.²³ The "soluble" forms were prepared by diluting nitric acid solutions and adjusting the pH to 2. Americium oxide was suspended in water a few days before administration. Curium oxide was administered either as a fresh suspension or after standing in water for a few months. Adults were administered 5 to 8 μ Ci contained in 0.5 to 1.0 mL of solution by gavage. Two day old rats received about 2 μ Ci in a volume of 0.05 to 0.1 mL. Excreta were collected from the adult rats and the animals were killed at 7 days and the tissues sampled for analysis. The data for the adult rats are given in Table D-XVIII.

rats²¹, a similar finding was observed but the hematologic data indicated that the animals must have received an inadvertent administration of iron. From these data the authors conclude that rats and mice are distinctly different in regard to the effect of iron deficiency on their plutonium absorption and retention.

Sullivan²⁰ studied the influence of species by the use of rats, guinea pig (herbivorous) and beagle dogs (carnivorous). Nitric acid solutions of ²³⁸Pu and ²⁴¹Am were adjusted to pH 1.5 with NaOH and either injected intragastrically into rats and guinea pigs or mixed into a 100 g bolus of meat and fed to dogs. Animals were housed in metabolism cages and the urine and the feces were collected separately. The distribution of the absorbed plutonium in the dogs is given in Table D-XIV. The concentration in the testes was higher than that in any tissue with the exception of the skeleton. In total quantities the skeleton

TABLE D-XIV
DISTRIBUTION OF GASTROINTESTINALLY ABSORBED
PLUTONIUM-238 IN DOGS

	<u>Percentage of Ingested Dose. x 10³</u>	
	<u>Females^a</u>	<u>Males^b</u>
Skeleton	7	2.21
Liver	0.65	0.58
Kidneys	0.04	0.03
Spleen	0.02	0.02
Lung	0.02	0.03
Adrenal	0.01	0.001
Testes	-	0.04
Muscle	0.76	6.49
Blood	-	0.02
Urine	28	21
Feces	96 000	97 000
Total Percent	96.5	97.5
Recovered		

^aTwo dogs, killed four days after ingestion of ²³⁸Pu (65% IV-valence, as determined by TTA extraction).

^bFour dogs, killed three days after ingestion of ²³⁸Pu (95% IV-valence, as determined by TTA extraction).

TABLE D-XII
 ABSORPTION OF ^{239}Pu IN SELECTED TISSUES OF
 IRON-DEFICIENT AND CONTROL MICE

	<u>% Administered Dose</u>			
	<u>24 h</u>		<u>96 h</u>	
	<u>Control</u>	<u>Iron Deficient</u>	<u>Control</u>	<u>Iron Deficient</u>
Spleen	0.0015	0.0038	0.0003	0.0004
Kidney	0.0005	0.0048	0.0002	0.0008
Blood	0.0009	0.0055	0.0001	0.0004
Femur	0.0005	0.0021	0.0005	0.0031
Liver	0.011	0.025	0.01	0.012
Carcass	0.018	0.065	0.012	0.098

TABLE D-XIII
 ABSORPTION OF PU CITRATE BY IRON DEFICIENT
 AND CONTROL RATS

	<u>% Administered Dose</u>	
	<u>After 48 h</u>	<u>After 96 h</u>
Iron Deficient	0.036	0.030
Control	0.046	0.026

TABLE D-XI

GI UPTAKE IN THE RAT WITH
CHELATING AGENTS

	Retention or Excretion			
	Nitrate	DTPA	Citrate	DTPA-Citrate
Liver	2.1×10^{-4}	1.3×10^{-3}	1.8×10^{-2}	3.1×10^{-3}
Skeleton ^a	9.2×10^{-4}	1.6×10^{-3}	7.4×10^{-2}	4.8×10^{-3}
Total Retained	1.1×10^{-3}	2.9×10^{-3}	9.2×10^{-2}	7.9×10^{-3}
Urine	0.014 ^b	0.79	0.059	1.7
Body + Urine	0.0011	0.79	0.15	1.7

^aEstimated as 23 times the femur content.

^bBelieved to have been contaminated from feces and not included in total.

half of the mice were killed at either 24 or 96 hours after administration. The analysis data from selected tissues are given in Table D-XII.

The mean plutonium content of the iron-deficient mice was higher than the iron-replete mice at 24 hours. By 96 hours, the soft tissue content in the iron-deficient mice had decreased markedly and the bone had increased, suggesting translocation from soft tissue to bone. Total absorption after 24 hours was $0.108 \pm 0.021\%$ in iron deficient mice and $0.028 \pm 0.007\%$ in iron-replete mice. Corresponding values after 96 hours were $0.117 \pm 0.021\%$ and $0.024 \pm 0.005\%$.

In a follow-up study with rats, Eagan¹⁸ fed two groups of weanling rats an iron deficient diet until they were about 70 or 100 days of age. At that time, the rats were administered freshly prepared ²³⁹Pu citrate at a pH of 3.5 by intragastric gavage. One group was killed at 48 hours after administration and the other 96 hours later. Littermates, maintained on a control diet, were gavaged with plutonium and killed at the same time as the experimental group. There was no difference in the uptake of plutonium in the control and iron deficient groups as is shown in Table D-XIII. In the previous study with

TABLE D-X

UPTAKE OF PLUTONIUM IN THE RAT FROM
0.5% SODIUM CITRATE SOLUTION

<u>Tissue</u>	<u>Uptake-% of Administered Dose</u>	
	<u>Non-Fasted</u> <u>Animals</u>	<u>Fasted</u> <u>Animals</u>
Gut	0.39	27
Lung	0.01	0.04
Skin	0.19	0.73
Liver	0.02	0.07
Skeleton	0.06	0.07
Carcass	0.04	0.17

The effect of chelating agents on the uptake of plutonium nitrate was investigated by Baxter and Sullivan.¹⁶ Four groups of 5 female, albino rats were used with solutions administered by gavage. The first group received plutonium nitrate; the second, plutonium in a 10% solution of DTPA; the third, plutonium in a 4.5% sodium citrate solution; and the fourth, plutonium in a solution containing both 10% DTPA and 4.5% sodium citrate. The gavage solutions were at pH <2 with one or two mL of solution administered. The total quantity of plutonium was 1270 μ Ci. The quantity excreted in the urine was measured for two days following administration and the retention in the liver and femur was determined after 5 days. These results are given in Table D-XI.

The uptake in the liver and skeleton of the nitrate is similar to that measured in other experiments with the uptake from the 4.5% citrate about 80 times greater. However, the retention of the DTPA chelate is only about 2 - 3 times greater than the nitrate, in accordance with the view that this stable chelate is not metabolized and is readily excreted. The combination of both citrate and DTPA results in an increase in retention of about 7 times over that of the nitrate, indicating that a small portion of the plutonium was bound in the more readily metabolized citrate.

Ragan¹⁷ investigated the uptake of plutonium by iron deficient mice to investigate the possibility that the known association of plutonium transport in the body with physiologic, iron-binding compound could influence the absorption from the GI Tract. Two groups of ICR white mice were fed either an iron-deficient or an iron-replete diet between 11 and 40 days of age. Animals of both groups were then given 15 μ Ci of ²³⁹Pu citrate by gastric intubation and

Richmond et al.¹⁴ in a study of the effects of high specific activity microspheres used as heat sources in thermoelectric generators, fed two male, 35 month old, beagle dogs forty microspheres contained in a piece of frankfurter once per week for 25 weeks. Each microsphere had an average diameter of 189 ± 21 μ m and contained about 0.55 mCi of ^{238}Pu . Thus the total ^{238}Pu fed was about 0.55 Ci. Upon completion of the feeding, the dogs were sacrificed and the plutonium content of the liver, spleen and mesenteric lymph nodes were measured. Results are given in Table D-IX.

It was noted that the highest concentration was probably in the mesenteric lymph nodes. They speculated that the deposited plutonium may represent small pieces (fines) associated with the microspheres or fragments formed before or after feeding. These could then have been moved to the blood by phagocytosis.

Foreman et al.¹⁵ studied the effect of prior x-ray on the absorption of plutonium from the GI Tract. Male, adult, Sprague-Dawley rats were irradiated with 250 to 2 000 R with a 250 kvp x-ray machine at a dose rate of 55 R/min. Two days after irradiation each animal was administered Pu(IV) in a 0.5% sodium citrate solution by stomach tube. Both fed and fasted animals were used to determine the effect of fasting on the absorption. The results indicated no significant effect of the x-ray on the absorption so that the values were pooled for controls and irradiated animals. These results are given in Table D-I.

TABLE D-IX
RETENTION OF ^{238}Pu FOLLOWING
FEEDING OF MICROSPHERES

Dog	Organ	% Retained
80M	Liver	1.02×10^{-6}
81M	Liver	9.2×10^{-7}
80M	^a Lymph Nodes	1.54×10^{-7}
81M	^a Lymph Nodes	1.14×10^{-7}
80M	Spleen	1.73×10^{-8}
81M	Spleen	2.30×10^{-8}

^a Several Nodes.

TABLE D-VIII
ABSORPTION OF ^{238}Pu FROM MICROSPHERES
BY MINATURE SWINE

Tissue	Batch Number			Crushed
	1	2	3	
	(% of Administered Dose)			
Liver	9.5×10^{-8}	2.3×10^{-7}	3.0×10^{-5}	2.7×10^{-7}
Spleen	6.0×10^{-9}	4.3×10^{-9}	8.2×10^{-7}	1.1×10^{-8}
Kidneys	4.4×10^{-8}	9.8×10^{-9}	3.2×10^{-6}	4.0×10^{-8}
Lung	5.0×10^{-9}	4.2×10^{-9}	3.8×10^{-7}	-
Heart	4.0×10^{-9}	2.1×10^{-9}	1.3×10^{-9}	-
Muscle	6.4×10^{-7}	1.8×10^{-7}	1.0×10^{-5}	-
Ovaries	1.9×10^{-8}	4.3×10^{-10}	7.0×10^{-11}	-
Skeleton	3.5×10^{-7}	2.6×10^{-6}	2.8×10^{-5}	7.1×10^{-7}
Other	5.2×10^{-6}	7.4×10^{-9}	4.3×10^{-8}	1.6×10^{-6}
Total Tissue	6.5×10^{-6}	3.0×10^{-6}	4.6×10^{-5}	3.0×10^{-6}
Urine	7.9×10^{-8}	6.0×10^{-7}	-	4.0×10^{-6}

In a comparison study with $^{239}\text{PuO}_2$ microspheres it was found that the average uptake in the liver and skeleton was about $0.15 \times 10^{-3}\%$. Once again there was considerable variability in the results and the authors noted that the highest measured value for ^{238}Pu absorption was only a factor of two lower than the average value for ^{239}Pu .

In a further study of the ingestion of material from microspheres,¹² cleaned $^{239}\text{PuO}_2$ microspheres were ground under ethanol to produce irregularly shaped particles mostly $1 \mu\text{m}$ on the longest dimension with a range in this size from 10 to $0.01 \mu\text{m}$. These particles were administered in capsules by stomach tube to adult female pigs held in metabolism cages. After 14 days the animals were sacrificed and the tissues analyzed. The average results for the three pigs are given in Table D-VIII, last column. While it was expected that the absorption from this crushed material would be considerably higher than for the microspheres, the tissue results are comparable to, or slightly below the values for the intact spheres. This was attributed to a possibly faster passage time through the GI Tract for the crushed material allowing less time for absorption.

TABLE D-VI

RETENTION OF PLUTONIUM IN THE DOG
AFTER ADMINISTRATION IN CITRATE

<u>Organ</u>	<u>Concentration</u> (%/gram)
Lung	0.008
Liver	0.063
Skeleton	0.050
Lymph Nodes	0.033
Turbinates	0.014

TABLE D-VII

RETENTION OF ORALLY ADMINISTERED
 ^{239}Pu AND ^{241}Am IN ADULT RATS

<u>Organ</u>	<u>% Deposited</u>	
	^{239}Pu	^{241}Am
Liver	0.015	0.2
Spleen	0.004	0.01
Kidney	0.01	0.02
Femur	0.03	0.04

TABLE D-V

RETENTION OF PLUTONIUM IN THE LIVER
AND SKELETON OF MINATURE SWINE

Amount Administered (μ Ci)	Time To Autopsy (Days)	<u>% Of Administered Dose</u>		
		<u>Liver</u>	<u>Bone</u>	<u>Total</u>
613	694	0.0003	0.0003	0.0006
579	664	0.0001	0.0002	0.0003
615	29	0.0002	0.0003	0.0005

72 $\pm$ 3% ultrafilterable through Visking tubing. The dog was sacrificed after 3 days and the tissues were analyzed. These results are given in Table D-VI.

Taylor¹⁰ performed a comparative study of the deposition of plutonium and americium by several routes of administration after noting that rats given 2.95 μ Ci ²³⁹Pu/kg by intravenous administration developed about 80% bone tumors in 12-18 months while very few bone tumors were observed in similar groups of animals given 2.5 or 5.8 μ Ci/kg of ²⁴¹Am. Deposition in the tissues of both ²³⁹Pu and ²⁴¹Am following oral administration in 0.02 N HNO₃ are given in Table D-VII. These values were read from a bar graph in the paper.

The use of ²³⁸Pu in the form of microspheres as a thermal power source for the generation of electricity in space was the impetus for study of the passage time through the gut of experimental animals and the measurement of the fraction absorbed from the gut by Smith et al.^{11,12,13} In the first study, three batches of production-run microspheres were used with the third batch cleaned ultra-sonically to remove fines. The microspheres were administered to minature swine by gavage and the animals were maintained in metabolism cages for 14 days following administration. Total urine output was measured during this period and the tissues were sampled at necropsy. A summary of these results is given in Table D-VIII.

In reviewing Table D-VIII it must be remembered that the quantity of plutonium administered to each animal ranged between 0.3 and 1.1 curies. The values given in the table are averages and do not fully reflect the wide variation found between individual animals. Particularly puzzling was the very high absorption and transfer to tissue in batch 3 which had been cleaned. However, the average transfer to tissue in each batch of three animals varied from $5 \times 10^{-5}\%$ to $3 \times 10^{-6}\%$.

TABLE D-IV

EFFECT OF VALENCE AND CITRATE ON THE RETENTION
OF PLUTONIUM AFTER INTRAGASTRIC ADMINISTRATION

Anion	pH	Valence	% Administered Dose Retained					
			At 4 Days			At 60 Days		
			Bone	Liver	Soft Tissue	Bone	Liver	Soft Tissue
Nitrate	1	68% IV	0.17	0.09	0.015	0.14	0.01	0.01
Nitrate	1	100% VI	0.94	0.81	0.12	1.3	0.27	0.09
Nitrate	2	90% III	0.0041	0.0016	0.006	0.01	0.0004	0.0009
		10% IV						
Nitrate	2	7% III	0.0027	0.0015	0.0008	0.0026	0.0002	0.0003
		93% IV						
Nitrate	2	96% IV	0.0007	0.0005	0.0001	0.0010	0.0001	-
Nitrate	4	97% IV	0.0011	0.0005	0.0001	0.0003	0.00003	-
Citrate	2	99% IV	0.017	0.009	0.003	0.0037	0.0008	0.0008
Citrate	2	96% IV	0.18	0.084	0.023	0.14	0.013	0.0008
		4% VI						
Citrate	2	85% IV	0.26	0.14	0.012	0.21	0.026	0.0019
		15% VI						

the concentrations in other organs were negligible in the 480-695 day time period at which the organs were analyzed. Plutonium (IV) nitrate was used at a pH of 2. Quantities of the plutonium administered and the fraction in the two organs are given in Table D-V.

The results of a continuous feeding experiment are given by Moskalev et al.⁸ in the form of equations. Plutonium-239 was administered in a 1% solution of sodium citrate (pH = 6.4) by placing the solution on the root of the tongue where it was quickly swallowed. The oxidation state was not determined but was believed to be IV. The results were given in the form of accumulation equations relating the quantity in the organ, q , to the time over which the feeding occurred.

$$\begin{aligned}\text{Skeleton } q &= 0.0478t^{0.878} \\ \text{Liver } q &= 0.0129t^{0.801} \\ \text{Kidneys } q &= 0.00245t^{0.835}\end{aligned}$$

The animals used were adult rats. Unfortunately, the actual data were not given so that the scatter and goodness of fit cannot be examined.

Ballou et al.⁹ administered 580 μCi of ^{239}Pu in a 4% citrate solution buffered at pH 3.5 to a single female beagle weighing 12 kg. The solution was

In an extension of these experiments, Weeks et al.⁴ investigated the effect of several variables on the uptake including the possible mass influence, the use of a different species of animal and the influence of the valence state of the plutonium. For the mass effect studies, ²³⁹Pu was used in concentrations ranging from 0.00015 µg/mL to 1.1 µg/mL resulting in total quantities fed ranging from 0.019 to 140 µg. Solutions and procedures were the same as for the earlier ²³⁸Pu work. Results of this experiment are given in Table D-II.

These results, in combination with those obtained earlier with ²³⁸Pu at low mass levels, indicate no statistically significant effect of concentration in the range of 10⁻⁵ µg/mL to 1 µg/mL.

In order to investigate the dependency of the uptake on the species of animal, a single dose of plutonium (IV) nitrate solution, pH2, was administered by stomach tube to three female pigs weighing between 30 and 45 kg and to 15 young-adult, female, Sprague-Dawley rats. The pigs received 20 mL of a solution containing 0.45 µg/mL and the rats received 1 mL each of the same solution. Single rats were sacrificed at 2, 3, 4, 5, and 7 days after administration and the remaining 10 rats and the pigs were sacrificed at 9 days after administration.

There was little difference between the results from the rats sacrificed before 9 days and those sacrificed at 9 days so that the data were pooled. Summary results of these experiments are given in Table D-III.

TABLE D-II

RETENTION OF PU IN THE RAT AFTER CHRONIC ADMINISTRATION
AT DIFFERENT MASS LEVELS

Conc. in Solution <u>Fed</u> (µg/mL)	Total Pu <u>Fed</u> (µg)	<u>Pu Retained</u>		
		<u>Skeleton</u> (%)	<u>Soft Tissue</u> (%)	<u>Total</u> (%)
0.00015	0.019	0.0022	0.00001	0.0022
0.0021	0.27	0	0	0
0.011	1.4	0.0011	0.00036	0.0015
0.11	13	0.0028	0.00017	0.0030
0.22	27	0.0028	0.00035	0.0032
0.44	55	0.0024	0.00018	0.0026
1.1	140	0.0028	0.00046	0.0033

gavage. Initially one dose was administered per day but, to shorten the time of the experiment, the feedings were later increased to two doses per day, then to three and finally to four. The total time of feeding was 9-1/2 months during which time 517 doses were administered. The solution used was the nitrate at a pH of 2 with a volume of 1 mL per dose. At the end of the feedings, 30 rats in the highest dose group were set aside and sacrificed at 90, 180, and 250 days to determine if loss had occurred from the tissues during the period of feeding. All other animals were sacrificed at 4 days after the feeding had stopped and the tissues were analyzed. The results of this experiment are given in Table D-I.

There was no detectable difference among the values found with the exception of Group IVb which appeared to have a higher retention. This was attributed to experimental error and the low levels of activity present in the animals which were difficult to detect with the instruments then available.

TABLE D-I
RETENTION OF ^{238}Pu NITRATE IN TISSUES OF RATS
FED FOR 9-1/2 MONTHS

Group	Total Fed (d/m)	Time Sacrifice (days)	Retention in Tissues		
			Skeleton (\$)	Soft Tissues (\$)	Total (\$)
I	18 820	4	^a ND	ND	ND
II	56 700	4	0.00194	ND	0.00194
III	188 710	4	0.00246	0.00044	0.00290
IV	580 000	4	0.00253	0.00022	0.00275
IVa	Same	90	0.00234	0.00020	0.00254
IVb	Same	180	0.00208	0.00080	0.00288
IVc	Same	250	0.00226	0.00050	0.00276
IV-all	Same	-	0.00234	0.00021	0.00261

^aND - Not Detectable.

APPENDIX D

TRANSURANIUM ELEMENT ABSORPTION FROM THE GI TRACT - A REVIEW

J. W. Bealy

I. INTRODUCTION

The absorption of plutonium from the gastrointestinal tract (GI Tract) has been taken as 0.003% since the issuance of the presently used Maximum Permissible Concentrations (MPCs) for air and water by the National Council on Radiation Protection and Measurements (NCRP)¹ and the International Council on Radiological Protection (ICRP).² This value resulted from the extensive chronic feeding experiments at Hanford^{3,4} in which the quantities remaining in the skeleton and liver after feeding for 9-1/2 months were measured. These data also provided the basis for the assumption by both the NCRP and the ICRP that 90% of the plutonium is deposited in the bone.

Recently, there have been proposals to increase this value and to make it more responsive to various environmental conditions. The Environmental Protection Agency⁵ proposes values of 0.1% for non-oxide compounds, 0.1% for ²³⁸Pu as oxide, 0.01% for ²³⁹Pu as oxide and 0.5% for either, when biologically incorporated. There was, however, no attempt to justify these values by a review of available data. Larsen⁶ has measured the valence state of plutonium in water and suggests that this treatment will oxidize Pu(IV) to Pu(VI) resulting in GI uptakes some three orders of magnitude greater than that of the Pu(IV). In addition, other transuranium elements are becoming of greater importance and values for the transfer across the GI tract are badly needed for these materials.

This report reviews the information that is available on the uptake from the GI tract under various experimental conditions and derives values to be used in estimating radiation doses.

II. REVIEW OF THE DATA

One of the more extensive experiments on the deposition of plutonium following chronic administration was that of Katz et al.³ A primary objective was to measure the uptake at mass levels approximating that of the Maximum Permissible Concentration in water. Young, adult Sprague-Dawley rats in four groups, with 20 males and 20 females in each group, were administered ²³⁸Pu by

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possible at present. K_d , the distribution coefficient, is the most widely used generalized measure of mobility in soil. But it is recognized that this parameter, and the usual laboratory methods for its measurement, are not adequate to account for all the factors discussed above as they affect mobility in-situ. Probably the most problematic aspect of mobility to account for by K_d is the movement of particulate forms. The most notable finding of the many recent studies of organic/biological modifications appears to be that while these processes may have a significant role in increasing the release rate from the waste matrix, there is little or no expected increased mobility in natural soil/water systems over that exhibited by natural thorium.

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than that predicted by simple chemical solubility.^{20,28,30,31} Whether the solubility of PuO_2 involves a reversible reaction so that true equilibrium can be achieved in the presence of water has been questioned,³⁰ but some sort of equilibrium is achieved, if only with the hydrous oxides precipitated from solution. The rate of dissolution is dependent on many factors including temperature, oxidizing, reducing, or complexing agents and specific activity of the Pu isotopes.^{20,30} Tests for the presence of oxidized species of Pu have been made,³² revealing the presence of substantial amounts of Pu(V) or Pu(VI) in neutral solutions contacting high-fired PuO_2 . A model has been proposed³¹ which accounts for the observed differences in solubility between ^{238}Pu and ^{239}Pu as due to an aggregate recoil effect. This phenomenon is the formation of small (colloidal dimension) particles by alpha emission recoils, in which the recoiling uranium atom carries $\sim 10^4$ atoms of Pu from the oxide surface per effective recoil.³¹

The oxidation processes appear to be due to radiolysis and the formation of hydrogen peroxide.^{20,30,32} In an experiment²⁰ in which plutonium in the form of PuO_2 microspheres (10^{-11} M Pu(IV)) was added to a solution of H_2O_2 (10^{-6} M) along with 6.6×10^{-10} gm ^{242}Pu in the tetravalent state as an aqueous phase plutonium, a steady dissolution and formation of oxidized ^{239}Pu was observed, while the ^{242}Pu in the bulk solution was not observed to be oxidized. This suggests that impurities or atmospheric oxygen are not involved, but that oxidation occurs in the intense radiation field right at the surface of the oxide particle.²⁰ In the presence of organic solutes such as EDTA or citrate only the Pu(IV) oxidation state was observed, perhaps due to the reducing properties of these solutes or to interference with oxidation at the surfaces. From dissolution rate studies³³ and the recoil model³¹ it has been estimated that the dissolution of PuO_2 particles is very slow, on the order of hundreds or thousands of years. Still, this mechanism could be important in determining the nature of the source term of TRU in buried waste.

IV. DISCUSSION

Several mechanisms for possible mobilization from a waste matrix and factors affecting mobility in the subsurface have been reviewed briefly here. Naturally, it would be useful if these qualitative descriptions could be reduced to quantitative form, perhaps in terms of functional relations between release and transport rates and the many factors discussed, but this does not appear

TABLE D-XVII

DISTRIBUTION OF ^{238}Pu IN NEONATAL SWINE GAVAGED AT 1 DAY OF AGE

Time of Necropsy, days	Percent of Gavaged ^{238}Pu Dose					
	3	7	9	10	12	21
Dose, $\mu\text{Ci/kg}$	25	29	480	355	27	26
No. of Pigs	1	1	2	2	2	2
<u>Tissue</u>						
Skeleton	15	14			13	16
Liver	6.7	6.9	3.4	3.45	5.0	6.4
Muscle	4.7	5.1			4.0	1.67
Kidneys	0.62	0.43	0.15 ^b		0.28	0.23
Spleen	0.20	0.15			0.14	0.13
Heart	0.15	0.05			0.06	0.10
Lungs	0.51	0.49	0.06 ^b		0.48	0.61
Testes	0.04	0.02	0.02 ^b	0.01	0.02 ^b	0.04 ^b
Lymph Nodes					0.65	0.90
Skin	3.3	2.2			1.4	1.5
GI Tract	51 ^c	4.1	15	27	1.4	0.79
GI Content		1.6	9.1	2.3	0.08	0
Carcass ^a			75	38		
Total Pu Retained (excluding GI tract)	31	30	79	41	26	29

^aEntire carcass, excluding GI tract and contents.^bSingle measurements.^cGI tract, including content.

TABLE XVIII

DISTRIBUTION OF RADIONUCLIDES IN ADULT RATS SEVEN DAYS AFTER
ADMINISTRATION BY GAVAGE OF NITRATES AND OXIDES OF ^{233}U , ^{237}Np ,
 ^{238}Pu , ^{241}Am , ^{244}Cm , AND ^{253}Es

Radio- Nuclide Nitrates	Number of Rats	Percent of Gavage Dose				
		Skeleton ^a	Liver	Skeleton + Liver	Urine	Skeleton + Liver + Urine
^{233}U	11	0.03	0.003	0.03	0.13	0.16
^{237}Np	11	1	0.06	1.1	0.4	1.5
^{238}Pu	12	0.006	0.001	0.007	0.02	0.03
^{241}Am	11	0.012	0.003	0.015	0.05	0.06
^{244}Cm	14	0.04	0.004	0.04	0.08	0.12
^{253}Es	6	0.014	0.001	0.015	0.02	0.04
les						
^{241}Am	6	0.003	0.01	0.013	0.11	0.12
^{244}Cm	3	0	0.003	0.003	0.03	0.03
	11 ^b	(0.02)	(0.014)	(0.04)	(0.06)	(0.1)

^aSkeleton estimated as 23 X single femur content.

^bValues in parentheses obtained using curium oxide suspended in water for 4 months prior to administration. Values not in parentheses were obtained using freshly suspended material.

the intestine - both in the contents and within the intestinal wall. At the high levels used for the experiments providing autoradiographs (doses administered were not specified but were higher than those used for the absorption experiments) substantial damage was observed, particularly in the lower small intestine.

TABLE D-XIX

DISTRIBUTION OF RADIONUCLIDES SEVEN DAYS
 AFTER ADMINISTRATION BY GAVAGE TO TWO-DAY-OLD RATS OF
 NITRATES AND OXIDES OF ^{237}Np , ^{238}Pu , ^{241}Am , ^{244}Cm , AND ^{253}Es

Radio- nuclide	Number of Rats	Percent of Gavage Dose				Total ^b Absorbed
		Skeleton ^a	Liver	Intestine		
				Wall	Contents	
Nitrates						
²³⁷ Np	10 ^c	3.6	0.10	5.5	7.5	6.6
²³⁸ Pu	11	2.6	0.3		13	2.9
²⁴¹ Am	8	4.5	0.3	11	11	5
²⁴⁴ Cm	7	1.8	0.2			2
²⁵³ Es	8	4.4	0.4	0.8	6.8	5.1
Oxides						
²⁴¹ Am	10	0.14	0.02	0.8	0.8	1.1
²⁴⁴ Cm	5	1.9	0.16	2.2	2.8	1.7
	(4) ^d	(2.3)	(0.16)	(7.0)	(11.)	(1.8)

^aSkeleton estimated as $0.22 \times \text{body wt (g)} \times \text{concentration in femur (}\mu\text{g/g)}$.

^bTotal absorbed estimated as sum of separate analyses on femur, liver, and residual carcass (not including intestinal wall or contents).

^cGavaged at 8 days of age.

^dValues in parentheses obtained using curium oxide suspended in water for 4 months prior to administration. Values not in parentheses were obtained using freshly suspended material.

Mahlum and Sikov²³ followed up a finding by Ballou²⁴ that very young rats retained a substantially greater fraction of orally administered plutonium than older rats at 21 days post-feeding. They administered nonpolymeric plutonium(IV) citrate, with a 14-fold citrate excess, by gavage to rats of 1, 20, 21, and 35 days of age. The 20-day age was chosen because it was one day before weaning while the 21 days is one day after weaning. Animals were sacrificed at times ranging from 1 h to 20 days post-administration. Data from this experiment for the animals at 4 hours post feeding and 6 days post feeding are given in Table D-XX. These data were obtained by reading from the bar charts in the report so that some error exists.

The data indicate that there was a higher initial absorption at 4 h in the one-day-old rats than in the older animals. However, there was no significant difference in the absorption at the older ages. The liver of the one-day-old rat had a pronounced uptake that was much greater than the older rats. At 6 days post administration, the retention was higher in the 1 and 20 day old rats than in the others. There was a tendency for the plutonium to translocate to bone in the pre-weanling rats which appeared to decrease with age.

Buldakov²⁵ investigated the uptake of ²³⁹Pu by three suckling pigs that were 2.5 months old and weighted 10-12 kg. 33 μ Ci of plutonium were administered in a 1% citric acid solution at pH 6.5 and the animals were sacrificed one day later. The results are given in Table D-XXI.

The plutonium concentration in the skeleton was determined by the analysis of 11 different bones with the total weight of the skeleton taken as 20 times the weight of the femur. The weight of the muscle was found to be 30% of the body weight. (This decreased to 20% at 640 days.)

In discussion of these results, Buldakov noted that the retention value does not differ significantly from that obtained with a 3% solution but is three times as high as after administration of a 0.5% citrate solution to rats. This may be due to the younger age of the suckling pig.

Moskalev et al²⁶ reported on studies to determine the influence of age on the absorption of ²⁴¹Am. Non-inbred white rats with weights of 15-17 g for 7-day old, 45 $\pm$ 5 g for 30-day old and 180 $\pm$ 15 g for adult animals were used. The rats received a peroral administration of a solution of ²⁴¹Am chloride at a pH of 3 in doses of 30 μ Ci for adult rats and 8 μ Ci for the younger animals. The animals were sacrificed on the first, second and fourth day following the administration. It was found that the absorption depended upon the age of the

TABLE D-XX

EFFECT OF AGE UPON ABSORPTION OF PLUTONIUM
CITRATE IN THE RAT - 3 RATS PER GROUP

Age (days)	% of Administered Dose					
	4 h post feeding			6 days post feeding		
	Total	Liver	Femur	Total	Liver	Femur
1	0.43	0.14	0.0043	0.35	0.13	0.019
20	0.14	0.019	0.0032	0.16	0.007	0.011
21	0.10	0.015	0.0040	0.04	0.005	0.005
35	0.13	0.013	0.0078	0.06	0.002	0.004

TABLE D-XXI

RETENTION IN ORGANS OF SUCKLING PIG FED
 ^{239}Pu IN 1% CITRIC ACID

Organ	Retention (%/organ)	Distribution (% of Total Retained)
Muscle	0.007	3.8
Liver	0.012	6.5
Kidneys	0.0009	0.49
Spleen	0.0010	0.54
Lungs	0.0027	1.46
Ovaries	0.00026	0.14
Skeleton	0.161	87.07
Total	0.185	100

animal, with 0.88% absorbed for the one-week old rats, 0.34% for the month-old rats and 0.07% for the adult rats.

The absorption of curium and californium by both newborn and adult rats was investigated by Sullivan.²⁷ Solutions of ^{244}Cm and ^{252}Cf were made by diluting a nitric acid solution and adjusting the pH to 3. The $^{244}\text{Cm}_2\text{O}_3$ had been suspended in water 4 months prior to being mixed with 5% carboxymethyl cellulose and administered by gavage. Two day old rats were administered 2 μCi in 0.1 mL and adults were administered 5 μCi in 0.5 mL by stomach tube. All animals were sacrificed 7 days after administration for analysis of tissue. Excretion and distribution data are given in Table D-XXII. These data, while preliminary, show relatively low uptake in the adult and high uptake in the newborn.

Sullivan²⁸ provided additional data on the uptake of plutonium and neptunium in the adult and young rat and also included data on the uptake of "biologically bound" plutonium. Solutions were prepared by diluting nitric acid solutions to a concentration of 20 $\mu\text{Ci/mL}$ and adjusting the pH to 2. The oxides were administered as water suspensions of equivalent activity. Two day old rats received 0.05 to 0.1 mL, 8 day old animals 0.1 to 0.2 mL and adults 0.5 mL. Higher doses were also administered intragastrically or intraperitoneally to 2 or 8 day old rats which were killed 5 hours later and fed, in toto or lower small intestine only, to fasted adult rats. The dose administered to the adult was determined by analysis of other injected animals and uneaten bone fragments were analyzed so that their content could be subtracted from the administered dose. All animals were killed at 7 days after gavage and the tissues analyzed. The data for the adults are given in Table D-XXIII.

The authors concluded that the skeleton and liver contained the majority of the absorbed activity. The biologically incorporated neptunium was absorbed 10 to 20 times less efficiently than the inorganic neptunium but the biologically incorporated plutonium appeared to be incorporated 1 to 10 fold more efficiently depending upon the groups compared. They also concluded that the urinary output was not affected by contamination from the feces because the urine of the restrained rats was more radioactive than that of the unrestrained controls.

The data for the young rats are given in Table D-XXIV. Once again, the authors conclude that the levels retained in the skeleton are greater and the amount in the intestine lower with increasing age. They also noted that the skeletal estimates for the young animals are quite uncertain and that more significance should be attached to the values for the total absorbed.

TABLE D-XXII

DISTRIBUTION OF RADIOACTIVITY IN ADULT AND NEONATAL RATS
GIVEN $^{244}\text{Cm}(\text{NO}_3)_3$, $^{244}\text{Cm}_2\text{O}_3$, OR $^{252}\text{Cf}(\text{NO}_3)_3$ BY GAVAGE

Tissue or Excreta	Percent of Administered Dose (Average Values for at Least Six Rats)				
	$^{244}\text{Cm}(\text{NO}_3)_3$		$^{244}\text{Cm}_2\text{O}_3$		$^{252}\text{Cf}(\text{NO}_3)_3$
	Adult	Neonate	Adult	Adult ^a	Neonate
Liver	0.005	0.16	0.02	0.003	0.18
Skeleton ^b	0.07	1.8	0.03	0.002	3.1
Carcass ^c		6.2			4.1
Lungs				0.0	0.006
Skin					0.15
Urine	0.07		0.07	0.1	
Feces	95.8		91.7	94.4	
Total Absorbed	0.15	6.3	0.12	0.11	4.3
Total Recovered	96	6.3	91.8	94.5	4.5

^aNo significant radioactivity was found in the intestine, kidney, spleen, blood, or muscle.

^bAdult skeletal content = single femur content x 23, neonate skeletal content calculated from an analyzed weighed femur on the assumption that total skeleton was 22% of total body weight.

^cIncludes skeleton.

TABLE XXIII

DISTRIBUTION OF RADIONUCLIDES IN ADULT RATS SEVEN DAYS AFTER
ADMINISTRATION BY GAVAGE OR FEEDING

Radio- Nuclide	Form Administered	Number of Rats and Sex	Percent of Administered Dose				
			Skeleton ^a	Liver	Skeleton + Liver	Urine	Skeleton + Liver + Urine
²³⁷ Np	Nitrate	15F	0.63	0.07	0.7	0.45	1.15
²³⁷ Np	Biologically ^b Incorporated	6F	0.03	0.003	0.03		
²³⁷ Np	Nitrate	7M	0.54	0.02	0.56		
²³⁷ Np	Biologically ^b Incorporated	7M	0.05	0.003	0.05		
²³⁸ Pu	Nitrate	15F	0.007	0.003	0.01	0.02	0.03
²³⁸ Pu	Nitrate ^c	3F	0.003	0.0003	0.003	0.02	0.02
²³⁸ Pu	Nitrate ^d	3F	0.003	0.0004	0.003	0.01	0.01
²³⁸ Pu	Biologically ^e Incorporated	4M	0.03	0.0007	0.03		
²³⁸ Pu	Biologically ^f Incorporated	3M	0.01	0.004	0.01	0.03	0.04
²³⁹ Pu	Nitrate	6F	0.016	0.0007	0.017	0.005	0.022
²⁴¹ Pu	Nitrate	6F	0.018	0.009	0.027	0.005	0.032
²³⁸ Pu	Oxide	6F	0.004	0.0006	0.0046	0.007	0.012
²³⁸ Pu	Biologically ^g Incorporated	5F	0.009	0.0003	0.009		
²³⁸ Pu	Oxide ^h	5F	0.01	0.0	0.01	0.015	0.025

^aSkeleton estimated as 23 x single femur content.^bRadionuclide as nitrate administered by gavage to 8-day-old rat that was killed and fed in toto to adult.^cRats maintained in restraining cages after gavage.^dControl rats maintained in metabolism cages for comparison with (g).^eRadionuclide as nitrate injected intraperitoneally into 2-day-old rat that was killed and fed in toto to adult.^fRadionuclide as nitrate administered by gavage to 2-day-old rat three days later the ileum was fed to adult.^gRadionuclide as oxide injected intraperitoneally to 8-day-old rat that was killed and fed in toto to adult.^hSNS-grade ²³⁸PuO₂.

TABLE D-XXIV

DISTRIBUTION OF RADIONUCLIDES IN YOUNG RATS SEVEN
DAYS AFTER ADMINISTRATION BY GAVAGE

Radio- Nuclide	Form Administered	Number of Rats	Dogs Age	Percent of Administered Dose				Total ^b Absorbed
				Skeleton ^a	Liver	Intestine Wall Contents		
²³⁷ Np	Nitrate	5	2	1.0	0.02	18.9	13.7	0.8
²³⁷ Np	Nitrate	6	4	0.7	0.01	11.3	9.8	0.5
²³⁷ Np	Nitrate	10	8	3.6	0.10	5.5	7.5	1.2
²³⁸ Pu	Nitrate	7	2	2.7	0.2	0.7	0.5	2.2
²³⁹ Pu	Nitrate	5	2	0.6	0.06	4.6	3.0	0.4
²³⁸ Pu	Oxide ^c	10	2	0.0	0.03	0.4	0.3	0.05
²³⁹ Pu	Oxide ^d	11	3	0.01	0.002	0.4	0.1	0.02

^aSkeleton estimated $0.22 \times \text{body wt (g)} \times \text{concentration in femur (\%g)}$.

^bTotal absorbed estimated as sum of separate analyses on femur, liver, lung, and residual carcass (not including intestinal wall or contents).

^cSMS-grade ²³⁸PuO₂.

^dWeapons-grade ²³⁹PuO₂.

In a continuation of the effects of age and biological incorporation on the uptake of actinides, Sullivan²⁹ reported on additional administrations. Techniques and solutions or biological material administered were similar to those described previously. Data for the adults are given in Table D-XXV. The author noted that the retention and urinary excretion were similar for both ²³³U and ²³²U and that the quantity of ²⁴¹Am absorbed after gavage was similar to that of the hexavalent uranium. Feeding of ²³³U incorporated in newborn rats resulted in a 2- to 5-fold increase in the quantity retained in the liver and a two-fold increase in the amount excreted in the urine. The effect of biological incorporation was similar for uranium and americium.

The results from the study of newborn rats with U and ²⁴¹Pu are given in Table D-XXVI. It may be noted that the ²³³U nitrate is absorbed to about the same extent as ²³⁴Pu nitrate and very little is retained in the mucosa after

TABLE D-XXV

DISTRIBUTION OF RADIONUCLIDES IN ADULT RATS 7 DAYS
AFTER GAVAGE OR FEEDING BIOLOGICALLY INCORPORATED NUCLIDES

	No. of Rats	Contents as Percent of Dose Administered				
		Skeleton ^a	Liver	Skeleton + Liver	Urine	Skeleton, Liver and Urine
²³² U - Nitrate	6	0.015 ^b ± 0.005	0.007	0.016	0.06	0.066
²³³ U - Nitrate	4	0.014 ± 0.006	0.0008 ± 0	0.015	0.007 ± 0.02	0.085
²³³ U - Nitrate (Biologically Incorporated in Gaviged Neonates)	4	0.014 ± 0.009	0.002 ± 0.001	0.016	0.15 ± 0.02	0.116
²³² U - Nitrate (Biologically, Incorporated by Neonates Injected IP)	2 ^b	0.021 ± 0.02	0.004	0.025	0.15 ± 0.02	0.175
²⁴¹ Am - Nitrate ^c	11	0.012 ± 0.003	0.003	0.015	0.05 ± 0.006	0.065
²⁴¹ Am - Nitrate (Biologically Incorporated in Gaviged Neonates)	6	0.02 ± 0.006	0.01 ± 0.001	0.03	0.05 ± 0.08	0.08

^a Skeleton estimated to be 23 times femur radioactivity.

^b Statistical variation is seen except range is shown for groups of 2 animals.

^c Data from reference 28.

TABLE D-XXVI

DISTRIBUTION OF RADIONUCLIDES IN NEWBORN RATS 7 DAYS
AFTER GAVAGE

Radionuclide	Dose (μ Ci)	No. of Rats	Content as Percent of Gavage Dose ⁺ Sem				Total Absorbed
			Skeleton ^a	Liver	Intestine Wall	Content	
²³² U - Nitrate	2	5	6.2 $\pm$ 0.6	0.054 $\pm$ 0	4.5 $\pm$ 0.7	2.5 $\pm$ 0.4	6.7 $\pm$ 0.4 ^d
²³³ U - Nitrate	2	10	2.1 $\pm$ 0.2	0.02 $\pm$ 0	0.7 $\pm$ 0.1	0.5 $\pm$ 0.1	2.0 $\pm$ 0.2 ^d
²³⁷ Pu - Nitrate	0.05	10	1.3 $\pm$ 0.7 ^b	0.14 $\pm$ 0.01	40.6 $\pm$ 2.3	10.7 $\pm$ 1.4	2.9 $\pm$ 0.3 ^c

^a Skeleton estimated as 0.165 x body wt (g) x concentration in femur (%/g).^b Entire skeleton counted on 3 rats.^c Determined by whole-body counting after removal of the intestine.^d Determined by counting carcass after removal of the intestine.

gavage of 2-day old rats. Although the absorption of ^{234}U and ^{233}U are similar in the adult, up to three times as much ^{232}U as ^{233}U is absorbed by the newborn. In addition, five times as much ^{232}U is retained in the gut wall. The low dose of ^{241}Pu administered showed that the absorption was similar to that measured for ^{239}Pu earlier when quantities 40 times higher in dose and 30,000 times higher in mass were administered. However, a much higher percentage of ^{241}Pu was retained in the gut wall. To investigate the possibility of a mass effect on this phenomenon, groups of two or three rats from two different litters were administered four different doses of ^{238}Pu at two different ages. These results are given in Table D-XXVII. The author concluded that there was little effect of dose on the adsorption from the gut. However, the retention in the intestine wall and contents was markedly reduced at the highest dose level indicating a saturation of the mechanisms responsible for this retention. It was also noted that the 4-hour old animals absorbed more ^{238}Pu from the gut than did the twenty-four hour old animal.

In order to further investigate the possible effects of incorporation into foodstuffs, Sullivan and Garland³⁰ administered alfalfa, that had been grown on soils containing ^{238}Pu , to rats and guinea pigs. The alfalfa was grown to maturity on the contaminated soil and the leaves, or leaves and stems, were dried, ground and pelleted. In addition, alfalfa grown on non-contaminated soil was mixed with a plutonium nitrate solution to produce ^{238}Pu concentrations equivalent to those in the alfalfa grown on contaminated soil (60,000 d/m per gram of plant tissue). A plutonium nitrate solution (pH2) was also administered by gavage. All animals were placed on an alfalfa ration one week prior to administration.

The rats were killed five days after administration and the pelt and viscera were removed and discarded before analysis of the tissues. Results for the rat administrations are given in Table D-XXVIII.

Incorporated plutonium was absorbed to a twofold greater extent than the mixed plutonium ($p = 0.11$). The incorporated plutonium also was absorbed ten times greater than the solution ($p = 0.08$).

The guinea pigs were used in a similar experiment except that leaves and stems, as well as leaves only, were fed. Results of this experiment are given in Table D-XXIX.

When leaves and stems were fed, the incorporated plutonium was absorbed four times greater than when the plutonium was mixed with the feed ($p = 0.13$).

TABLE XXVII

DISTRIBUTION OF PLUTONIUM-238 IN NEWBORN RATS KILLED 7 DAYS AFTER GAVAGE

Administered Dose (μ Cl)	No. of Rats	Age at Gavage (h)	Content as Percent of Gavage Dose ^a				Total ^c Absorbed
			Skeleton ^b	Liver	Intestine Wall	Contents	
0.0036	2	24	6.2 $\pm$ 1.2	0.5 $\pm$ 0.1	90 $\pm$ 14	39 $\pm$ 1.4	10 $\pm$ 1.3
	2	4	7.7 $\pm$ 0.1	0.6 $\pm$ 0.1	83 $\pm$ 17	29 $\pm$ 6	29 $\pm$ 3.8
0.037	1	24	3.3	0.24	64.9	28	2.7
	2	4	3.4 $\pm$ 0.4	0.4 $\pm$ 0.01	51 $\pm$ 1	25 $\pm$ 6	4.8 $\pm$ 0.2
0.39	3	24	1.9 $\pm$ 0.2	0.2 $\pm$ 0.03	33 $\pm$ 7	26 $\pm$ 3	1.6 $\pm$ 0.2
	3	4	4.2 $\pm$ 1.0	0.7 $\pm$ 0.2	27 $\pm$ 6	24 $\pm$ 6	4.7 $\pm$ 1.4
4.14	3	24	2.1 $\pm$ 0.2	0.4 $\pm$ 0.03	1.3 $\pm$ 0.2	6.8 $\pm$ 1.4	2.4 $\pm$ 0.3
	3	4	4.7 $\pm$ 0.8	0.9 $\pm$ 0.2	1.7 $\pm$ 0.6	3.7 $\pm$ 3.1	5.9 $\pm$ 1.1

^a Statistical variation is sem, except range is shown for groups of 2 animals.

^b Skeleton estimated as 0.165 x body wt (g) x concentration in femur (%/g).

^c Total absorbed is sum of carcass, liver, lung, and femur analyses, and does not include gut or gut contents.

TABLE D-XXVIII
 ABSORPTION OF PLANT-BOUND ^{238}Pu BY RATS

<u>Form Administered</u>	<u># Animals</u>	<u>% Administered Dose</u>		
		<u>Liver</u>	<u>Carcass</u>	<u>Total</u>
Incorporated- Leaves	3	0.016	0.068	0.084
Mixed - Leaves	3	0.006	0.032	0.038
Solution	2	0.002	0.006	0.007

TABLE D-XXIX
 ABSORPTION OF PLANT-BOUND ^{238}Pu BY GUINEA PIGS

<u>Form Administered</u>	<u># Animals</u>	<u>% Administered Dose</u>			
		<u>Liver</u>	<u>Skeleton</u>	<u>Carcass</u>	<u>Total</u>
Incorporated Leaves and Stems	2	0.036	0.032	-	0.068
Leaves	3	0.017	-	0.029	0.046
Mixed Leaves and Stems	2	0.008	0.011	-	0.018
Leaves	3	0.023	-	0.076	0.099
Solution	3	0.014	-	0.019	0.033

However, when leaves only were fed, the mixed plutonium showed a higher absorption which was not statistically different from either the plutonium incorporated in the plant material or mixed with the plant material.

In order to obtain further data at higher tissue concentrations, guinea pigs were fed leaves with incorporated Pu, leaves with mixed Pu and solution by gavage for ten days. Animals were killed two days after the last administration. The results of the tissue analyses are given in Table D-XXX.

When leaves and stems were fed, there was a four-fold increase in the uptake of the incorporated plutonium as compared to the mixed plutonium ($p = 0.12$). When leaves only were fed, incorporated plutonium was absorbed to about twice the extent of mixed plutonium ($p = 0.04$). Absorption of the gavaged plutonium was higher than for any of that fed with alfalfa, except for that incorporated in leaves and stems, but the difference was not significant. The authors note that the variability of the data preclude a quantitative evaluation of the increased uptake although it does appear that such an increase occurs with incorporated plutonium.

Zalikin et al.³¹ studied the absorption of ^{241}Am following peroral administration as the chloride in a study of the distribution following several methods of administration. Five white rats with an average weight of 200 g were used. After 4 days, the liver contained 0.015% of the dose. From the distribution noted following intravenous administration, it was noted that the liver contains 47.5% of the ^{241}Am at this time. Assuming that the same distribution applied to the quantity in the body following oral absorption, it was concluded that about 0.03% was absorbed.

In a study of several other actinides, Hungate et al.³² studied the retention of ^{253}Es and ^{249}Bk at 4 and 21 days after intragastric administration of these nuclides to 400-g male rats. The nuclides were contained in 1 mL of 0.2 N HCl. Retention of ^{253}Es in the total carcass was $0.036 \pm 0.02\%$ at 4 days and $0.0044 \pm 0.001\%$ at 21 days. For ^{249}Bk the corresponding values were 0.009% and 0.004%. While the retentions for individual organs were not given, the authors indicated the distribution to be similar to an intravenous study described in the paper. In this distribution, the skeleton content was about 30%, the liver about 1.5%, the kidney about 1% and the spleen about 0.6% for ^{253}Es . For ^{249}Bk , the skeleton contained 40%, the liver about 3%, the kidney about 1.2% and the spleen about 1.2%. The influence of the high acidity and the

TABLE D-XXX

ABSORPTION OF CHRONICALLY INGESTED ^{238}Pu BY GUINEA PIGS

Group	Annual	Form Administered	10-Day Dose Administered (dis/min)	Quantity Absorbed					
				Liver		Skeleton		Carcass	
				(dis/min)	(%)	(dis/min)	(%)	(dis/min)	(%)
GP-6	1	Incorporated	587.000	80	0.014	76	0.013		
	2	(Leaves and	499.000	282	0.057			402	0.081
	3	Stems)	575.000	262	0.046	6.36	0.111		
	Avg.			208	0.039	356	0.062		
GP-7	1	Mixed	666.000	84	0.013			155	0.023
	2	(Leaves and	663.000	52	0.008			111	0.017
	3	Stems)	661.000	35	0.005	56	0.009		
	Avg.			57	0.009			133	0.020
GP-8	1	Incorporated	595.000	103	0.017			156	0.026
	2	(Leaves)	597.000	52	0.009			162	0.027
	3		580.000	55	0.010	162	0.028		
	Avg.			70	0.012			159	0.027
GP-9	1	Mixed	558.000	35	0.006			143	0.026
	2	(Leaves)	557.000	42	0.008	50	0.009		
	3		505.000	26	0.005			60	0.012
	Avg.			34	0.006			102	0.019
GP-10	1	Solution	625.000	77	0.012	153	0.025		
	2	(Gavaged to	625.000	91	0.015			448	0.072
	3	Animals fed	625.000	34	0.005			85	0.014
		Leaves)						119	0.019
GP-11	1	Solution	625.000	50	0.008			125	0.020
	2	(Gavaged to	625.000	302	0.048	216	0.035		
		Animals fed						175	0.028
		Leaves and Stems)						518	0.083
Gr-C	1	None	0	0.7		0		0	
	Avg.			176	0.028			346	0.035

use of the chloride ion on these results cannot be assessed, but it is noted that the uptakes are in the same range as for the other actinides.

III. CONCLUSIONS

These data permit a derivation of uptake values, at least in animals. Some of the following conclusions are relatively well based while others are considerably more uncertain. One important uncertainty that cannot be resolved at this time is the question of species difference. While the comparison among the animals tested does not indicate large differences, there will be no proof that man's behavior in regard to uptake is the same as that of the animals used until experimental data on man become available.

Summary tables of the data from the various investigations, broken down as to compound fed and nuclide, are given in Tables D-XXXI through D-XXXIII so that a ready comparison of the data is possible. In this section we will use these tables, as well as those in the body of the report to discuss the importance of a number of factors that could influence uptake.

A. Effect of Quantity Fed

Since most feeding experiments are done with relatively large doses so that measureable quantities of transuranic element may be deposited in the organs, the question as to whether these experiments describe the uptake at the low concentrations typical of the environmental situation can be considered. Fortunately, this was one of the early questions considered and the studies of Katz et al.⁷ and Weeks et al.¹⁸ show that over a range of 10^{-5} $\mu\text{g/mL}$ to 1 $\mu\text{g/mL}$, in chronic feeding of rats with 1 mL doses, there are no statistical differences in the retention in the organs. The lower end of this range is below the present occupational MPC. It appears, therefore, that any effect of concentration is so small as to be negligible, at least in the range of environmental conditions.

B. Effect of Valence

As was noted in the introduction, the question of the influence of the valence state of plutonium on the uptake from the GI Tract has been of interest in the past few years. The primary data available has been that of Weeks et al.⁴ (Table D-IV). If one assumes the 96% IV for the nitrate pH2 case and the 99% IV for the citrate pH2 case to represent the real absorption of Pu IV, then

TABLE D-III
UPTAKE OF PLUTONIUM FROM NITRATE SOLUTIONS

Material Administered	Time of Loading (Days)	Time to Sacrifice (Days)	Species	# Animals	Ref.	g Administered Dose $\times 10^4$						Total Tissue	Total
						Skeleton	Liver	Soft Tissue	Urine				
^{238}Pu - pH 2	209	3	Rat	40	3	19.4	2.6	-	-	-	-	-	-
						24.6	1.6	-	-	-	-	26.1	1.9
						23.4	1.4	-	2.1	0.5	-	22	6.5
						22	2.5	-	0.1	6.5	-	0	130
^{239}Pu - pH 2	209	3	Rat	20	4	0	35	-	0	120	-	15	26
						11	0.5	-	3.6	24	-	30	3.5
						20	2	-	1.7	2	-	32	2
						20	2	-	3.5	1.5	-	26	2
						24	2	-	1.0	0.5	-	33	2
						20	2	-	4.6	0.5	-	22	6.5
						15	3.0	3.6	1.3	3.3	0.5	32	6
						20	3.7	9.3	3.2	2.1	0.0	-	-
^{239}Pu - pH 2	A ^a	9	Pig	3	4	15	4.6	2.1	0.9	-	-	60	-
			Rat	15	4	20	20	10	2	-	100	20	290
^{239}Pu - pH 2	A	5	Rat	5	16	100	20	10	2	-	80	20	270
			Rat	21	20	100	20	80	150	-	-	30	550
^{238}Pu - pH 1.5-055 IV	A	3	G. Pig	10	20	100	40	270	120	-	100	110	360
			G. Pig	5	20	100	30	6	10	-	200	100	320
			Dog	2	20	70	4	5	1	-	210	100	300
			Dog	4	20	20	-	10	-	-	200	-	300
^{238}Pu - pH 2	A	7	Rat	12	22	60	-	30	-	-	200	-	230
			Rat	15	20	70	-	3	-	-	200	-	130
^{238}Pu - pH 2	A	3		3	20	30	-	4	-	-	100	-	220
				3	20	30	-	7	-	-	50	-	320
^{239}Pu - pH 2	A	7	Rat	6	20	160	-	90	-	-	50	-	-
			Rat	6	20	100	-	20	-	-	-	70	-
^{241}Pu - pH 2	A	7	Rat	2	30	-	-	140	-	-	-	330	-
^{238}Pu - pH 2	A	5	Rat	3	30	-	-	110	-	-	-	480	-
^{238}Pu - pH 2	A	5	G. Pig	3	30	-	-	200	-	-	-	550	-
^{238}Pu - pH 2	10	2	G. Pig	3	30	350	-	200	-	-	-	-	-
^{238}Pu - pH 2	10	2	G. Pig	2	30	350	-	200	-	-	-	-	-

^a Route

TABLE XXIII

PLUTONIUM UPTAKE FROM CITRATE SOLUTIONS

Material Administered	Time of Feeding (Days)	Time to Sacrifice (Days)	Species	# Animals	Ref.	% Administered Dose x 10 ⁴								Total Tissue	Total
						Skeleton	Liver	Soft Tissue	Urine						
²³⁹ Pu - 4% Citrate - pH 3.5	A ^a	3	Dog	1	9	400	-	200	-	-	-	-	-	830	-
²³⁹ Pu - 5% Citrate	A	-	Bat	40	15	400	300	200	200	400	500	-	-	-	-
						1 300	600	200	200	00	100	-	-	-	-
						200	0	300	100	400	0	-	-	-	-
4.5% Na Citrate	A	5	Bat	5	16	740	600	180	190	-	-	590	410	910	1 500

^aAcute

TABLE D-XXIII
UPTAKE OF OTHER NUCLIDES

Material Administered	Time of Feeding (Days)	Time to Sacrifice (Days)	Species	# Animals	Ref.	1 Administered Dose x 10 ⁴									
						Skeleton	Liver	Soft Tissue	Urine	Total Tissue	Total				
²⁴¹ Am Nitrate pH 2	A ^a	7	Rat	11	22	120	-	30	-	-	500	-	150	-	600
²⁴¹ Am Oxide	A	7	Rat	6	22	30	-	100	-	-	1 100	-	130	-	1 200
²⁴¹ Am Chloride	A	4	Rat	-	19	-	-	150	-	-	-	-	300	-	-
²⁴¹ Am Chloride	A	1-4	Rat	-	26	-	-	-	-	-	-	-	700	-	-
²⁴¹ Am Nitrate	A	7	Rat	11	29	120	30	30	-	-	500	60	150	-	650
²⁴¹ Am Nitrate pH 1.5	A	7	Rat	11	20	140	40	30	5	-	440	60	-	-	610
		3	G. Pig	9	20	60	20	80	20	-	210	40	-	-	350
		3	G. Pig	6	20	40	10	70	10	-	60	10	-	-	170
²³² U Nitrate	A	7	Rat	6	29	150	50	70	-	-	500	-	-	-	660
²³³ U Nitrate	A	7	Rat	4	29	140	60	8	0	-	700	200	-	-	850
²³³ U Nitrate pH 2	A	7	Rat	11	22	300	-	30	-	-	-	-	-	-	1 600
²³⁷ Np Nitrate pH 2	A	7	Rat	11	22	10 000	-	600	-	-	4 000	-	-	-	15 000
²³⁷ Np Nitrate pH 2	A	7	Rat	15	28	6 300	-	700	-	-	4 500	-	-	-	11 500
²³⁷ Np Nitrate pH 2	A	7	Rat	7	28	5 400	-	200	-	-	-	-	-	-	-
²³⁷ Np Nitrate pH 2	A	7	Rat	14	22	400	-	40	-	-	800	-	-	-	1 200
²⁴⁴ Cm Nitrate	A	7	Rat	3	22	0	-	30	-	-	300	-	-	-	330
²⁴⁴ Cm Oxide Fresh	A	7	Rat	11	22	200	-	140	-	-	600	-	-	-	940
²⁴⁴ Cm Oxide Aged	A	7	Rat	-	22	700	-	50	-	-	700	-	-	-	1 500
²⁴⁴ Cm Nitrate pH 3	A	7	Rat	-	27	300	-	200	-	-	700	-	-	-	-
²⁴⁴ Cm ₂ O ₃	A	7	Rat	-	27	20	-	30	-	-	1 000	-	-	-	1 100
²⁵² Cf Nitrate	A	7	Rat	-											

^aAcute

the uptake of the other valence states can be estimated by assuming that each one acts independently. The ratio of uptake for the III valence state compared to IV in bone ranges from 6 to 42, in liver from 3 to 30 and in the total animal from 10 to 20. For the VI valence state in citrate the bone ratios range from 100 to 900, the liver from 100 to 400 and the total animal from 90 to 700. This examination would, indeed, imply an increase in the uptake for both the III and the VI valence state over that of the IV.

However, Sullivan has recently been studying this situation and has found factors that make these earlier results questionable.³³ In the first place the animals were starved before administration and, following administration, until sacrifice. Secondly, the oxidant used was $K_2Cr_2O_7$ which could influence uptake by itself or by acting as a holding oxidant. In order to explore this situation he performed experiments with rats to determine the effect of these variables as well as the use of ozone as an oxidant. While his data are not as yet published so that we do not wish to include them here, they show the effect of valence change to be small, if present at all. We will, therefore, not consider such an effect to be of importance.

C. Uptake by Adult Animals

As can be seen from Tables D-XXXI through D-XXXIII, there is considerable scatter in the data available. For example, the highest value for retention of plutonium administered as the nitrate in the skeleton is over an order of magnitude greater than the lowest. A complicating factor in the comparison and analysis of the data is that the same tissues were not analyzed for all of the animals. An additional uncertainty is introduced by the significant quantities excreted in the urine in many of the later studies where the urine was measured. This material excreted in the urine was absorbed from the GI Tract although it was excreted before deposition in tissue. This raises the question as to whether this fraction will behave differently in man and be deposited in tissue rather than excreted. Once again, however, the fractions excreted in the urine and deposited in tissue appear to be similar in the three species examined (rat, dog, and guinea pig) so that it appears that the material absorbed through the GI Tract is more easily excreted in the urine than that injected intravenously. This view is supported by the comparison between the urine data for humans after intravenous administration, with similar data for dogs given by Ballou et al.⁹ Here the excretion rates of the dog and the human for the first 100 days

were similar (within a factor of about 5 at the later times). This indicates that for similar methods of administration the results are similar. Since our interest is in the quantity deposited in tissue and not the total absorption, we will consider the deposition to be the same in man as in the animals and the uptake will be considered as that deposited, without the urine included.

One other question must be considered before uptake values can be derived. This is the difference between ^{238}Pu and ^{239}Pu . In studies with microspheres of "heat source" plutonium containing about 80% by mass of ^{238}Pu , it has been shown that the material upon standing in water becomes more soluble and the uptake of this material in animals upon inhalation is increased. This has been attributed to the high specific activity of the heat source plutonium leading to ejection of a few atoms by recoil following a disintegration. It is sometimes assumed that the ^{238}Pu will be more soluble in all forms. However, for plutonium in the environment (unless it is in the form of actual particles of heat source plutonium which would result in high concentrations) or for plutonium in solution this specific activity should make little difference. This has been confirmed by the experiments of Katz et al.³ and Weeks et al.⁴ where both ^{238}Pu and ^{239}Pu were administered as nitrate with no difference in the retention in the organs. Even for the oxide, the uptake from solution should provide a maximum value because here the material is already soluble. We will, therefore, consider the uptake for these two isotopes to be the same.

Several averages for the uptake from nitrate solution can be derived from the data in Table D-XXXI. If all of the values are used, one obtains an average weighted for the number of animals in each experiment of 0.0044% for the retention in the skeleton. Use of the values through reference 18, which includes mostly the chronic feedings gives an average of 0.0023% while the remainder of the values give 0.01%. The apparent increase in the more recent experiments is unexplained, although the presence of some values among this group of 0.002% and 0.003% indicate that the higher uptake is not general in nature. From this compilation we would feel that the ICRP value of $0.0027\%^{2}$ to the skeleton is reasonable as a realistic case. However, with the presence of the higher values in the table, we recommend a value of 0.05% for this calculation in order to hedge some of the uncertainties.

Because of the limited number of cases in which skeleton, liver and total tissue were measured, we did not attempt to find values by averaging. Instead, the weighted averages were used to obtain the ratio. In this procedure four

values, in which both the liver and skeleton were high, were eliminated since it was assumed that these values may have indicated the presence of cross-contamination. The skeleton on these cases used averaged 0.0063% while the liver averaged 0.0012% leading to a liver/skeleton ratio of 0.18 appreciably different from the ICRP value of 1.³⁴ However, such a ratio is believed to be more representative of the actual uptake in the two organs from oral intake. Data on the uptake in other tissues are sparse so that no estimate is made. Instead, because the skeleton and liver dose are of primary interest in environmental calculations, only the uptake and retention in these two organs is estimated. The values derived are 0.005% to the skeleton and 0.001% to the liver for the reasonable case. For a maximum case, we will consider the higher uptake values measured to be applicable and use the value of 0.02% to the bone and 0.004% to the liver.

There are additional data for the citrate administration over that given in Table D-XXXII but they are expressed as concentration in an organ or the quantity in a single bone. No attempt was made to reduce these data to a total organ basis. From the data available, it would appear that a larger fraction of the plutonium administered as citrate goes to the liver and the soft tissue than in the case of the nitrate administration. The averages for the animals listed are 0.06% to bone, 0.02% to liver and 0.04% to soft tissue.

The oxide data have not been reviewed to derive an uptake value because, in the practical case, there is no way of distinguishing between the oxide and other forms. We would feel, therefore, that it is more appropriate to use the value for the uptake and retention given for the nitrate.

The ²⁴¹Am data for skeleton and liver lead to uptakes of 0.01% and 0.005%, respectively. These values will be used as a reasonable case. The ICRP has recently adopted a new set of GI absorption values for use in occupational situations.³⁵ These are 10^{-5} for insoluble plutonium compounds and 10^{-4} for soluble plutonium compounds. For ²⁴¹Am, the uptake through the GI Tract is given as 5×10^{-4} . For both nuclides the partition between liver and skeleton is again taken as 45% to each as was used in ICRP-19.³⁴ These values are in reasonable agreement with the ones derived in this document although the americium uptake is somewhat higher in the new ICRP values.

It is noted in Table D-XXXIII that the uptake of ²³⁷Np nitrate approaches 1% in the skeleton and 0.1% in the liver. The urine is also an order magnitude or so higher than the other transuranium elements. In situations

where this nuclide occurs, these high absorption values may make it a significant contributor to the hazard.

D. Uptake in the Young Animal

There is a reasonable body of information on the uptake by the young animal, and the data all show a marked increase in the absorption and deposition in the organs over that measured for the adult. A summary of the data available is given in Table D-XXXIV. As can be seen, the uptake is on the order of 1 to 5% with indications from Table D-XXVII that there is a difference between 4 hours and 24 hours of age. While only a few points are available, ^{241}Am appears to behave in a similar manner. One experiment, on the change in the uptake of plutonium with age, is given in Table D-XX. There appears to be some difference between pre-weaning (20 days) and the post-weaning (21 days) in the data for total retention and retention by the femur for the animals sacrificed at 6 days post-feeding but this difference does not occur at the 4-hours post-feeding time.

TABLE D-XXXIV
RETENTION OF TRANSURANICS IN THE YOUNG RAT

Ref.	Table	Material (days)	Age at Gavage (days)	Time to Sacrifice (days)	<u>1 Administered Dose</u>		
					<u>Skeleton</u>	<u>Liver</u>	<u>Total</u>
<u>Plutonium</u>							
21	XVI	Nitrate	2	1	1.1	0.16	1.2
				8	1.56	0.21	1.8
22	XIX	Nitrate	2	7	2.6	0.3	2.9
23	XX	Citrate	1	0.17	-	0.14	0.43
				6	-	0.13	0.35
28	XXIV	Nitrate	2	7	2.7	0.2	2.2
					0.6	0.06	0.4
29	XXVI	Nitrate	-	7	1.3	0.14	2.9
29	XXVII	Nitrate	0.17	7	7.7	0.6	29
					3.4	0.4	4.8
					4.2	0.7	4.7
					4.7	0.9	5.9
			1	7	6.2	0.5	10
					3.3	0.24	2.7
					1.9	0.2	1.6
					2.1	0.4	2.4
<u>Americium</u>							
26	-	Chloride	7	1-4	-	-	0.88
27	XIX	Nitrate	2	7	4.5	0.3	2.9

The data on plutonium in the pig (Table D-XVII) indicate a much larger uptake, which ranges from 13 to 16% in the skeleton and 3 to 7% in the liver, with total retention (excluding the GI tract) of 25 - 80%. The pig data also indicates a significant retention of 15% to 25% of the gavaged dose remaining in the GI Tract at times as long as 10 days after administration. The possible significance of this type of retention to the dose to the GI Tract of the young individual is not clear at this time, but further work is required to assess whether this represents the behavior in the human better than the rat data as well as to determine the need for including a dose to the GI Tract for the young animal in the assessments.

The application of these data to the human is uncertain because the correspondence between the uptake at a given age in the human and these animals is unknown. There is apparently a decrease in the uptake with age of the animal to some point where the animal becomes an "adult" but this point in the human is not known. Very young humans have only indirect access to ingestion of environmental transuranium elements and their organ sizes increase with age providing some dilution effect. An assumption would be to consider the uptake to be 5% with a linear decrease in the uptake to one year of age.

E. Biologically Bound Transuranics

A possible increase in the uptake of transuranics that have been biologically incorporated into foodstuffs has been a question for some years. Data are now becoming available from studies at Battelle-Northwest with the information presently available given in Tables D-XXIII, D-XXV, D-XXVIII, D-XXIX, and D-XXX. Much of these data were obtained on only a few animals per experiment so that they have considerable uncertainty.

We believe that these data must be examined in light of the internal controls run with each experiment, rather than using uptake from other experiments with solution, as the basis. For this reason, we calculated the ratio of the average of each experimental point to its respective control in the given experiment. These ratios are given in Table D-XXIV.

As can be seen, the results are variable with the acute feeding ranging up to a ratio of 13 for rats and 4 for guinea pigs. The chronically fed guinea pigs, however, had ratios generally less than 1. This could have resulted from high controls in this particular experiment as well as from higher uptake from the incorporated or mixed foods.

TABLE D-XXIV

RATIO OF UPTAKE OF INCORPORATED OR MIXED PLUTONIUM TO CONTROLS

Material	Ratio			
	Skeleton	Liver	Carcass	Total
<u>Rats</u>				
2 Day Old Rat	10	2.3	-	-
Ileum	3	13	-	2
2 Day Old Rat	3	1	-	-
Incorporated-Leaves	-	8	11	12
Mixed-Leaves	-	3	5	5
<u>Guinea Pigs</u>				
Incorporated-Leaves & Stems	-	3	-	-
Mixed Leaves and Stems	-	0.6	-	-
Incorporated-Leaves	-	1	2	1
Mixed-Leaves	-	2	4	3
<u>Guinea Pigs-Chronic</u>				
Incorporated-Leaves and Stems	-	1		2
Mixed Leaves and Stems	-	0.3	0.2 ^a	0.5
Incorporated-Leaves	-	1	0.3	0.8
Mixed-Leaves	-	0.5	0.2	0.5

^aControls for animals fed leaves used.

It is impossible, at this time, to determine the degree of increase in uptake from biological incorporation or mixing of the plutonium with the food. The data would seem to indicate that mixing is less effective than incorporation, perhaps by about a factor of two. The data are inconclusive at this point so that a recommendation as to any allowance for incorporated plutonium cannot be given.

Only one experiment has been done with ^{241}Am and it shows an increase for incorporated material by about a factor of two over the nitrate feeding. Since this is within the range of the values for plutonium, we will again conclude that recommendations are not possible.

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APPENDIX E

RESUSPENSION

J. W. Healy

Resuspension from soils and subsequent inhalation of the resuspended material, has long been considered the chief source of exposure to transuranium elements deposited in soils. In spite of the obvious importance of this pathway, research has been limited so that a reasonable prediction of the resulting concentrations and inhalation is difficult to attain. In fact, Slinn¹ indicates that he does not trust resuspension factors, rates or velocities to within many orders of magnitude. In the following we will review the data available and attempt to arrive at as reasonable an answer as possible.

I. TYPES OF RESUSPENSION CONSIDERED

The overall resuspension problem can be divided into three types for conceptual understanding and calculations.^{2,3} These are: 1) wind-driven resuspension; 2) mechanical resuspension; and 3) local resuspension. For wind-resuspension the energy required to dislodge the particles arises from the wind and the particles then disperse downwind depositing on surfaces at a rate depending upon the aerodynamic properties of the particles and the nature of the terrain. Both mechanical resuspension and local resuspension result from mechanical disturbance of the soil. However, in the mechanical resuspension case the concern is with concentrations downwind following dispersion and deposition, while the local resuspension describes the exposure in the immediate vicinity of the individual before dispersion occurs.

Another type of resuspension that will be considered only briefly is transfer resuspension. This involves the transfer of the contaminant from its place of deposit to another place where inhalation may be more probable. Unfortunately data to describe this type of resuspension are extremely limited.

Each of these types of resuspension will be considered in deriving the best estimate of exposure to an individual in an area contaminated by the spread of transuranics in the soil.

II. RESUSPENSION MODELING

There are three basic techniques in use for resuspension modeling: the

resuspension factor; 2) the resuspension rate; and 3) the mass loading approach. Each method has its strengths and its weaknesses, particularly in view of the state of the technology at this time. We will describe each of the techniques and discuss its advantages and disadvantages.

A. The Resuspension Factor

The resuspension factor is defined as the ratio of the concentration in the air at a reference height (usually one meter) to the quantity of the contaminant per unit area on the surface of the ground. The usual units are m^{-1} . Its chief advantages are its simplicity and the fact that most measurements in the past have been expressed in this form so that values are available for calculation. The chief disadvantages are that it is a completely empirical formulation so that it is difficult to extrapolate from one terrain to another and it ignores the distribution of the contamination over the area and the size of the contaminated area involved. Thus, the denominator contains the local quantity on the ground while the numerator is an undefined function of the air concentration resulting from upwind contaminated areas and activities.

A problem common to this model, as well as all others, is the uncertainty resulting from the depth to be used in assessing the quantity per unit area to be used in the denominator. For a uniform profile in the soils, the quantity per unit area will increase in direct proportion to the depth of the sample used. For non-uniform profiles the estimate of the quantity per unit area will also change depending upon depth used. For wind resuspension it can be assumed that the appropriate depth is small, perhaps a millimeter or less, although it is likely that this depth may be variable depending upon wind speed and the degree of saltation allowed by the size of the area and the nature of the soils. For mechanical disturbance, the depth will be some function of the depth to which the disturbance occurs; the function depending upon the relative ease with which the particles can escape from the soil to the atmosphere.

Mishima⁴ has tabulated resuspension factors measured over a variety of conditions. It is frequently noted that these values range over eleven orders of magnitude. However, the values quoted represent both outdoor and indoor conditions, with and without mechanical disturbance, and at various times after the contaminant has been deposited. In a brief review of Mishima's table it is noted that the values for mechanical disturbance range from about $2 \times 10^{-6} \text{m}^{-1}$ to $7 \times 10^{-5} \text{m}^{-1}$ (with one value of 10^{-3}m^{-1} based on uranium contaminant with dust

stirred up and sampling at a height of 1 foot, ignored). For periods of no activity, with relatively freshly deposited material, the values generally range from 10^{-8} m^{-1} to $2 \times 10^{-6} \text{ m}^{-1}$ while for aged material they range from $6 \times 10^{-10} \text{ m}^{-1}$ to 10^{-13} m^{-1} . It is difficult to generalize these numbers because the exact value will depend upon the degree of disturbance, the placement of the sampler, the meteorological conditions at the time and the nature of the soils. However, many of the measurements were made in desert areas with low soil moisture where resuspension would be expected to be highest.

An apparent reduction in the resuspension factor with time was indicated by Wilson et al.⁵ from measurements at the Nevada Test Site. Here samples were taken at three different distances from the center of a safety shot starting about one month after the contamination occurred. The authors noted that the data were too erratic to establish half-times for the decay of the air concentration beyond a very crude estimate. This estimate was made by plotting the medians of the data at each sampling distance and provided a value of five weeks for the concentration half-time. This value was used by Langham⁶ in assessing future hazards from plutonium contamination. A somewhat larger half-life of 45 days was used by Kathren⁷ in a study of acceptable contamination levels for plutonium in soils. Anspaugh et al.⁸ measured the decrease in air concentration with time immediately following a cratering event in Nevada and following the venting of an underground shot. For the cratering event, sampling was carried out for six weeks following the event with a measured half-time of 38 days. The venting experiment started three months after the event and continued for 9-10 months with the most predominant radionuclides being the isotopes of ruthenium. Here a half-life of 66 days was found.

However, the consequences of continuing such half-times over a long period were pointed out by Healy⁹ and Anspaugh¹⁰. Healy noted that samples taken at the same location as those by Wilson et al., but about one year after the conclusion of the Wilson study,¹¹ gave values up to several orders of magnitude greater than would be predicted by the 35 day half-life. Anspaugh¹⁰ indicated that the functional nature of the decrease in resuspension rate with time cannot be confidently extrapolated, and previously published models should not be applied to calculations many years after the contaminating event. He also cited two sets of measurements at the Nevada Test Site where the area had been contaminated with plutonium by high explosive detonations some 20 years earlier. These studies gave values for the resuspension factor of $3 \times 10^{-10} \text{ m}^{-1}$ and

$2 \times 10^{-9} \text{ m}^{-1}$. These data indicate unequivocally that resuspension does occur after this period of time although predictions using the short half-life following deposition would result in unmeasurable values of air concentration.

Anspaugh et al.¹² used the available information to derive an empirical expression for the resuspension factor that allows the resuspension factor to decrease with time. In their derivation they used four constraints; 1) the apparent half-time of decrease during the first ten weeks should approximate a value of 5 weeks; 2) this half-life should about double over the next 30 weeks; 3) the initial resuspension factor should be 10^{-4} m^{-1} ; and 4) the resuspension factor 17 years after the contaminating event should approximate 10^{-9} m^{-1} . The value for the resuspension factor in the aged source resulted from 23 individual air concentration measurements at a location contaminated with plutonium 17 years earlier with the average resuspension factor found to be 10^{-9} m^{-1} . An expression that approximates these constraints is given as

$$R(t) = 10^{-4} \exp(-0.15\sqrt{t}) + 10^{-9}, \quad (1)$$

where $R(t)$ is the resuspension factor (m^{-1}) and t is the time since the contaminating event (days).

Oksza-Chocimowski¹³ provided a "generalized" model for the change in half-time of the resuspension factor which allows the ratio of the resuspension factor at time zero to the resuspension factor at a long time [$R(0)/R(=)$] to vary.

$$T_{1/2}(t) = A \ln \left(1 + B + Ct^D \right) + \frac{t \ln 2}{\ln \frac{R(0)}{R(=)}}, \quad (2)$$

where

- A = constant coefficient (days),
- B = constant,
- C = constant coefficient (days^{-D}),
- D = constant exponent,
- $R(0)$ = initial resuspension factor, m^{-1} ,
- $R(=)$ = final resuspension factor, m^{-1} ,
- t = time since contaminating event (days).

This expression was evaluated for a range of values of the constants with limiting values corresponding to ratios of $k(0)/R(=)$ of 10 and 10^{11} . However, it would appear that data are not available to make adequate choices for a given area.

In both of these models, the constants are evaluated based upon data from a limited number of areas and conditions. In Anspaugh's model, for example, it is apparent that the final resuspension factor after long aging is based upon wind resuspension only in a desert area. Whether disturbance in the area would cause an increase is unknown although it appears likely to do so. Sehmel¹⁴ made measurements down-wind from the original oil-storage pad at Rocky Flats and related the concentrations found to the 2.1 power of the wind speed. Subsequent measurements were higher, however, due to the digging of a ditch between the oil storage area and the sampler. This work also involved increased vehicular activity. This disturbance caused about an order of magnitude increase in concentrations at the samplers which persisted after the work was complete, presumably because of a change in the character of the surface. The half-time for decrease over the next seven months appeared to be about nine months. Thus, it is possible that disturbance can not only increase the concentration at the time that it occurs, but could result in increased wind-driven resuspension factors for some time thereafter.

Sehmel^{15,16} has also provided data on the wind-borne resuspension of a submicrometer tracer, calcium molybdate, that was applied to a test area by spraying. In this experiment, using cascade impactors arranged to operate in different wind conditions, he noted that the resuspension factor at 1.8 meter height increased as the 6.5 power of the wind speed. However, this slope was determined by drawing the line through the bottom end of uneven wind speed ranges; a procedure that could well result in an overestimate. He also noted, for this system, that there seemed to be little, if any, decrease in the resuspension over a period of three years. Whether this is due to a "preaging" by the application in water is unknown.

In concluding the discussion of the resuspension factor, it is apparent that this empirical approach does not inherently incorporate many of the variables and present estimates are relatively crude. In particular, the present estimates appear to be based upon short term experiments with little attempt to provide a factor applicable to long-term (say one year) averages.

B. The Resuspension Rate

The resuspension rate is defined as the fraction of the contaminant present on the ground that is resuspended per unit time by either winds or mechanical disturbance. Once obtained, it can be used to describe concentrations at any point around a non-uniform contaminated area by the use of point-source dispersion and deposition equations and integration over the area. This potential use was illustrated by the application to an area contaminated with plutonium by a safety shot⁹ and the inverse use at the same area to obtain resuspension rates from measured air concentrations.¹⁷ It was introduced for use in resuspension calculations by Healy and Fuquay,¹⁸ although in a crude form.

Slinn¹ has pointed out that the resuspension rate can be converted to a resuspension velocity by multiplying by the ratio of the quantity of contaminant per unit area and dividing by the volumetric concentration of the contaminant in the soil. Such a velocity is analogous to the deposition velocity with, however, a negative sign when conditions are such that net resuspension occurs.

Resuspension rates have been measured for a given area by three techniques: 1) measurement of air concentrations resulting from a known pattern of a tracer material on the ground and inferring the resuspension rate from height profiles giving the total transport or from use of dispersion equations in known meteorology; 2) measurement of air concentrations from an existing contaminated area and obtaining the resuspension rate as given in 1; and 3) measurement of natural dust fluxes and relating these fluxes to some association between the concentration of the contaminant in the soil and the dust flux. The last method has been used only for wind resuspension.

1. Wind Resuspension

A detailed body of knowledge exists on the mechanisms of the movement of soils by wind through the classic studies of Bagnold¹⁹ on desert sands and the detailed studies of Chepil and others^{20,22-33} on agricultural soils. We will not review these studies in detail since much of the information is applicable to the limited condition of erosion of erodible soils. There are, however, data and concepts applicable to the resuspension process, at least for the limited conditions of agricultural soil, and a brief review of these is in order.

The relationship between erosion and winds is complex with a large number of variables affecting the outcome. Chepil²⁰ listed the most important of the

factors as related to three categories. These are given in Table 1. In the following discussion we will briefly describe some of the more important findings applicable to the general problem of resuspension from the extensive work on soil erosion.

Soil movement across an eroding field is primarily from movement of the smaller particles, usually less than about 1 mm in size. There are three mechanisms for movement with the particular size for each somewhat dependent upon the wind speed. The heaviest particles move by surface creep or movement along the surface. Chepil²⁰ noted that these grains were too heavy to be moved by the direct pressure of the wind but were propelled by the impacts of smaller grains moving in the second method of movement, saltation. In saltation the grains, after being rolled by the winds, suddenly leap almost vertically. Some grains rise only a short distance while others, depending upon the wind speed can rise to several feet. They are then carried forward by the winds and settle by gravity until they strike the ground. Chepil²⁰ attributes this sudden rise to the spinning of the grain as it rolls along the surface with the Bernoulli effect causing a difference in pressure at the top and bottom of the grain. The third method of movement is, then, suspension of the small soil particles by the wind. In the latter case the particles must be small enough to be kept airborne by the turbulent forces in the atmosphere overcoming the force of gravity. Since the turbulence varies with atmospheric stability and, to some extent with

TABLE I
MOST IMPORTANT FACTORS IN WIND EROSION
OF AGRICULTURAL SOILS - AFTER CHEPIL²⁰

I. Air	II. Ground	III. Soil
1. Velocity	1. Roughness	1. Structure
2. Turbulence	2. Cover	a. Organic Matter
3. Density	3. Obstructions	b. Lime Content
a. Temperature	4. Temperature	c. Texture
b. Pressure	5. Topographic	2. Specific Gravity
c. Humidity	Features	3. Moisture Content
4. Viscosity		

wind speed, one would expect larger particles to be suspended in strong turbulent winds and that these particles would settle out as the winds decrease. However, the fine particles can remain suspended for long times and can cover large distances. It is the suspension fraction, and particularly the smaller particles, that is of interest in resuspension since resuspension is defined as the suspension of a previously deposited contaminant.

The air flow characteristics over the surface of the soil are important in transmitting force to the soil grains and in determining the velocity at which they start moving. In a neutral atmosphere (i.e., temperature decrease with height is adiabatic) the velocity profile is logarithmic with height so that one can write²¹

$$u_z = \frac{u_0}{K} \ln \left(\frac{z}{z_0} \right) . \quad (3)$$

Here, u_z is the wind speed at a height z , z_0 is the height at which the wind speed is zero, u_0 is the friction velocity (or drag velocity) and K is the Van Karman constant which has been found by integration to equal 0.4. z_0 is a characteristic of the surface, increasing as the roughness of the surface increases. The friction velocity is of importance in determining the force exerted on any object protruding above the laminar layer of the atmosphere and has been shown to be the characteristic wind speed that affects soil movement.^{19, 22, 23}

Bagnold¹⁹ has shown that in severe erosion conditions, the profile of wind speed with height is changed by the momentum transfer to the particles in saltation. In order to induce movement of the soil it has been observed that the wind velocity must exceed some threshold value. Chapil²² has studied the movement of particular sizes of grains and has noted that the curve of grain diameter times the specific gravity versus threshold friction velocity for initial movement has a minimum at about 0.15 mm (150 μ m), with a friction velocity of about 0.15 m/sec. That is, grains smaller than this size and grains larger than this size require higher friction velocities to initiate any movement. The stability of the finer grains is illustrated by Bagnold's¹⁹ simple experiment of placing a pile of talcum powder on a smooth surface and exposing it to winds. It will be found that the layer is stable at relatively

high wind speeds. However, a few grains of larger particles sprinkled on the pile will result in rapid dispersion at a wind speed much lower than would serve to disperse the particles without this added factor. It is believed that the relative stability of the small particles is due to the fact that they do not protrude above the laminar layer so that no drag force is exerted upon them. The minimum in the curve, then, is due to the balance between the increasing drag force as the particle increases in size and the increasing downwind force from gravity as the particle becomes larger. Above about 0.15-0.2 mm the threshold velocity required to initiate movement increases as the square root of the product of the particle diameter and its specific gravity.^{19,20}

As a result of this threshold friction velocity curve, it is apparent that direct aerodynamic pickup of small particle sizes from the soil is unlikely. Instead, the process of saltation is the key to producing the suspended fraction because the impact of these particles as they strike the ground provides the energy to propel the smaller particles above the laminar layer into the wind stream where they are transported by eddies in the wind. Thus, in the talcum powder example, the sand particles sprinkled on the talcum powder serve the function of the saltating particles. In a field, knolls, ridges, sand pockets, or other areas most exposed to the wind and/or containing the easily erodible grains, start to erode at a lower velocity than the rest of the field. Once it starts it spreads downwind with the bombarding action of the particles in saltation causing movement in other parts of the field that normally would not be eroded.²¹ The threshold velocity of the field is, therefore, the threshold of the most exposed or most erosive spots in the field. Since the avalanching effect of saltation increases down the field in the direction of the wind, the length of the field is also a factor in the degree of erosion.

An important consequence of the role of saltation in the production of resuspension is that there will be no dust, or particles flowing in suspension, unless the wind speeds are great enough to produce saltation under the conditions of the field. This places a threshold condition on the wind speeds required to resuspend particles from the ground.

Bagnold¹⁹ measured the rate of soil flow for desert sands and found that these rates could be described by an equation of the form of

$$q = C u_*^3 (\rho/g),$$

where q is the rate of soil flow in grams per centimeter width per second, ρ is the density of the air, g is the acceleration of gravity, and C is a constant which differs for differing soils and forms of erosion. On the desert sands, Bagnold concluded that the flow in suspension was small as compared to saltation and surface creep, being only about 1/20th of the total flow. Chepil^{20,23} made similar measurements on agricultural soils in a wind tunnel. His results indicated that all three methods of flow appear to follow Bagnold's law of the cube of the wind velocity, at least for the soils tested, a fine sandy loam and a heavy clay soil. The constant C for total soil flow varied widely for different soils with the range found in these experiments being 1.0 to 3.1. Chepil also measured the proportion of each type of flow on four widely different soils. These results are given in Table II.

It is apparent that the fraction of the total flow carried in suspension on agricultural soils is considerably higher than on desert sands (presumably because of the availability of the smaller particles). These studies showed that the logarithm of the flow plotted against height was essentially a straight line and that relative concentrations of soil particles at different heights remained the same with wind velocities ranging from 13 to 30 miles per hour (6 - 13.5 meters/seconds). Presumably, then, the relative flows also remained constant.

Chepil²³ also provided the sizes of the particles in the soils studied between <0.1 and 0.83 mm. A plot of the relative suspension flow against the fraction of particles <0.1 mm is given in Figure 1. The use of any other particle size range or cumulative percentages gave erratic results. This may

TABLE II
RELATIVE PORTION OF THREE TYPES
OF FLOW ON DIFFERENT SOILS (AFTER CHEPIL²³)

Soil Type	% Of Flow In		
	Surface Creep	Saltation	Suspension
Soepte Heavy Clay	24.9	71.9	3.2
Haverhill Loam	7.4	54.5	38.1
Hatton Fine Sandy Loam	12.7	54.7	32.6
Fine Dune Sand	15.7	67.7	16.6

Fig. 1. Suspension flow versus percent of grains in soil <0.1 mm.

indicate the importance of the fraction of the smaller particles in the soil in producing the suspension fraction.

An important factor in the suspension fraction is the aggregate state of the smaller particles in the soil. Particles in the submicron range rarely exist as such in the soils because they tend to either clump together or to adhere to larger particles resulting in small aggregates. In fact Chepil²² states that particles smaller than 0.005 mm (5 μ m) do not exist as such in ordinary soils, for they are aggregated into larger individual grains. He adds also that single grains or aggregates 0.05-0.5 mm in diameter have little or no cohesive properties and are easily carried by the winds. This means that contaminants in the soils, either as fine particulates or absorbed on the surface of soil particles will largely exist as soil aggregates and will behave in the same manner as the soil. Chepil²⁴ demonstrated the aggregation of material carried in suspension at heights of 4 to 8 feet in a dust storm by

sizing particles by sedimentation in CCl_4 , a non-polar solvent that tends to preserve aggregates, and then repeating in water following dispersion with sodium hexametaphosphate. The curves show the percentage of particles smaller than a given value reaching zero at about 5 μm diameter in a 1954 storm and about 10 μm in a 1955 storm. By contrast, the dispersed samples showed 15-25% of the particles smaller than 5 μm .

An important factor in aggregation is the moisture content of the soil. This has been investigated by Chepil²⁵ and by Bisal and Hsieh.²⁶ Chepil²⁵ has provided a formulation for the soil flow taking into account the increased resistance to movement due to the cohesion of the absorbed water films. However, it is noted that the water content of a field can decrease rapidly following a rain due to the drying actions of the winds.

There are many other factors that influence the erosion of the field such as the presence or absence of ridges, the quantity of vegetative cover and the presence or absence of a surface crust. These have been combined to give a soil erosion equation. 27,28

$$E = f(I', C', K', L', V). \quad (5)$$

Where E is the potential average annual soil loss in tons per year; I' is the soil and knoll erodibility; C' is the local wind erosion climatic factor; K' is the soil ridge roughness factor; L' is the field length factor and V is the equivalent quantity of vegetation. There have been mathematical relations established between the individual variables. The relationships, however, are so complex that the individual factors are evaluated separately in a form where combinations of the factors can be evaluated graphically.

We will not attempt, here, a complete discussion of this equation along with numerical factors, but will discuss each of the factors in a qualitative fashion because it appears that many of these could be of importance in any case of wind resuspension.

The soil and knoll erodibility consists of two terms. The first, the soil erodibility index is related to the percentage of dry aggregates greater than 0.84 mm in diameter. The higher this percentage the lower the soil erodibility. Conversely, the higher the percentage of aggregates less than 0.84 mm in

diameter the greater the erodibility. The knoll erodibility is expressed as the percent of the level ground erosion that occurs at various slopes of the knolls in the field. This factor ranges from 0 for a level field to about 650% at the top of a knoll having a slope of 10% and about 360% from that portion of the windward slope where the drag velocity is the same as the top of the knoll (about the upper third of the slope). A surface crust stability factor is usually ignored because the crust disintegrates rapidly because of abrasion once the wind erosion starts.

The soil ridge roughness is a measure of the surface roughness other than that caused by clods or vegetation. It has been found that ridges of 2-4 inches in height are the most effective in controlling erosion with the erosion for ridges of this height about 50% of that over a smooth surface.

The wind erosion climatic factor includes the influence of wind speed and moisture. The rate of soil movement varies directly as the cube of the wind velocity. In this factor the mean annual wind velocity, corrected to a standard height of 30 feet, is used. Since atmospheric wind velocities are normally distributed, the probability of obtaining high winds is higher with higher mean velocities. The rate of soil movement varies approximately as the square of effective soil moisture. The wind erosion climatic factor has been given for a number of locations by the U. S. Department of Agriculture²⁹ for each month of the year. To illustrate the differences in this factor from one place to another, we have given rough ranges for four locations. For Washington State, the climatic index varies from about 1 to 50 with the highest values in the months of March through May. For eastern New Mexico along the north-eastern border, the climatic factor remains the highest in the nation throughout the year. Here, values range from about 70 to 300. For the state of Ohio the factor ranges from 1-10 and for Georgia about 1-5. Thus, there are widely differing wind and moisture factors throughout the country with the east, in particular, having low factors as compared to the west. It could be predicted that the resuspension of contaminants from the soil will also be lower in the areas of low climatic factor for erosion.

The field length factor again has two parts: The distance across the field and the sheltered distance. The distance across the field is measured along the prevailing wind erosion direction. On an unprotected field the rate of soil flow is zero on the windward edge and increases with distance downwind until, for a large field, the flow reaches a maximum that the wind of a given velocity can

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The field length factor again has two parts: The distance across the field and the sheltered distance. The distance across the field is measured along the prevailing wind erosion direction. On an unprotected field the rate of soil flow is zero on the windward edge and increases with distance downwind until, for a large field, the flow reaches a maximum that the wind of a given velocity can

maintain. The sheltered distance is that distance along the prevailing wind erosion direction that is sheltered by any barrier.

Vegetative cover is an important factor in controlling wind erosion. Three different factors are included in the equivalent quantity of vegetative cover. The first is the quantity of the vegetative cover expressed as clean, air-dried residue. The second denotes the total cross-sectional area of the vegetative material. The finer the material and the greater the surface area, the more it reduces the wind velocity and the more it reduces wind erosion. The third factor is the orientation of the cover. The more erect the vegetation, the higher it stands above the ground, the more it reduces wind velocity near the ground and the lower the erosion.

Gillette and his collaborators have been studying the vertical flux of dust from agricultural soils under the influence of the winds.³⁴⁻³⁹ In a study of particle size distribution at 1.5 and 6.0 m along with the horizontal velocity, it was concluded that the upper limit of the ratio of the settling velocity to the friction velocity for aerosols having the potential for long range transport is approximately 0.2 or slightly greater.³⁷ Also by studying the sizes of the aerosols produced at different values of the friction velocity, it was concluded that the dominant injection mechanism of soil aggregates <10 μm in diameter is not direct aerodynamic entrainment.³⁷ It was noted that the size distribution of the vertical flux (expressed as $dN/d(\log r)$) was proportional to r^2 for particle sizes greater than 2 μm . A similar particle size distribution was noted for the parent soil when the size distribution was measured with liquid freon dispersal. Since the dielectric constant of liquid freon is close to that of air it tends to preserve the aggregate state of the soil. However, if the aggregates are broken up, the number of particles less than 10 μm greatly increases,³⁷ showing once again the importance of aggregation of the smaller particles in the soil.

In a study of the vertical flux above several soils, Gillette³⁶ noted that the production of a flux of particles <10 μm in size increased more rapidly with friction velocity on a soil that had a higher percentage of silt and clay than it did on a soil with a relatively low percentage of these materials. On both soils, extrapolation of the curves to the point of intersection indicates a threshold friction velocity of about 0.18 m/sec, about the same value as for the impact threshold discussed earlier. For the soils containing 3.5% clay (<1 μm) and either 0.5 or 1.0% silt (<25 μm) the vertical flux of particles <1 μm

increased as the ratio of the friction velocity to the threshold friction velocity raised to the 5.14 power. For the soil containing 3.5% clay and 1% silt, the vertical flux increased as the 9.62 power of the ratio of friction velocity to the threshold friction velocity. This provides a model for the vertical flux of

$$F_A = \text{Const.} (U_e / U_{e \text{ Threshold}})^P. \quad (6)$$

For the two soils evaluated, the constant at the point of intersection was about $6 \times 10^{11} \text{ cm}^3 \text{ sec}^{-1} \text{ cm}^2$ or, assuming an aggregate density of 2.5 g/cm^3 , about $1.5 \times 10^{-6} \text{ g sec}^{-1} \text{ m}^{-2}$. An important conclusion from these studies was that the increase in the vertical flux at powers of the friction velocity much greater than the observed cube for the horizontal flux was due to the break-up of the aggregates by sandblasting and release of the smaller particles to suspension.

In a further study of the vertical flux resulting from eight different soils,³⁹ a regular pattern of increase with friction velocity appeared to exist with, however considerable spread. A curve fitted by eye by the present author to the bulk of the data indicated an average increase of vertical flux as the fifth power of the friction velocity with a constant at the assumed threshold friction velocity as given by the earlier study of two soils. A comparison of the size distribution of a parent, sandy-soil with the size distribution of the horizontal flux to a height of 1.3 cm indicated the size distribution of the horizontal flux is very similar to the size distribution of the parent soil aggregates. The aerosol at 1 meter height showed a mode for the particles greater than $20 \text{ }\mu\text{m}$ centered around $50 \text{ }\mu\text{m}$. As height increases the particles less than $20 \text{ }\mu\text{m}$ become an increasingly larger proportion of total aerosol. It was also noted that the concentration of larger particles increases with the windspeed.

Travis^{40,41} has developed a model for the redistribution of wind-eroding, soil-contaminant mixtures using the Gillette relationships for the vertical and horizontal flux.^{34,35} This model assumes that the contaminant is closely associated with the soil aggregates and moves in the same manner as the soil. Since it incorporates only studies from eroding agricultural soils, it should be limited to these soil conditions.

Shinn et al.⁴² made measurements of the vertical profile of dust in the atmosphere at two sites and related these profiles to the vertical dust flux by the eddy-correlation method. The two sites were an area at the Nevada Test Site where plutonium contamination had occurred during a series of non-nuclear tests over 20 years earlier (GMX Area) and in an agricultural field in West Texas. Measurements were made simultaneously of the dust flux by an optical particle detector and the mean wind and temperature profiles.

It was noted that the mass-concentration particle size distributions at both sites had a maximum at about 4 or 5 μm (MMAD) and this decreased by an order of magnitude at 1 μm and 10 μm . This distribution does not agree with Chepil's⁴³ or Sehmel's⁴⁴ data that show significant quantities of particles much greater than 10 μm at elevations up to 30 m above the ground. Shinn⁴¹ indicates that this could be due to reaggregation after sampling because the sedimentation velocity of such particles would be greater than u_g . However, this remains an uncertainty that requires investigation.

The wind profile measurements showed the roughness length (z_0) at the Texas site to be 0.044 cm and at the GMX site to be 2.0 cm. This resulted in a drag coefficient referenced to the wind speed at 2 m, (u_0/u_{200}), to be 0.05 at Texas and 0.10 at the GMX site. It was noted that the dust profiles in this study, as well as in previous ones, fit a power law with exponents of either -0.2 or -0.35. This is due to the fact that all are nearly bare surfaces and the measurements were made in dynamic neutral atmospheric stability conditions. The dust flux calculation was parameterized by several simple relations.

$$F = K \frac{dx}{dz}, \quad (7)$$

where F is the flux, Z is height, x is the dust concentration, and K is the eddy diffusivity. Since

$$K = u_g K_z, \quad (8)$$

where k is the Van Karman constant ($k=0.4$). Since the dust concentration follows a power-law distribution with height

$$\frac{dx}{dz} = \frac{P}{z} x \quad (9)$$

Since the power, P , is about 3 and the concentration over the heights from 0.7 m to 2 m deviates only about 20% from the 1 m reference velocity, one obtains, by combining the above

$$F \approx -0.12 u_* x_1 \quad (10)$$

The data at GMX and Texas give values for x_1 of

$$\text{GMX } x_1 = 6.1 u_*^{2.09} \text{ and} \quad (11)$$

$$\text{Texas } x_1 = 522 u_*^{6.37} \quad (12)$$

These values then result in fluxes of

$$\text{GMX } F = 0.73 u_*^{3.09} \quad (13)$$

$$\text{Texas } F = 62.6 u_*^{7.38} \quad (14)$$

A tentative model of the upward dust flux was derived from profile and soil erosion data from a number of locations. This was titled the Gillette and Shinn model and is expressed by:

$$F = -F_0(u_0/u_0)^{\gamma+1}.$$

(15)

Where F is the flux, the reference wind speed $= 1$ m/sec, F_0 is a reference dust flux at $U_0 = 0$ and γ is the power in the dust profile. From a series of experiments by Gillette³⁸ a tentative relationship between the parameters in the Gillette-Shinn Model and the soil erosion index was derived. This is given in Figure 2.

An initial attempt to assess resuspension from the ground was made by Healy and Fuquay¹⁸ and Healy⁹ using data from Hilst^{45,46} who was using zinc sulfide particles to estimate the effects of various surfaces on wind erosion. At this time, the high rate of increase with wind speed due to breakup of aggregates was not known and it was assumed that the rate increased as the square of the wind speed. Later, the rates were converted to a cube relation with the wind speed,² although it was noted that, for these results, the square appeared to give less

Fig. 2. Tentative Relation for Parameterization of Dust Flux for the Gillette-Shinn Model.

variance in the data. This indicated a pickup rate of $5 \times 10^{-9} u^3$ per second. Since the wind speeds were measured at a 2 m height⁴⁵ and the Hanford area, the site of the experiments, has a z_0 of about 2 cm, this value would be about $4 \times 10^{-11} u^3$ per second. This can be only a preliminary estimate, however because of the alteration of the natural roughness feature by the change in courses.

Schmel^{15,47,48} has studied the resuspension of a tracer, submicrometer calcium molybdate, sprayed as a suspension over a lightly vegetated area with a roughness height of 3.4 cm. The area sprayed was within a circle of 29 m radius with a sampling tower in the middle. The average surface concentration on the ground was 0.62 g of molybdenum per square meter. Measurements were made with a cowled impactor, which always faced into the wind, at heights to 6.1 meters with some measurements made at specific wind speeds as measured at a height of 2.1 m above the ground. Resuspension rates were calculated from a mass balance calculated from the profile. Those particles depositing in the cowl were considered as "non-respirable" while those entering the impactor were considered as "respirable." The impactor separated the "respirable" particles into nominal diameters for unit density spheres of 7, 3.3, 2.0, 1.1 μ m and smaller particles on the back-up filters.

In these experiments, the resuspension rates ranged from about 10^{-11} to 10^{-7} sec^{-1} .⁴⁷ Plots of all of the data showed that the resuspension rates increased with the 1.0 to 4.8 power of the wind speed. However, these results included wind intervals with wide speed increments so that an uncertainty as to the actual wind speed to be used existed. When these were eliminated, the resuspension rates for all sizes in the impactor and the back-up filter increased with wind speed to the 4.8 power. It may be noted, however, that these experiments were run over a finite period of time so that differences in stability would occur. This would cause differences in the wind profile and a change in u_g . Thus, if the majority of the lower speed winds were in the stable condition, where u_g would be lower than for the neutral condition, and if the majority of the higher speed winds were in the unstable condition, where u_g was greater than for the neutral condition, an exaggeration of the slope of the curve would occur. Since lower wind speeds frequently occur in the stable condition with higher wind speeds in the unstable condition, it can be postulated that such an effect has influenced these relationships, albeit to an unknown degree. It is noted, however, that dust loadings by the same technique

appear to increase as the 2.9 power of the wind speed for the higher wind speeds.⁴⁸

The data from the back-up filters which showed an increase as the 4.8 power of the wind speed and for which both the small interval and wide interval of sampling fitted the curve gave a fit to

$$R.R. = 1.96 \times 10^{-13} u^{4.82}, \quad (16)$$

where R.R. is the resuspension rate. If one assumes that the logarithmic wind profile existed throughout the period this becomes:

$$R.R. = 2 \times 10^{-16} u_e^{4.82}, \quad (17)$$

when the threshold velocity is ignored.

Schmel⁴⁹ also measured the resuspension rate of zinc sulfide tracer particles from an asphalt surface. Average resuspension rates were determined to range from 5×10^{-9} to $6 \times 10^{-8} \text{ sec}^{-1}$ for average wind speeds from 2 to 9 mph (0.9 to 4.2 m/s). The dependency of resuspension rate upon wind speed was not determined but there was some indication that wind gusts greater than about 15 mph (7 m/s) rapidly suspended particles.

In a similar experiment, but in an area of cheat grass, a resuspension rate of $3.4 \times 10^{-8} \text{ s}^{-1}$ occurred in an area of surface roughness of 4 cm and a friction velocity of 0.52 m/s.⁵⁰ After truck traffic that removed 0.35% of the ZnS, the surface roughness was reduced to 3 cm and a resuspension rate of $1.25 \times 10^{-9} \text{ s}^{-1}$ was measured in a friction velocity of 0.51 m/s.

Schmel⁵¹ measured the resuspension rate of 10 μm diameter uranine particles deposited on the inner surface of an aluminum tube with a 2.93 cm inside diameter. The resuspension rates range from 10^{-6} to 10^{-3} s^{-1} and were dependent upon air flow rates and resuspension time. Orgill et al.⁵² measured the resuspension of DDT from forests in the Pacific Northwest. The DDT was sprayed in the early morning with low wind speeds and sampling was conducted by an aircraft-mounted sampler for five days. Calculated resuspension rates on three

of the sampling days were 1.0×10^{-8} , 2.5×10^{-8} and 7.7×10^{-8} . Sehmel^{48,50} proposed a correlation between resuspension rate and the roughness height z_0 using the data from the experiments on the aluminum tube, the ZnS from the asphalt surface, the Mo tracer from desert soil and the DDT from the forest. This curve shows a decrease in resuspension rate from the aluminum tube data to the Mo tracer with the asphalt surface falling on the lines between these two points. However, the DDT from the forest, with a large roughness height, had an increase by 2.5 orders of magnitude from the soil data. It is not clear how these resuspension rate data were corrected for the differing wind speeds or values of u_g that existed in each of the experiments. It is further noted that the surfaces and, possibly, mechanisms of wind pickup from the surface were different. For example, the resuspension of DDT from the forest could have been primarily as a result of the mechanical movement of leaves, needles and branches rather than the types of force found on the soil surface.

There have been many measurements of the air concentration in or near an area contaminated with radioactive materials, but most of these are not suitable for estimating resuspension rates or dependency on wind speed because of the lack of detailed meteorological data at the time of the measurement or the lack of a detailed knowledge of the configuration and contamination level of the source. Anspaugh et al.^{17,53} studied the resuspension of plutonium at the GMX area in Nevada using an ultra-high flow rate sampler so that samples could be obtained in a relatively short time while meteorological conditions were relatively consistent. The resuspension rates measured ranged from 2.7×10^{-12} to $4.8 \times 10^{-10} \text{ s}^{-1}$. There was a strong correlation between the resuspension rate and u_g^3 . Dividing the resuspension rate by u_g^3 greatly reduced the variance found in the resuspension rate itself. This result is consistent with Shinn's value for the dependence of dust flux on u_g at this location. The value of the normalized ratio to u_g^3 ranged from 1.5×10^{-11} to 10^{-9} . It is noted that the pattern of plutonium in the soil used for these derivations was determined by FIDLER measurements of the ^{241}Am gamma⁹ converted to plutonium concentrations by statistical comparison of the plutonium and americium content of soil samples.⁵⁴ This should provide an estimate of the plutonium in the surface layers of the soil so that correction for plutonium that has migrated to some depth is not required.

The dust samples used by Shinn⁴² in parameterization of the dust flux in this area were also analyzed for plutonium.⁵³ The profile of plutonium

concentration at a distance 100 m from ground zero agreed with the shape of the dust flux to a height of about 1 m and then showed lower concentrations than expected. At the 730 meter point the deviation is smaller. The deviations were as would be expected for a limited source with rather abrupt discontinuities. However, the close fit of the plutonium concentrations to the dust profile at the lower two heights indicates that the plutonium concentration at a height <1 m is closely coupled to the ground concentration even though the soil contamination is less by 2 orders of magnitude at the greater distance downwind.

Saltation fluxes were also measured at the GMX Area.⁵³ Values ranged from 3×10^{-7} to 8×10^{-6} g cm⁻² sec⁻¹ which are 10^{-3} to 10^{-4} of those measured by Chapil for wind-eroded fields. In this connection, Shinn⁵⁶ points out that the Nevada Test Site, the location of the GMX Area, is unique in that the natural resuspension rate due to convective winds is very low compared to more erodible sites in the Western U.S. He has concluded that the natural desert shrubland, covered by a "desert pavement", or the dry lakes, covered by a crust after a rain, are not subject to wind erosion unless they are physically disturbed.

Schmel has measured resuspension of plutonium at Rocky Flats and of plutonium and cesium at the Hanford Plant.^{16,47} However, the source areas are poorly characterized so that estimates of resuspension rates can not be made. Various values of the power of the increase of concentration with wind speed ranging from unity to 9.3 have been obtained in these experiments so that it is difficult to draw conclusions from these data.

2. Mechanical Resuspension

Mechanical resuspension is that caused by forces other than the wind. Such forces could range from the movement of small animals on the surface, through humans walking, to the movement of heavy equipment or plows across the ground. There are several differences between mechanical resuspension and wind resuspension, chief of which is the fact that the resuspending force is independent of the wind speed (although dilution downwind will increase at higher wind speeds). Instead, the resuspension rate will depend upon the magnitude of the force applied as well, perhaps, as the nature of the force. A second difference is the depth from which resuspension can possibly occur. In the case of wind resuspension, the layer from which particles can contribute to the air stream is limited by the depth to which the saltating particles can cause ejection. For mechanical disturbance the depths over which the forces can

be applied varies with the means of disturbance but could reach depths of one or two feet in plowing. Of course, the probability of resuspension is not the same at all depths but no data are available to indicate possible variations. The extreme example would be the excavation of a hole, such as a basement, where material could be ejected into the air from considerable depths in the ground.

Another difference arises from the fact that most mechanical disturbances are a point source. That is the disturbance occurs over a fairly limited area at any one time. There could be multiple disturbances that could result in an approximate area source or the disturbance could move with time resulting in an average that resembled a line source or an area source. An example of the line source would be traffic moving along a road. The average resuspension rate from the road would be the product of the resuspension per vehicle times the number of vehicles divided by the time over which this number of vehicles passed. An area source would be the average result of a farmer plowing a field and producing a resuspension rate at each point. Here, the average resuspension rate would be the instantaneous resuspension rate that occurs at each point divided by the time required to plow the field. Such relationships allow the derivation of calculational methods for finding the average concentration downwind if the resuspension rate from the disturbance can be defined.

There are also similarities with wind resuspension. One would expect the dust flux to be greater in fields that contain a larger quantity of small aggregates. Many of the factors, such as moisture or vegetative cover that inhibit erosion, would be expected to minimize mechanical disturbance. Thus, in digging in contaminated soil it is common practice to keep the soil damp to minimize resuspension. However, local areas of low saltation (vegetated strips in the field) will not affect the mechanical resuspension rate.

There are a few measurements of mechanical resuspension that can be used to give an order of magnitude estimate of the rate of resuspension under different condition. Sehmel⁴⁹ measured the resuspension caused by an individual walking along a 50 foot length of asphalt 10 feet wide that had been previously seeded with zinc sulfide tracer. He reports that 1×10^{-5} to 7×10^{-4} (at wind speeds of 3 to 18 mph) of the tracer was resuspended per walk through. Assuming a walking speed of 3 mph this would result in resuspension rates of $9 \times 10^{-7} \text{ s}^{-1}$ to $9 \times 10^{-6} \text{ s}^{-1}$. Such values have much uncertainty because of the width of the seeded area but it is noted that they are about two orders of magnitude greater than the wind resuspension rates with wind speeds of 2 to 9 mph.

In a continuation of these same experiments Sehmel⁶⁰ reports the results of driving vehicles in the adjacent lane and through the tracer material. Both a car and a three-quarter ton truck were used. His values reported as fractional resuspension per pass were converted to a resuspension rate through use of the length of the seeded area and the speed of the vehicle. The resuspension rate varied from 10^{-8} s^{-1} to $8 \times 10^{-3} \text{ s}^{-1}$ depending primarily on the speed of the vehicle. Sehmel⁶⁰ had reported that the resuspension was proportional to the square of the speed. The resuspension rate caused by the truck was greater than that caused by the car, presumably because of the greater turbulence from the truck. A rapid weathering of the particles was also noted. In this calculation it was assumed that there was no removal by the winds during the 30 days of the experiment so that the latter rates are the lower limit of the resuspension rates. However, the resuspension rate 30 days after application was two to three orders of magnitude lower than the initial rate. These data indicate that there will be rapid depletion of the source for materials deposited on such pavements if any significant traffic occurs.

Sehmel⁵⁰ also performed a similar experiment with the zinc sulfide tracer placed on a strip of cheat grass. The course was the same size as the asphalt course. A 3/4 ton truck was driven through the area at different speeds. The results indicated a relatively high resuspension rate of $3.7 \times 10^{-5} \text{ s}^{-1}$ on the first pass at 2.2 m/s. For the second pass at 6.7 m/s, the resuspension rate had decreased to $4 \times 10^{-6} \text{ s}^{-1}$ while for the third pass at 13.4 m/s the rate increased to $8 \times 10^{-6} \text{ s}^{-1}$. With the final pass at 17.9 m/s the resuspension rate increased to its highest value of $9.4 \times 10^{-5} \text{ s}^{-1}$. The high resuspension rate at the low speed was caused by removing the most readily suspendible tracer from the cheat grass. By the time the higher speeds were attained it is likely that this more readily suspendible material was removed and the resuspension was from the soil surface.

Milham et al.⁶² described the results of air sampling during the agricultural preparation of two fields having small concentrations of plutonium accumulated 25-30 years earlier as the result of a release from a nearby stack. Samples were taken at several locations during operations in the field. Healy² converted these to approximate resuspension rates by use of the field sizes and meteorological parameters given in the paper. These results are given in Table III.

TABLE III

RESUSPENSION RATES FROM AGRICULTURAL OPERATIONS

	Estimated Resuspension Rates (s^{-1})			
	North Field		South Field	
<u>Milham</u> ⁶²	7.6 m	30.5 m	7.6 m	30.5 m
Bushhogging	9×10^{-8}	2×10^{-7}	1×10^{-6}	8×10^{-8}
Disking	4×10^{-8}	6×10^{-8}	-	-
Subsoiling	7×10^{-7}	3×10^{-8}	3×10^{-8}	3×10^{-9}
Fertilizing	2×10^{-8}	3×10^{-8}	1×10^{-8}	1×10^{-8}
Planting	1×10^{-6}	4×10^{-8}	6×10^{-7}	2×10^{-8}
<u>Meyers</u> ⁶³				
Rototilling	9×10^{-8}			

A somewhat similar experiment was performed by Meyers.⁶³ Here the plutonium was applied to a small field in the form of digested sewage plant sludge containing a small amount of plutonium. The sludge was allowed to dry for a period of four weeks without rain and the area was rototilled. The rototiller was 2 m wide with the dust cover removed and was pulled behind a tractor. The sampling results were, again, converted to an approximate resuspension rate by Healy² with the result given in Table III.

C. Mass Loading

The mass loading concept is an attempt to by-pass the details of the soil characteristics and the resuspension process and to relate directly measured soil concentrations of the contaminant to the air concentration by use of the mass of soil particulates in the air. Thus, the air concentration of the contaminant is given by the product of the concentration of the contaminant in the soil and the concentration of the soil particulates in the air. If the quantity of particulates in the air is known from other data, one need, theoretically, only measure the soils in the region to provide an estimate of the air concentration of the contaminant.

Two parameters, the dust loading in the atmosphere and the appropriate concentration of the contaminant in the soil are needed to provide estimates by

this method. Healy⁹ used an average value of $120 \mu\text{g}/\text{m}^3$ of dust in a generic analysis of limits for plutonium in the soil. This was derived from the Federal Secondary Standard for particulates in the air expressed as a geometric mean of $60 \mu\text{g}/\text{m}^3$ assuming a geometric standard deviation of 2. Anspaugh¹⁰ explored a reasonable mass loading in several ways. The lower bound is quoted as about $10 \mu\text{g}/\text{m}^3$. Examination of the data on the levels in mine atmospheres which have led to a considerable prevalence of pneumoconiosis in the workers indicate that standards on the range of $1\text{--}10 \text{ mg}/\text{m}^3$ have a very small, if any, margin of safety. He also quotes some British data that indicate that dust levels in excess of $1 \text{ mg}/\text{m}^3$ could lead to considerable public health problems. He also used the data on ambient mass loading for 1966 from the National Air Surveillance Network to show that the average for urban stations ranged from $33 \mu\text{g}/\text{m}^3$ to $254 \mu\text{g}/\text{m}^3$ with a mean for all nonurban locations of $38 \mu\text{g}/\text{m}^3$. For the nonurban stations the average ranged from $9 \mu\text{g}/\text{m}^3$ to $79 \mu\text{g}/\text{m}^3$. From these studies he chose an average of $100 \mu\text{g}/\text{m}^3$ as reasonable for predictive purposes.^{12,17} The Environmental Protection Agency⁶⁴ also examined the data from the nonurban stations from the National Air Surveillance Network for the years of 1964-1965. Their map of these data indicate values ranging from $9 \mu\text{g}/\text{m}^3$ in southern Montana to $56 \mu\text{g}/\text{m}^3$ in Western Pennsylvania, $57 \mu\text{g}/\text{m}^3$ on the Southern Oregon Coast, and $59 \mu\text{g}/\text{m}^3$ on the North Carolina Coast. However, the prevalence of high values in the East would indicate the possible inclusion of industrial particulates in these samples. The EPA used a value of $100 \mu\text{g}/\text{m}^3$ in calculation of their screening level.

Several uncertainties appear in the use of the data from the National Air Surveillance Network. The first was pointed out above in that the particulates that are collected can include a portion of those generated by industrial operations so that the values could be high. The second problem arises from the fact that the samplers are frequently in positions, such as on top of buildings, so that they do not measure the air actually breathed by people. Associated with this question is the potential for people engaged in various activities to generate their own dust. This would result in local concentrations in excess of the ambient value measured by the network. However, the $100 \mu\text{g}/\text{m}^3$ value still appears reasonable from the standpoint that it is an average over a full year and people do not work or play in dusty operations all of the time. For example, if we assume that an individual spends eight hours per day, 5 days per week for 50 weeks per year in a concentration five times the maximum value noted

in the ambient air measurements or $300 \mu\text{g}/\text{m}^3$, the average concentration through the year would be only $115 \mu\text{g}/\text{m}^3$. While some individuals, such as farmers, work longer hours during the week, their exposure to dust is limited to fewer weeks per year and a portion of their time in the field is during periods of high moisture or vegetation in the soil where dusty conditions are limited.

Associated with the question of the concentration of soil particles in the air is the question of the origin of the particles. Once airborne, the smaller particles can travel very long distances. For example, Carlson and Prospero⁶⁵ have reported the movement of dust from the Sahara desert over the Northern equatorial Atlantic Ocean and Clayton et al⁶⁶ have reported evidence of transport across the Pacific Ocean to Hawaii. Since most contaminated areas are relatively small in area, one would expect that only a fraction of the dust in the air would originate from the area. Because of preferential deposition of the larger particles from sources some distance away, this "background dust" would contain a higher percentage of the smaller particles that are more readily deposited in the lung than the dust originating from the local, contaminated area. Thus, Anspaugh and Phelps⁶⁷ report that measurements at the GMX area with Anderson high-volume cascade impactors for about one month indicate the mass distribution of sizes are about $1.6 \mu\text{m}$ MMAD with a σ_g of about 15, while the plutonium and ²⁴¹Am had an AMAD of about $3 \mu\text{m}$ with a σ_g of about 7. It was also noted that the average activity of the soil was about one-third of that found in the soil in close proximity to the sampler. It is noted that even higher activity was upwind.

From this, we conclude that a direct comparison of the size distribution of contaminated particles in the air with those in the soil is probably valid only for very large areas. For the more usual size of contaminated area, the dilution of the total mass in the air, particularly in the smaller particle sizes, could be significant. It is noted, however, that for resuspension by mechanical disturbance this dilution may be of lower importance because of the frequently higher concentrations resulting from such disturbances.

The second question, that of the appropriate concentration of the contaminant in the soil, is more subtle. As was discussed earlier, those soil particles that are carried in suspension are the smaller ones because the larger ones will settle rapidly. Thus, if the concentration of the contaminant in the soil fraction containing the small particles is greatly different from that in the other particle sizes, it would appear that the concentration predicted by

the mass loading approach using the total soil concentration would theoretically be low.

Tamura⁶⁸ has analyzed the particle sizes and their associated plutonium content in samples from several existing plutonium contaminated areas and have shown that fractionation of the plutonium content by particle size does exist. Analyses were done using water as the suspending agent and the effect of this, as compared to the carbon tetrachloride used by Chepil or the liquid freon used by Gillette, on the aggregate size is unknown. However, in two samples from the NTS the aggregates less than 20 μ m had three and five times greater plutonium concentration than the total soil mass. (At the NTS, the bulk of the activity appears to be in the 20-53 μ m size range.) In a bottom sediment from the canal at Mound Laboratory the fraction lower than 20 μ m had 1.8 times higher concentration than the total; at the flood plain in ORNL the fraction lower than 20 μ m had 1.1 times the total soil concentration; and a sample from Rocky Flats had about three times the total soil concentration in the fraction less than 20 μ m. It is of interest that these distributions reflect both the method of contamination as well as the soil type. At the Nevada Test Site the plutonium was mechanically dispersed by explosive material and the particle size distribution reflects the largest amount of the plutonium in the 53-125 μ m size range, although the highest concentration was in the smaller particle sizes. The Mound and ORNL samples reflect the distribution expected by adsorption on the smaller particles in the sample while Rocky Flats is intermediate reflecting, perhaps, some adsorption as well as direct contamination of the larger soil particles by the plutonium bearing oil that was the source of the contamination.

The Environmental Protection Agency⁶⁴ proposed the use of an "enrichment factor" to include these data in resuspension calculations. - This is defined as the summation of the products of g_i , the ratio of the fraction of the total activity contained within the size increment i to the fraction of the total mass in the size, and f , the fraction of the airborne mass within each increment of particle size in the air. For the distribution in soil sizes at Rocky Flats, they calculate an enrichment factor of 1.5. Tamura⁶⁵ has defined a "soil plutonium index" which accounts for the size distribution as well as the lung deposition. This is given as

where SI is the soil plutonium index, SA is the soil activity factor, LD is the lung deposition factor, and RA is the resuspendible activity factor. The soil activity factor is the fraction of the activity in a given mass fraction divided by the mass fraction for particles less than 100 μ m. (Tamura used 125 μ m in evaluating this factor because this was the sieve size used in his analysis.) Values of this factor range from 3.14 for the ORNL sample to 7.27 for the Rocky Flats sample. The lung deposition factor is the deposition in the pulmonary region as defined by the ICRP Task Group on Lung Dynamics.⁶⁹ The final factor, the resuspendible activity factor, is the fraction of the total soil plutonium index activity in the resuspendible fraction. Indices derived from available data give 0.52 for Area 13 at the Nevada Test Site, 1.26 for the Rocky Flats area, 1.18 for Mound Laboratory and 0.69 for ORNL.

Another approach to use of the smaller particles is that of Johnson et al.⁷⁰ His sampling technique was to brush the surface dust into a container. The 5 μ m or smaller particle sizes were then separated from the sample after aggregates had been broken up and plutonium analyses were performed on this fraction. He found that the concentration in these small particles was 4 to about 300 times larger than in similar samples taken to a depth of 1/8 inch. His conclusion was that these results provided a better indication of the hazard than the conventional sample although he did not explore mechanisms of breaking down the aggregates found in the soil, which severely limit the quantities of particles of this size found in natural soils, nor did he examine pathways of this material to man.

It may be noted that these relations have never been actually tested to show their validity. The work on soil erosion indicates the many additional factors that will influence wind erosion and resuspension. These include the soil texture, the moisture content of the soil, the presence or absence of vegetation in vegetative residues, and the characteristic surface roughness. In the case of the Nevada Test Site, the desert pavement undoubtedly has more influence on either wind or mechanical resuspension than the other factors. We would believe that wind erosion, in particular, is more complex than these relations would indicate. However, it is possible that such concepts may be more applicable to mechanical disturbance.

became adjusted to the absence of the ice. Although evidence is scant, the vegetation of the Sangamon Stage probably was much like that of today.

By 70 000 B.P., temperatures had again cooled, and another ice age, known as the Wisconsin Stage, began, which lasted until 10 000 B.P. This glacial stage was much like the Illinoian except that in the Middle West, the ice did not extend as far south and terminated just south of what is now Lake Michigan. We know many more details about the fluctuations and history of retreat of the ice during the Wisconsin Stage, but the same features accompanied the ice advance and retreat that must have been present during the Illinoian Stage, including a low sea level, drier climate in the east and resultant dunes and loess deposits, and freshwater lakes in the Great Basin. The ice began to retreat about 18 000 B.P., and by 11 000 B.P., the continental ice had retreated to Lake Superior and the St. Lawrence Valley. During the retreat, the Great Lakes formed in deep valleys and behind dams of glacial debris left by the ice. The time of maximum warmth was probably about 5 000 B.P., by which time Canada must have been free of ice. We believe that increased aridity in the Southwest caused accelerated dune activity at this time. The climate continues to fluctuate, and it is apparently somewhat warmer today than it was in the 18th century.

Constant or Rapidly Recurring Processes of Change

Weathering and erosion go on almost constantly wherever deposition is not taking place. Within a time span of 250 000 years, these processes may make substantial changes in the landscape.

Weathering involves the breakup of rock into individual mineral grains as well as the formation of soil in the upper five feet of the weathered zone. In the humid southeastern United States, certain kinds of rock may be overlain by more than 100 feet of unconsolidated material formed by weathering. As erosion transports the weathered material into the streams, new weathered material and soil form. The average lowering of the land by these processes is believed to be about one foot in 10 000 years. In 250 000 years, this lowering amounts to some 25 feet. Lowering is somewhat more rapid in the semiarid climates of the West. In detail, the rate of lowering differs in different regions and locally in different parts of the landscape. A nearly level plateau surface, for example, is not lowered as rapidly as its bordering slopes. The north-facing

A direct test of the mass loading technique has been made by Anspaugh et al.^{12,17} The measured concentrations of a number of nuclides in the air were compared to a concentration calculated from the quantity of the nuclide in the soil assuming a mass loading of $100 \mu\text{g}/\text{m}^3$. These results are given in Table IV.

The agreement between calculated and predicted is good. Of course the sources for the natural isotopes are large in area. However, the values at Nevada show reasonable agreement between calculation and prediction. It is believed that the soil concentration at the point of sampling was used for the predicted values. If this was the case, the discrepancy between the two results at ground zero is explainable on the basis that the concentration in the soil is highest at this point and the measured dust arose from surrounding areas of lower concentration.

TABLE IV

A COMPARISON OF THE PREDICTED CONCENTRATION IN AIR USING
A MASS LOADING OF $100 \mu\text{g}/\text{m}^3$ TO MEASURED CONCENTRATIONS
(AFTER ANSPAUGH ET AL.^{12,17})

Location	Nuclide	Air Concentration	
		Predicted	Measured
<u>GMX Site, USARC</u>			
<u>Nevada Test Site</u>			
NE, Anspaugh et al. ⁶⁷	²³⁹ Pu	7.2×10^{-3} pCi/m ³	6.6×10^{-3} pCi/m ³
GZ, Anspaugh et al. ⁶⁷	²³⁹ Pu	0.12 pCi/m ³	0.023 pCi/m ³
<u>Lawrence Livermore Laboratory</u>			
Gudiksen et al. ⁷¹	²³⁸ U	150 pg/m ³	52 pg/m ³
Gudiksen et al. ⁷²	²³⁸ U	150 pg/m ³	100 pg/m ³
Silver et al. ⁷³	²³⁸ U	150 pg/m ³	86 pg/m ³
Silver et al. ⁷³	⁴⁰ K	10^{-3} pCi/m ³	9.8×10^{-4} pCi/m ³
<u>Argonne National Laboratory</u>			
Sedlet et al. ⁷⁴	²³² Th	320 pg/m ³	240 pg/m ³
Sedlet et al. ⁷⁴	natU	215 pg/m ³	170 pg/m ³
<u>Sutton, England</u>			
Hamilton ⁷⁵	natU	110 pg/m ³	62 pg/m ³

III. DISCUSSION

This review covered concepts and numerical values related to the resuspension problem and did not include the important conceptual and modeling studies that have been carried out by several individuals including Amato,⁷⁶ Trevino,⁷² Horst⁷⁸ or Slinn.¹ This was done deliberately in order to focus on the non-mathematical aspects of the problem and to attempt to bring the factors of importance into focus.

It is apparent that a gratifying amount of progress has been made on determination of resuspension in the past few years. The studies of Anspaugh et al.^{12,17} at the Nevada Test Site have shown the feasibility of measurement of the resuspension rate in a contaminated area and his application of the mass loading concept has added greatly to the understanding of this model. The work of Gillette^{32,39} and Shinn⁴² on the dust flux have given new insights into methodology and the phenomena concerned. The studies of Sehmel^{47,48}, with the tracer particles, have given values that are extremely useful for application. Slinn¹ has provided parameterization concepts that aid in understanding.

This is not to say that additional work is not needed. Further studies of both the dust flux and resuspension rate at contaminated areas in various regions and types of soil are definitely needed along with models, such as that of Gillette and Shinn⁴², that provide relationships between the resuspension and readily measured parameters that can be used to estimate resuspension rates. Concurrent studies of the dust flux and resuspension of a contaminant are badly needed, particularly in undisturbed areas apart from agricultural soils. The dust flux model requires assumptions as to the connection between resuspension of a contaminant and the dust flux. The only check point now available is the measurements at the GMX area of the dust flux by Shinn et al.⁴² and the plutonium resuspension rate by Anspaugh et al.¹⁷ In particular, additional data are needed on resuspension by mechanical disturbance. Few appropriate experiments are available and it is frequently difficult to interpret them in a manner that provides useful results. A particular area of concern for which very few data are available is the possibility of contamination while playing and working in an area with subsequent transfer to a place where inhalation is more probable. Extreme examples of this would be pulling a contaminated garment over one's head or contaminating pillows or other bed clothing. While one could feel that this could not be a major source of exposure, we cannot tell until the experiments are done.

A primary purpose of this review was to choose resuspension parameters to be used in the calculation of dose to individuals in a contaminated area. The study has reinforced our previous prejudice that the resuspension factor is not the method for use. This is because of the failure of this method to account for many of the variables and because the conditions of the measurement are seldom described in sufficient detail to allow intelligent extrapolation to areas different from those in which the measurements were made. There is some usefulness to this technique, however, in describing the exposure of the individual causing the disturbance.

The resuspension rate has been our favorite method because of the capability of integrating over a contaminated area using accepted dispersion and deposition parameters to provide concentration isopleths around the area. For a specific situation where the soil and meteorological parameters can be defined, this is still the preferred method and the state of the art is rapidly approaching sufficient detail that this can be done.

However, for a generic study, the mass loading approach seems to be best. The work of Tamura,⁶⁸ the EPA⁶⁴ and Johnson⁷⁰ all indicate that even this approach requires revision for the distribution of contamination in the soil. However, as has been pointed out there are factors that tend to compensate for this, such as the size of the area, and the magnitude of the correction factor proposed by the EPA⁶⁴ and Tamura⁶⁸ is only on the order of 1.5 to 2, a value that tends to get lost in the noise of the other uncertainties. In addition, the success shown by Anspaugh in predicting the concentrations of several nuclides in widely different climates and soil types is encouraging.

Anspaugh¹⁰ used a mass loading of $100 \mu\text{g}/\text{m}^3$ in his comparison. The measured values were primarily ambient air and included no component for mechanical disturbance. In view of the agreement found in his study, we would propose the use of $200 \mu\text{g}/\text{m}^3$ for generic studies to make allowance for these other types of exposure. It is noted that this seems to be unrealistically high when compared with air sampling results. This may well be due to the factors proposed by Tamura with the actual mass loading of little importance as compared to the correlation found by Anspaugh using an arbitrary value of $100 \mu\text{g}/\text{m}^3$.

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APPENDIX F

ENVIRONMENTAL CHANGES IN THE UNITED STATES IN THE LAST QUARTER MILLION YEARS

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Environmental changes of a dramatic nature have taken place repeatedly in the last 250 000 years in the United States. These changes can be grouped in three classes. The first and most dramatic has to do with worldwide changes in climate. Another class consists of processes that go on continuously or at repeated short intervals, such as stream erosion and deposition, and weathering and soil formation. A third category relates to processes operating within the crust, such as volcanic and tectonic activity. The history and nature of the changes are best known for the last 60 000 years. The details become less clear as one looks farther back in time. However, many changes have been repeated, and details of the earlier ones can be inferred.

Climatic Changes and Their Effects

Within the last quarter million years, the earth has experienced two major cold, or glacial, periods separated by a warm period and followed by the present warm period. It is generally believed that 250 000 years B.P. (before present) the earth was near the end of a warm cycle. The warm cycle was followed by an ice age (known in the United States as the Illinoian Stage) that lasted until about 150 000 B.P. Ice accumulated in the northern part of the North American continent until it spread south as far as the course of the present Missouri and Ohio Rivers, covering all of New England, the Great Lakes, and almost all of Illinois and the Northern Plains. Ice caps covered all but the high peaks in the Rocky Mountains, Cascade, and Sierra Nevada Mountains. The ice scoured the rocks beneath it, deepening and widening valleys or forming depressions. The material excavated was deposited as moraines and outwash or was left behind as irregular deposits of till and outwash when the ice melted. Some of the depressions left by the ice retreat became filled with sediments and are now nearly level plains underlain by silt and clay. Examples are the fertile plain of the Red River Valley of Minnesota and North Dakota, and the former bed of Glacial Lake Agassiz, which was formed during a later ice age. Marine deposits

were laid down when the sea entered areas that had been submerged by the weight of the ice, as in the Champlain and St. Lawrence Valleys.

Many changes took place outside the ice-covered areas. Great rivers drained the continental ice sheet, including the Missouri, Mississippi, and Ohio. At that time, the Mississippi was not a meandering stream as it is today; it flowed across a broad flood plain in a series of anastomosing channels. In winter this plain was dry, and the wind lifted clouds of silt and fine sand, depositing this material as loess on the hilly landscape east of the river. Because a part of the earth's sea water was transferred to the land in the form of ice, sea level was lower than now by about 300-400 feet. Along the Atlantic Coast, the shoreline in places may have been more than 100 miles out from the present shore, exposing a sandy plain which became covered with vegetation.

At the peak of the cold period, the climate of the eastern United States is believed to have been rather dry. The vegetation of the preceding period migrated southward, and the hardwoods were replaced by northern species such as pine and spruce, probably forming savannah with large areas of herbaceous plants. The northwesterly winds that dominated the weather blew sand and silt across the area, especially on the Coastal Plain, forming scattered dune and loess areas. The crest of the Blue Ridge Mountains and the higher peaks to the west became treeless tundra and probably had fields of perennial snow. In the western United States, in the Great Basin and in the Columbia Plateau to the north, although the climate was dry, the low temperatures slowed evaporation, and closed basins became lakes, many of them interconnected by fresh-water streams. Great Salt Lake, for example, was much larger than now and drained to the sea through the Snake River Plain.

About 150 000 years B.P., temperatures began to warm, and the climate eventually became much like that of today, but perhaps a bit warmer. The warm interval is called the Sangamon Stage. The ice retreated northward, clearing all of North America except perhaps for small glaciers in the western mountains. The rivers draining the ice east of the Rockies became narrower and meandered across only a part of their flood plains, as they do today. Sea level rose as the ice melted, eventually to a level as much as 15 to 20 feet higher than today, flooding some areas that are now dry. As the weight of the ice had depressed the land under it, some of the northern part of the continent was flooded by the sea to a greater depth, until eventually the level of the land

slopes of a valley wear downward more rapidly and are steeper than the south-facing slopes, because they are wetter.

The effects of weathering and erosion through time are visible in formerly glaciated regions. In Illinois, the glacial deposits of the Illinoian Stage extend farther south than do those of the Wisconsin Stage, so that the topography developed on the two may be compared. On the younger deposits (18 000 years old), the topography is hilly, and many undrained depressions hold swamps or lakes. The landscape is probably very little changed since it formed at the ice margin. The deposits laid down by the older ice (150 000 years or more old) have been worn down to a nearly flat or gently rolling plain, completely drained, and broken only by the stream valleys intrenched below the plain.

A dramatic form of erosion is the debris avalanching and flooding that takes place in mountain valleys. This catastrophic form of erosion generally occurs during unusual storms having rainfall as heavy as 5 to 10 inches an hour. Such storms are important processes in the Pacific Coast ranges as well as in the Appalachians. In the latter area, they probably recur at any one place every 500 to 1 000 years.

Deposition of sediment goes on in certain environments more or less continuously. The formation of deltas in lakes or along some ocean shores are examples. The Mississippi Delta is the best example. The river continuously carries large quantities of sediment which is spread out at its mouth as a delta plain. This environment through long periods of time is very unstable, as the distributary channels shift their position or are abandoned and new ones formed. The delta is also influenced by changes in sea level, building up or eroding as the level may change.

Many streams are bordered by flood plains and terraces. The flood plains are flooded on the average every one to two years. Higher, less frequent floods may cover terraces. The river channels on flood plains are subject to changes of position at frequent intervals.

Deposition and erosion are continuing processes along sea and lakeshores. The overall tendency is to straighten the configuration of the shore, eroding in some places, forming notches or cliffs, and depositing beaches in others. The shore environment is generally a high-energy environment subject to constant changes. Over a period of 250 000 years, the entire marine shoreline of the United States has been shifted many times as a result of changes in sea level.

The great lakes and other inland lakes have come and gone in response to climatic change, sedimentation, glacial movements, and other dynamic processes.

Another depositional process is the formation of sand dunes, which takes place along lakeshores or ocean shores wherever there is a large supply of sand. Dunes also form in arid lands generally downwind from stream valleys or other sources of loose sand. The process is continuous where conditions are suitable.

Volcanic and Tectonic Activity

At present, only Alaska and Hawaii have active volcanoes, but the lack of activity in the conterminous United States is abnormal and may be very short lived. For example, the Cascade Range contains volcanoes that have had a long history of violent eruptions—eruptions that have taken place within the last few thousand years as well as earlier. Lassen Peak, in northern California, had a violent eruption in 1914. We believe that the modern cone of Mount St. Helens has formed primarily in the last 2 500 years. Crater Lake was formed by an explosive eruption about 7 000 years ago that spread a sheet of ash more than 6 inches thick over an area 100 miles in diameter. Volcanoes in other western States have been repeatedly active during the last quarter million years. Less than 1 000 years ago, for example, an Indian pueblo near Flagstaff, Arizona, was destroyed by a lava flow. Large and violent eruptions took place in Yellowstone Park as recently as 100 000 years ago.

Tectonic activity, including tilting of the land and faulting, goes on more or less continuously. One example of tilting is found along the Pacific Coast where tilted marine terraces are common. Another example is the rise of land as the ice sheet retreated. Isle Royale in Lake Superior is now 150 feet higher than when it emerged from under the ice. In northern Maine, the land rose more than 500 feet in postglacial time.

Faulting is the differential movement of two adjacent parts of the earth's crust. The movement may be continuous or sporadic and may last millions of years. Active faults produce spectacular changes in the landscape, forming escarpments, trenchlike features, and ponds along their trace. Streams are offset, and other effects are produced. The most famous active fault in the United States is the San Andreas rift in California, along which the motion is mostly horizontal but which has a vertical component in places. The average rate of relative motion amounts to about half an inch per year. This rate is sufficient to have caused the land west of the fault to have moved northward

more than a mile in the last quarter million years. The trace extends out to sea near San Francisco, so presumably some formerly dry land is now beneath the sea. Many other faults are active, particularly in the Great Basin and northern Rockies, where they have changed the face of the landscape to varying degrees, particularly along the foot of basin ranges.

APPENDIX G

TRANSPORT OF TRANSURANICS BY SEDIMENTS IN SURFACE WATERS

J. C. Rodgers

A. Introduction and General Criteria

In the early years of the development of nuclear energy approximately 95 percent of the short-lived, low-level, liquid radioactive wastes were discharged into surface streams.¹ As a result, such river systems as the Columbia and the Tennessee became contaminated, and numerous radiological surveys were made to assess the impact of this disposal practice. Data from such studies provide most of the basis for current efforts to assess the impact of river systems as a vector to man from releases of radioactivity from shallow land burial sites by erosion processes.

Some general criteria for identifying stream conditions conducive to transport and concentration of radionuclides in sediments can be identified, but the development of quantitative factors suitable for limits assessment is not simple and clearcut.¹

In general terms, with respect to the sediment itself, the total available surface area of the sediments is an index of the potential for transport. Concentration of various particle size groups is a suitable indicator of surface area. The greater sorption capacity of the extremely fine sediments is somewhat offset² however, by the tendency of colloidal materials to remain in suspension except when flocculation occurs. The chemical and mineralogical nature of the sediments, particularly fines, are also factors to be considered. The rate and reversibility of sorption reactions could be important.

With respect to stream conditions, the nature of the concentration and load variation of sediments with time are important since they indicate the capacity of the stream to transport a given input of contaminant. For example, a stream with relatively uniform water discharge and high concentration of fine sediment would have a greater capacity to maintain the waste in suspension. But a stream with a large range of water discharge and sediment load, receiving the same contaminant input, would be expected to transport the waste in slugs: buildup of concentration in the bed-load is likely during low flow; then release and possible deposit on high banks or flood plains could occur during high flow. Channel geometry can be a significant factor in reconcentration. A meandering

channel is characterized by building of point bars on the inside bends. The pool-and-riffle type of stream accumulates contaminants in pool reaches during periods of low flow. Man-made control features, especially reservoirs, can have very significant effects such as trapping and then releasing contaminants bound to very fine sediments.

Evidently, then, the range of conditions encountered with regard to (1) the quantity of sediment and water discharge, (2) the time variation of sediment and water discharge, (3) the mineralogic composition of sediment, and (4) channel geometry and the nature and extent of man-made channel controls and reservoirs makes each radioactive waste disposal site and associated watershed unique with respect to the effects of sediment transport as a potential exposure vector. Clearly it is not possible to assess adequately the hazard potential of a given stream on the basis of any one of these parameters.

B. Large Scale Sediment Transport

In the following section, a simple analytic model of large-scale, stream-sediment transport of contaminants will be described as an aid in exploring the significance of the limited data which are currently available from two major watersheds. As would be expected, the large scale effect of the introduction of contaminated sediments in stream sediments is dilution and dispersion. But, as has been suggested earlier, on a smaller scale there is the possibility of reconcentration and buildup in some sediment fractions. This effect, which is much more difficult to model in a quantitative manner, will be examined in the next section.

Stream sediment transport is an exceedingly complex phenomenon, depending on such parameters as velocity, temperature and chemistry of the water, the width, depth, and other geometrical factors of the stream bed, and on the grain size distribution of the sediment particles being transported. Some of these parameters vary widely from stream to stream, and of course, even between various reaches of the same stream.

It should be kept in mind, in the following analysis, that it is the transport of particles of sediment (sand, silt, or clay) that is being described, not bulk water flow. Each particle executes a complex series of motions in the process of moving downstream, spending part of the time in the water column being carried along in the water currents, and part of the time at rest in the sediment bed. At a given moment, at a certain point on a stream.

part of the total sediment load is in suspension (the suspended load) and part is at rest (the bed load). For simplicity, the entire sediment load of the stream can be treated as though it were a moving "entity," like a dust cloud, having its own characteristic velocity and dispersion.

From this perspective, theoretical sediment transport models have been developed following either a deterministic approach using a classic dispersion equation³, or a statistical approach assuming that sediment particles execute a sequence of steps with a characteristic step length, and 'rests' with a characteristic rest period.² It has been shown that both approaches yield comparable predictions for sufficiently large transport distances (i.e., distances relative to the characteristic step length).⁴

Our interest at this stage is the development of a generalized formulation based on available data and applicable to large distances and long times, so that a deterministic model is the appropriate choice.

Consider the case of continuous injection of a contaminant into a stream bed sediment load. De Vries⁴ has represented the transport and dispersion process for bed load particles by the diffusion equation

$$\frac{\partial C}{\partial t} + U \frac{\partial C}{\partial x} = D_L \frac{\partial^2 C}{\partial x^2} \quad , \quad (1)$$

where C is the concentration of contaminant, U is the average downstream propagation velocity of sediment particles, and D_L is the longitudinal (downstream) diffusion coefficient. If to this we add a general concentration dependent loss term, λ , which will be taken to represent the combined effects of radioactive decay and whatever other physical losses that occur, such as irreversible deposition in stream bed or banks, the equation becomes,

$$\frac{\partial C}{\partial t} + U \frac{\partial C}{\partial x} + \lambda C = D_L \frac{\partial^2 C}{\partial x^2} \quad . \quad (2)$$

The integration of (2) over time T for C constant at x=0 represents the steady-state condition resulting from continuous injection of contaminants at the origin of the contaminant dispersal, and has the solution:⁵

$$C(x,t) = C_0 U / (4\lambda D_L)^{1/2} \exp \left\{ \frac{Ux}{2D_L} \right\} \int_0^{t=T} \frac{1}{t} \exp \left\{ -\frac{a}{t} - bt \right\} dt, \quad (3)$$

where $a = x^2/4 D_L$ and $b = U^2/4 D_L + \lambda$, and C_0 is the initial concentration. When $T \rightarrow \infty$ (3) becomes

$$C(x,\infty) = C_0 / (1 + 4\lambda D_L / U^2)^{1/2} \exp \frac{Ux}{2D_L} (1 - \sqrt{1 + 4\lambda D_L / U^2}). \quad (4)$$

The values of the transport velocity and downstream dispersion coefficient must, of course, be appropriate to the movement of sediments, and thus are not the same as the water and dissolved constituent values. Tracer studies supported by statistical analysis, and in-situ studies of contaminated sediments, both yield empirical estimates of U on the order of 10^{-4} to 10^{-3} m/sec, and D_L about $0.28 \text{ m}^2/\text{sec}$. (Table G-I).

Since the values of D_L and U are small and the value of λ is expected to be small for most TRU's, the expression for $C(x, \infty)$ can be simplified considerably by using the approximation that $\sqrt{1+x} = 1 + x/2$, for small x; and approximating $C_0 / (1 + 4\lambda D_L / U^2)^{1/2} = C_0$. Then

$$C(x,\infty) = C_0 \exp (-x\lambda/U). \quad (5)$$

TABLE G-1
SUMMARY OF MEASUREMENTS OF SOME STREAM
SEDIMENT PARAMETERS

<u>Channel</u>	<u>Bed Material</u>	<u>Mean U_c</u> (m/sec)	<u>$\frac{D_b}{U_c}$</u> (m ² /sec)	<u>References</u>
N. Loup River	0.29 mm sand tracer	2.5×10^{-4}	0.28	4
Danube River	0.24 mm sand tracer	8.0×10^{-5}	-	7
Velika Morava	0.36 and 0.60 mm sand tracer	8.3×10^{-5} 2.7×10^{-4}	-	7
Columbia River	In situ ⁶⁰ Co and ⁶⁵ Zn sediment bound	3.3×10^{-4} 9.7×10^{-4}	-	6

Thus, the principal sediment transport factors can be reduced, for the case of continuous release and transport over long distances and the large time frames being considered here, to sediment transport velocity U , and the combined loss rate λ . Since the magnitude of the sediment transport velocity has been given rough (order of magnitude) bounds by radioactive tracer study data, the combined loss rate parameter can be quantified using field data on the large scale spatial distribution of radionuclides bound to stream sediments under conditions of more or less continuous release. It is recognized that this procedure leaves much to be desired as far as an understanding of actual physical processes involved in the loss of contaminants from a flowing bed of sediments. The field data represent the combined effects of a large number of unknown process, which are likely very different from stream to stream. What these data do provide is a somewhat limited perspective on a range of values of λ .

As was previously noted, continuous releases over a long period of time occurred in the Columbia and Tennessee River systems for which some data pertinent to the sediment transport model exist.

Data of Nelson and others⁶ for long-lived activation and fission products released from the Hanford complex on the Columbia River suggest that a fairly

narrow range of loss rates applies to those radionuclides that adsorb fairly strongly on sediments. By assuming a constant average sediment velocity of 2.5×10^{-4} m/sec, values of λ on the order of $3-5 \times 10^{-9} \text{ sec}^{-1}$ yield a good fit by the model to data over a 322 km (200 mile) reach of river downstream from the McNary Dam (the first major sediment basin). These losses include the sedimentation effects of two dams as well as other processes in this cold, fast moving, gravel-bed river.

Data from studies of the fate of radionuclides released from the Oak Ridge operations to the Tennessee River Watershed are representative of quite a different system, one characterized by warmer water, greater sediment load (with different size distribution), more man-made stream controls, etc. The strongly sediment-bound radionuclide for which there is the best quality and quantity of data is ^{137}Cs . In fresh water environments, there appears to be a good correlation between ^{137}Cs behavior and plutonium behavior as far as sediment transport is concerned.⁸

The steady-state condition, after years of nearly continuous input and repeated episodes of scouring followed by deposition behind one of the eight dams along the river, appears to be a ^{137}Cs sediment concentration vs downstream distance pattern having an overall exponential form similar to the Columbia River over the first 161 km (100 mi) to 321 km (200 mi). But the remainder is characterized by a lower rate of reduction, probably due to the extensive system of dams in the lower reaches. Figure 1 illustrates the comparison of model projections and field data for these two river systems.

On the basis of these implied loss and transport rates it is evident that a long-lived TRU contaminant on stream sediments may build up to an equilibrium distribution, which has a fairly shallow down-stream concentration gradient, and may become a persistent source for a great many years after the input ceases. Evidence for such persistence of sediment-bound plutonium can be found in studies of ^{238}Pu in the Miami River watershed.⁹⁻¹³ As the result of a number of inputs from the Mound Laboratory facility, ^{238}Pu exists as a sediment-bound contaminant of the Great Miami River, not all of which can be fully identified and quantified. Two major inputs have been (a) the accidental release of ^{238}Pu due to pipeline rupture in 1969, which resulted in the input of contaminated sediments carrying many curies of ^{238}Pu to the nearby Miami-Erie Canal, and from there to the Miami River¹³, and (b) the weekly pulse release of low-level waste water from a holding tank containing small quantities (10-200 μCi) of

Fig. 1. Large-Scale Reduction in Sediment Concentration (Columbia and Tennessee Rivers).

plutonium.¹² Factors that have contributed to the present distribution and its observable effects will be discussed further below. The point to be made here in the context of persistent, large scale distributions is that nearly 10 years after the major contaminating event, the weekly releases of plutonium result in observable pulses which, at points up to 40 km down stream, account for only 10% to 20% of the weekly flux of ^{238}Pu passing downstream (35 $\mu\text{Ci/week}$ attributed to "background").¹²

C. Local Effects and Concentration Buildup

Although the large scale model just described would be useful for approximating the widest possible impact of sediment transport of TRU

radionuclides from a release point to the mouth of a river, local impacts, including the "worst-case" exposure condition expected near the point of discharge of contaminant to the stream, are not appropriately modeled at all.

As was observed above in the discussion of general aspects of sediment transport, the combined effects of the relatively greater sorptive capacity of the fine fraction of sediments (clays and silts) over the coarse fraction (sands), and different sized sediment particles (i.e., the duration of suspension is inversely related to particle size) combine to create the potential for reconcentration of radionuclides in certain reaches of streams. Here an estimate of the magnitude of this reconcentration process will be made.

Both ^{137}Cs and $^{238,239}\text{Pu}$ have been observed to attach preferentially to the smaller size classes in soils and sediments.^{14,15} For example, in Ohio soils, the most significant fractions in terms of total plutonium content are the 4 - 2 μm and <2 μm size class, which generally account for 25-35% of soil weight, but contribute 60-75% of soil plutonium.¹⁴ Due to the scarcity of field data on sediment transport of TRU's ^{137}Cs data will be utilized in support of what plutonium data there is.

A good example of ^{137}Cs data illustrating the reconcentration or buildup process in a watershed of limited extent and complexity is found in a study of a reservoir in Mississippi.¹⁵ Tables G-II and G-III summarize some of the data from the study. Soil samples were collected in 2.5 or 5 cm increments to a depth of 10 cm; sediment samples were collected in 5 or 10 cm increments to a depth of 60 cm.

TABLE G-II
CONCENTRATION OF ^{137}Cs UNDER LAND
USES OF POWERLINE WATERSHED

Land use	% of watershed	^{137}Cs Conc. ($\mu\text{Ci}/\text{m}^2$)
Oak-hickory	10	159.0
Pine	79	153.2
Grass	7	147.2
Eroded	4	8.9
Average ^{137}Cs		147.6

TABLE G-III
CONCENTRATION OF ^{137}Cs IN SEDIMENTS

<u>Depositional Area</u>	<u>^{137}Cs Conc. ($\mu\text{Ci}/\text{m}^2$)</u>
Stream Channel	2.3
& Overbank Deposits	2.3
Flood Pool	207.3
Conservation Pool	412.2

Evidently, the coarse-textured stream channel and overbank sand deposits had the lowest ^{137}Cs concentration in the whole watershed. But the concentration in the medium to fine textured sediments of the conservation pool (the portion of a reservoir at an elevation below the free flow pipe outlet) was 2.8 times the average ^{137}Cs concentration of the watershed when compared on a unit area basis, and two times the concentration in the medium textured sediments of the flood pool region of the reservoir.

Another example of similar processes at work is found in data on sediment concentrations of ^{137}Cs in Tennessee River silt downstream from the ORNL site.¹⁶ Table G-IV gives the approximate ratios of ^{137}Cs concentration in sediments at the dam, and just upstream, for several of the dams downstream from the entry of the Clinch River. Clearly these ratios are in the same range as the reservoir data.

Some insights into the mechanisms at work in the sorting processes revealed by data such as these can be found in a recent finite-element model of sediment transport of ^{137}Cs in the Clinch River.¹⁷ The modeling procedure involves simulating the transport of sediments within the water body. Models of dissolved and particulate interactions are used to process these results. Finally changes of river bed conditions are recorded, including river bottom elevation change, ratios of sand, silt, and clay, and distribution of radionuclide concentration in the river bed. Sand, silt, and clay are modeled independently since sediment movement and adsorption capacity vary significantly with particle size. The model includes effects of (i) convection and dispersion of sediments, (ii) fall velocity and cohesiveness, (iii) deposition on river bed, and (iv) tributaries. Table G-V shows values of the critical parameters of the model taken from Clinch River studies conducted in the mid-1960's.¹⁷ The

TABLE G-IV

RATIO ^{137}Cs CONCENTRATION IN STORAGE POOLS TO
CONCENTRATION IN TENNESSEE RIVER SEDIMENTS
UPSTREAM, 1961-1963^a

Location on Tennessee River	Ratio		
	1961	1962	1963
Entry of Clinch River	0.95	1.05	1.11
Watts Bar Dam	1.15	0.92	0.92
Chickamauga Dam	2.5	2.7	2.0
Hales Bar Dam	1.78	1.92	2.0
Guntersville Dam	2.5	2.67	4.0

^aBased on gamma count rate at surface of silt.

release rate is assumed to be continuous. Case 1 is the base case, case 3 is for increased deposition rate, and case 4 assumes increased river flow. The concentrations of released ^{137}Cs at White Oak Creek are assumed to be: 2.5×10^5 pCi/m³ in dissolved phase, 7.42×10^7 pCi/kg attached to silt, 7.42×10^8 pCi/kg attached to clay, and none attached to sand. This implies 98% of the release is via sediment transport, as expected.

Results of the simulation show that, on the one hand, the concentration of ^{137}Cs adsorbed on suspended silt remains steady after an initial rapid buildup from near White Oak Creek to 30 km downstream where the quantity of suspended silt drops dramatically due to deposition in the quieter, deep water. On the other hand, the ^{137}Cs adsorbed on suspended clays rapidly builds up to an order of magnitude greater concentration over the same reach, but then drops only slightly in the quieter water. The suspended clay does not begin to deposit appreciably until the flow almost reaches the mouth of the river (40 km) where the flow velocity is extremely low. It is the combination of the greater sorptive capacity of the silts and this natural sorting process that begins to account for buildup in areas of high sedimentation.

Data from the application of the model to the Clinch River conditions allow a tentative quantification of buildup effects. These values are approximate since only three particle sizes (and their corresponding properties) are modeled. In Table G-VI are tabulated results for reaches near the beginning, middle, and mouth of the river for the three cases of interest: baseline,

TABLE G-V

CONDITIONS FOR MODELING CONTINUOUS RELEASE
OF ^{137}Cs TO THE CLINCH RIVER

Parameters	Case 1 (Baseline)	Case 3 (4X deposition)	Case 4 (2X flow rate)
Discharge of Clinch R. (m^3/sec)	117.8	117.8	226.7
of White Oak Creek	1.3	1.3	1.3
of Poplar Oak Creek	7.4	7.4	7.4
of Emory River	45.0	45.0	45.0
River Sediment Sizes (mm)			
sand	0.125	0.125	0.125
silt	0.030	0.030	0.030
clay	0.002	0.002	0.002
Critical Shear Stress Erosion (kg/m^2)			
silt	0.070	0.070	0.070
clay	0.073	0.073	0.073
Erodibility Coeff. (kg/m^3)			
silt	0.15	0.15	0.15
clay	0.15	0.15	0.15
Critical Shear Stress for Deposition (kg/m^2)			
silt	0.005	0.020	0.005
clay	0.0025	0.010	0.0025
Radionuclide Adsorption Rate ($\text{m}^3/\text{kg-day}$)			
sand	200	200	200
silt	500	500	500
clay	1 500	1 500	1 500
Vertical Diffusion Coeff. (m^2/sec)	0.003	0.003	0.003
Size Distribution at White Oak Creek	sand 35%	silt 53%	clay 12%

TABLE G-VI
CLINCH RIVER MODEL RESULTS

Downstream Distance (km)	Suspended Sediment (mg/l)		Suspended ¹³⁷ Cs Activity (pCi/gm)		Total Activity/ Total suspended Sediment (pCi/gm)
	Silt	Clay	Silt	Clay	
Case 1. Baseline					
10	7.1	0.5	1x10 ³	8.6x10 ³	11.4/7.6x10 ⁻³ = 1 500
20	6.4	0.5	1x10 ³	8.6x10 ³	10.7/6.9x10 ⁻³ = 1 550
30	-0-	0.5	1x10 ³	9.5x10 ³	4.8/5x10 ⁻⁴ = 9 500
Case 3. 4 x deposition					
10	7.1	0.5	9x10 ²	8.5x10 ³	10.6/7.6x10 ⁻³ = 1 400
20	6.4	0.5	9x10 ²	8.0x10 ³	9.76/6.9x10 ⁻³ = 1 414
30	-0-	0.5	-0-	-0-	-0-
Case 4. 2 x River flow					
10	15	8	250	600	8.55/2.3x10 ⁻² = 372
20	14	7	250	600	7.7/2.1x10 ⁻² = 367
30	10	5	250	650	5.8/1.5x10 ⁻² = 383

increased deposition (increased critical shear stress for deposition), and increased river flow.

Judging from the total suspended activity in sediments there is a 6-fold buildup in the baseline case, and a 2% and 3% buildup in the other two cases. Recall that these results are derived under steady-state conditions (constant release rate, constant river flow). In fact, one would expect strong seasonal changes in both input rate and river flow, so that actual conditions might be expected to be somewhere between Case 1 and Case 4. Clearly flow rate very strongly influences the buildup effect.

With the same precaution in mind, a particularly useful way to express these results, which allows comparison with other data and permits impact assessment, is to compute the ratio K_s , the total sediment bound concentration of contaminant in areas of high deposition to the total annual release of contaminant to the river system (pCi/gm/Ci/yr). For the present case, the annual release rate of ^{137}Cs at White Oak Creek was 265 Ci/yr. Then from Table 6 results it can be seen that, for the baseline case, K_s ranges from 5.6 to 36; for increased deposition rate, K_s is approximately 5.3; and for the case of higher river flow, K_s is approximately 1.4.

Studies of ^{137}Cs concentrations in Clinch River sediments downstream from White Oak Lake for the period 1954-1961 have shown a strong correlation between the annual release (curies) and the concentration 1.8 km upstream of the mouth (correlation coefficient of 0.8).¹⁸ For the same data, the slope of the regression of concentration in the sediment on annual release, or K_s , is approximately 2 pCi/gm/Ci/yr. This result suggests that, averaged over fairly long times, the values of K_s lies near the low end of the range of values derived from the steady state model. Again, this would be expected since seasonal scour of varying intensity over the years is a normal aspect of river flow cycles, and accordingly deposits of clay-size sediments are resuspended and redistributed.

At this point one might wonder, in spite of the general agreement of the data reviewed thus far, whether the K_s estimates developed here could be generally and usefully valid since river size and sediment load vary so much. Part of the leverage for skepticism concerning the generality of these estimates derives from a perception of the process of erosional release followed by sediment transport as one of simple dispersion and dilution by uncontaminated sediments. On this account, variations in the flow and sediment load of a stream would seriously affect the dilution factor implied in the parameter K_s . Fortunately there are some data with which to test this perception of the process.

First, utilizing TVA data on the sediment loads in the Tennessee River during the period 1950-60, and data on ^{137}Cs concentrations in sediments, an attempt was made to compare the observed decrease in radionuclide concentrations in the downstream with that predicted by dilution by uncontaminated sediments.¹⁸

Some of the findings are tabulated in Table G-VII. Evidently the sediment dilution estimate gets progressively worse as more uncontaminated sediments are

TABLE G-VII
COMPARISON OF OBSERVED REDUCTION IN ¹³⁷Cs CONCENTRATION
IN TENNESSEE RIVER SEDIMENTS WITH PREDICTED
REDUCTION BASED ON SEDIMENT DILUTION¹⁸

<u>Downstream Distance from source (km)</u>	<u>Predicted Reduction Based on Sediment Dilution (% of initial Conc.)</u>	<u>Observed Reduction (% of initial Conc.)</u>
161	50	60
323	4.8	23
484	0.4	15
564	0.12	11

added. This directly contradicts the sediment dilution hypothesis. A second piece of evidence on this process comes from a mass balance study of fallout plutonium in a watershed near Sidney, Ohio.¹⁹ The Great Miami River at the USGS gaging station at Sidney exhibits sediment concentrations varying from near zero to over 1500 mg dry weight/liter. It was apparently expected that during periods of high sediment loading and high flow there would be a reduction in plutonium concentration in sediments. However, as some of the data displayed in Table G-VIII show, the sediment concentration of ^{239,240}Pu remained remarkably constant over a 6-fold increase in stream flow and sediment concentration. Of course this case is not one of simple dilution by uncontaminated water and sediments since the whole watershed is contaminated by fallout. But nevertheless, since the total sediment load is most definitely not uniformly labeled with contaminant due to the greater sorptive capacity of the fine sediment fraction, these data do tend to contradict the dilution hypothesis. Other mechanisms, described above as natural sorting processes, are apparently overriding dilution.

These findings, therefore, tend to support the view that the maximum buildup of sediment contaminant concentration occurs in the first reach of quiet water downstream from the release point as a complex result of natural sorting processes, and cannot simply be predicted from river flow and sediment load data. A range of possible values of K_s obtained from a number of measured

TABLE G-VIII

MIAMI RIVER PARAMETERS AND CORRESPONDING $^{239,240}\text{Pu}$ SEDIMENT
CONCENTRATIONS AT THE SIDNEY, OHIO GAGING STATION.¹⁹

<u>Date</u>	<u>Flow Rate (L/sec)</u>	<u>Sediment Concentration (mg/L)</u>	<u>$^{239,240}\text{Pu}$ in Suspended Sediment (pCi/g)</u>
12/16/74	53 500	64.9	14.7 $\pm$ 1.9
1/7/75	16 900	34.1	8.4 $\pm$ 2.3
1/15/75	19 050	47.8	19.4 $\pm$ 2.0
2/2/75	30 800	50.0	16.7 $\pm$ 2.2
2/12/75	12 700	15.7	11.4 $\pm$ 2.3
3/3/75	27 800	80.2	22.1 $\pm$ 1.5
3/20/75	47 800	83.0	20.5 $\pm$ 1.5
4/15/75	7 130	13.2	19.7 $\pm$ 3.8
5/28/75	4 920	74.5	6.5 $\pm$ 0.7

stream conditions and mathematical modeling of the Clinch River is 1.4 - 36 pCi/gm/Ci/yr.

D. Transport in Estuary and Ocean Sediments

To complete the analysis of the transport of TRU released to sediments from shallow earth burial sites, the effects occurring in estuaries and adjacent ocean waters must also be considered.

Some of the contaminated sediments from waste burial sites transported by large rivers will eventually be discharged into an estuary and finally the oceans. Radionuclide transport in the estuary zone is particularly difficult to fully characterize because of the complex fresh and salt water interactions possible under the varying conditions of river flow, tide, and wind. But certainly estuaries are sites of rapid sedimentation.

Studies of the Hudson River estuary²⁰, and the Savannah River Estuary²¹, have shown that, as in fresh water, attachment and transport with particulate is the major mode of transport in the salt water environment. As might be expected then, sedimentation rate is a critical factor in determining areas of highest concentration. In Table G-IX it can be seen that highest surface

TABLE G-IX
RADIONUCLIDES IN HUDSON ESTUARY SEDIMENTS

Collection Date	Location	Depth Interval (cm)	Activity (pCi/kg)	
			^{137}Cs	$^{239,240}\text{Pu}$
10/25/73	N. Y. Harbor	0-5	1 260 $\pm$ 38	32.7 $\pm$ 2.4
		5-10	1 795 $\pm$ 60	31.7 $\pm$ 3.2
		10-15	1 995 $\pm$ 56	43.3 $\pm$ 3.0
		15-20	960 $\pm$ 44	45.8 $\pm$ 3.2
		20-25	1 030 $\pm$ 40	26.4 $\pm$ 1.2
		25-30	370 $\pm$ 28	38.5 $\pm$ 1.3
		35-40	925 $\pm$ 49	48.5 $\pm$ 1.6
		40-45	1 190 $\pm$ 40	-
6/20/75	Subtidal Bank Main Naviga- tional Chan- nel	0-1	580 $\pm$ 25	12.9 $\pm$ 0.9
		1-3	525 $\pm$ 45	12.4 $\pm$ 0.8
		3-4	410 $\pm$ 20	11.1 $\pm$ 0.8
		4-5	335 $\pm$ 25	-
		5-10	-1 $\pm$ 15	-
8/29/73	Cove; shallow Embayment	0-5	2 475 $\pm$ 63	69.2 $\pm$ 6.4
		5-10	1 825 $\pm$ 68	52.1 $\pm$ 4.3
		10-15	210 $\pm$ 17	5.7 $\pm$ 0.4
		15-20	26 $\pm$ 17	-
		20-25	35 $\pm$ 23	-
		50-55	9 $\pm$ 14	-

sediment radionuclide concentrations were observed in coves, lowest in navigational channels, and intermediate levels in harbors.

Here, as in the case of reservoirs and rivers, there is evidence of sorting processes. If, for example, the navigational-channel sediment radionuclide concentration is taken as a typical bed load value, enhanced concentrations in certain cove or harbor areas of 2 to 5 times this value can be expected on the basis of these data. This range of values is quite consistent with the reservoir case. As the investigators note²⁰, there appears to be a close covariance between $^{239,240}\text{Pu}$ concentrations and ^{137}Cs concentrations in these sediments, extending the previous use of ^{137}Cs as a model of the physical transport behavior of sediment bound TRU such as plutonium.

The mixing of seawater and river water takes place throughout the estuary, and continues into the ocean over the continental shelf. Kautsky found local

highs in fallout radioactivity levels in seabed sediments off the outer mouth of the River Elbe²², and more complete studies on the Columbia River estuary and adjacent continental shelf by Forster have substantially confirmed this local elevation in radionuclide concentration in seabed sediments near the mouth of river (called the Kautsky Effect by Forster).²³

From the ¹³⁷Cs data for the Columbia River, its estuary and offshore sediments, it would appear that an amplified concentration in sediments, on the order of six times the concentration of the main channel of the estuary might be expected to occur in some locations out onto the adjacent continental shelf as far as 30 km beyond the mouth of the river.¹⁷ In the Savannah River estuary, the amplification is about nine times.²¹ The magnitude of the impact on a given estuary and adjacent ocean shelf will depend on the hydrologic parameters characterizing the stream discharging contaminated sediments into the system. For example, plutonium concentrations in the Newport River estuary sediments were found to be about three times higher than those in the Savannah River estuary despite the fact that the Newport River receives only fallout whereas the Savannah River estuary receives both fallout materials and discharges from the DOE Savannah River Plant.²¹

Since the estuary and nearby ocean waters are the ultimate sinks for the contaminated sediments transported through the river system, long-term concentration trends in these sediments are of concern. Not much information has been found regarding long term trends in the buildup of transuranics in bay and ocean sediments. The studies around the nuclear reactor near Bombay Harbor and bay indicate a steady increase over a period of five years with plutonium accumulation higher by a factor of 10-100 compared to the fallout levels observed in surface sediments from other areas near Bombay.²⁴ However, there are insufficient data to relate the observed concentrations to a source term.

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APPENDIX H

TRANSURANIC UPTAKE BY AQUATIC ORGANISMS

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Plutonium and the transuranium elements were, for years, considered to be unimportant in ecological transfers and food chains because of their low solubility and uptake when ingested by mammals. It is true that compared to cesium or strontium, the nuclides of concern with radioisotopes in the environs for years, the involvement of plutonium is small.

However, sufficient information has been developed in the past five to ten years to indicate that the transuranics do take part in aquatic food chains. Interpretation of environmental data is difficult due to differing degrees of surface contamination and/or gut loading for the fish. Thus, true biological incorporation relationships are often masked by these contributions which often indicate a higher degree of assimilation than actually occurs.

In order to assess the potential hazards from transuranic elements released to aquatic environments, some measure of this behavior, particularly as it leads to foods of interest to man, is needed. The following review of available data is intended to provide information upon which such an assessment can be made although it is not intended to be a complete review of the literature on the subject.

Fresh Water Systems

Emery et al.^{1,2,3,4} have described the behavior of plutonium and americium in a pond at the Hanford Plant. The pond is fed by a plutonium processing plant and laundry wastes with a flow of about ten m³ of water each minute. About 95% of the water leaving the pond is by percolation into the soils. The pond is described as an ultra-eutrophic system with most of the plant nutrients supplied by wastes from the laundry. Analyses of sediment from trenches leading to the pond indicated that much of the transuranium elements from historic releases were removed to the sediments of the trenches and thus did not reach the pond. This raises the possibility that leaching into the pond of plutonium and

americium that were deposited years earlier may constitute a supply of possibly soluble radionuclides that could affect the measured values.

The sediment contains >95% of the total plutonium pool in the pond. The potential availability of ^{238}Pu , $^{239-240}\text{Pu}$, and ^{241}Am were estimated by a series of extractions using sodium chloride, oxalate, and EDTA. Concentration ratios (the ratio of the concentration in the organism to the concentration in the water) for biota were corrected by the authors to account for the estimated available fraction from the sediments or water. However, for purposes of this presentation, we have chosen to present concentration ratios (CR) for plutonium and americium related to the total concentrations reported in sediments and water. Table H-I clearly demonstrates the difference in calculated CR values for aquatic biota when different sources of the transuranium element are assumed.

TABLE H-I
CONCENTRATION RATIOS FOR Pu AND Am
EXPRESSED WITH DIFFERENT BASES

Sediment	Base for Concentration Ratio					
	Sediment		Interstitial Water		Water	
	Pu	Am	Pu	Am	Pu	Am
Algae Floc	5	3	219	185	2×10^8	2×10^5
Snail	0.2	1	6	62	5×10^6	8×10^4
Submergent Cattail	0.2	0.6	7	34	5×10^6	5×10^4
Algae	0.1	-	6	-	5×10^6	-
Native Goldfish, w/o Gut	0.05	0.1	2	7	2×10^6	8×10^3
Emergent Cattail	0.02	0.05	0.8	3	5×10^5	3×10^3
Beetle (coleoptera)	0.009	0.01	0.3	0.7	3×10^5	8×10^2
Submergent Bulrush	0.003	-	0.1	-	1×10^5	-
Emergent Bulrush	0.003	0.001	0.1	0.2	1×10^5	3×10^2
Native Goldfish Muscle	0.002	0.005	0.06	0.3	5×10^4	4×10^2

The ratios of both ^{241}Am and ^{238}Pu to $^{239,240}\text{Pu}$ in organisms compared to those ratios in various potential environmental source compartments are presented in Table H-II.

A value of 1.0 would indicate that the acceptor compartment (biota) contained exactly the same nuclide ratio as the proposed source compartment (sediment, interstitial water, overlying water). Therefore, that compartment which exhibits ratios closest to one for both nuclides should be given primary consideration as the prime source of transuranics in this system. From Table H-II it can be seen that interstitial water most closely meets this requirement. The significance of these ratios is not clear because it is impossible to distinguish between possible actual separations of these nuclides and such factors as different inputs of the nuclides.

Marshall et al.⁵ provided an earlier assessment of the role of organisms in removal of plutonium from Great Lakes waters and the potential for plutonium to reach man via the food chain. Concentration ratios are reported in Table H-III for biota of the five Great Lakes, with emphasis on samples from Lake Michigan. High CR's in mixed plankton compared to zooplankton are thought to be, at least partially, due to plutonium in phytoplankton which predominate in mixed plankton samples. It is further suggested that much of the ^{239}Pu in zooplankton samples is due to phytoplankton in their digestive tracts. Food chain relationships between most of the species, shown in Table H-III are mentioned in the text of Marshall et al.'s. paper and are listed in Table H-IV.

TABLE H-II
COMPARISON OF NUCLIDE RATIOS IN POTENTIAL SOURCE
COMPARTMENTS AND BIOTA

	Sediment	Interstitial Water	Overlying Water
$^{238}\text{Pu}/^{239,240}\text{Pu}$	1.7-1.9	0.74-0.84	0.41-0.46
$^{241}\text{Am}/^{239,240}\text{Pu}$	2.3-2.8	1.7 -2.0	0.002-0.003

TABLE H-III
CONCENTRATION RATIOS FOR ²³⁹Pu IN GREAT LAKES BIOTA,
JUNE 1972 TO NOVEMBER 1973 (WET WEIGHT)

Lake	Sample	Concentration Factors over Water		
		Number	Mean ^a	Range
Superior	Mixed plankton	3	4 000 ± 900	2 680 - 5 700
	Zooplankton	1	630 ±	
	Smelt	1	6	
Michigan	Mixed plankton	22	5 700 ± 800	620 - 15 300
	Cladophora sp.	16	3 800 ± 500	1 060 - 6 930
	Zooplankton	9	350 ± 60	122 - 653
	Mysis relicta	7	760 ± 60	587 - 989
	Pontoporeia affinis	2	1 600	1 450 - 1 830
	Slimy sculpin	7	250 ± 60	128 - 560
	Chub	8	37 ± 3	21 - 50
	Alewife	7	25 ± 2	17 - 30
	Smelt	6	20 ± 4	6 - 33
	Perch	2	16	4 - 29
	Whitefish	2	14	5 - 23
	Coho	1	7	
	Chinook	1	4	
	Lake trout	2	1	1 - 2
Huron	Mixed plankton	2	4 460	3 340 - 5 680
	Alewife	2	165	25 - 305
	Smelt	1	13	
	Perch	1	24	
Erie	Zooplankton	3	500 ± 150	316 - 788
	Smelt	1	235	
	Perch	1	10	
Ontario	Mixed plankton	3	2 420 ± 200	2 030 - 2 670
	Alewife	1	176	

^aMean ± standard error.

TABLE H-IV
FOOD CHAIN RELATIONS

<u>Species</u>	<u>Primary Food Sources</u>
Zooplankton (general)	Phytoplankton
Mysis, Pontoporeia (crustaceans)	Periphyton near surficial sediment layer
Sculpin	Pontoporeia
Chub, Alewife, Smelt Whitefish	Zooplankton or mixture of zooplankton & benthic invert.
Perch	Mixture of invert. & fish
Coho salmon, Chinook salmon, lake trout	Smaller fish

The authors concluded that, although the concentration of plutonium in phytoplankton is several thousand times that in the water, the concentration in other organisms decreases by an order of magnitude in each successive link in the food chain leading to man.

Davis et al.⁶ measured the concentration ratios for neptunium-239, an isotope with a 2.3 day half life, in Columbia River organisms. This isotope resulted from the operation of the production reactors at Hanford. The results are given in Table H-V. In interpretation of these values it must be remembered that neptunium-239, with a short half-life, may well give values lower than for the neptunium-237 of interest in waste management. The factor is not known because the biological half-lives of the nuclide in the organisms are unknown.

Wahlgren and Marshall⁷ studied the distribution of residual fallout plutonium in Lake Michigan between the water and the various trophic levels of the food chain. They found the concentration ratio for plutonium in phytoplankton compared to water was about 5 000. A reduction in concentration by a factor of about 10 was observed at each trophic level consisting of zooplankton, planktivorous fish, and piscivorous fish. The top predators had concentrations only slightly greater than the lake water. However, the concentration in benthic fish was considerably higher than in the planktivorous fish. From their figure showing these levels, we would estimate the

TABLE H-V
CONCENTRATION RATIOS FOR ^{239}Pu
IN COLUMBIA RIVER WATER

<u>Species</u>	<u>CR (wet weight)</u>
Green algae	280
Sponge	40
Insect larvae	30

planktivorous fish to have a concentration ratio of about 20 over the water and the piscivorous fish a concentration factor of about two, while benthic fish showed a CR of approximately 300.

The major role of phytoplankton in plutonium kinetics in aquatic systems has been postulated to be one of removal of a significant fraction of plutonium from the water column.^{8,9} However, collection techniques are such that phytoplankton cannot readily be separated from inorganic suspended particulate matter. The reported concentration ratios for algae may, in fact, be high due to the inclusion of inorganic suspended particulate matter which would have a CR value of 10^5 . The correlation between percent silicon content and plutonium concentrations in phytoplankton samples was attributed to the predominance of diatom frustules in the samples analyzed.¹⁰ Wahlgren et al.⁸ reported a correlation between percent ash weight and plutonium concentrations. They also concluded that plutonium was associated with diatom frustules in the plankton samples. However, in neither case was the contribution of associated inorganic, suspended particulate matter to the observed plutonium concentrations measured. They also reported a distribution coefficient for suspended sediment materials of 3×10^5 . The inclusion of a small amount of those materials in the ash residue to phytoplankton samples may offer a plausible alternative interpretation of the observed correlations.

Edgington et al.¹¹ measured the plutonium and americium contents of Lake Michigan sediments, as well as some of their other characteristics. It has been shown that approximately 97% of the fallout plutonium that has entered Lake Michigan now resides in the sediments. Surface sediments in Lake Michigan now contain between 0.14 and 0.4 pCi per gram of dry sediment with the concentration in the water column less than 10^{-3} pCi per liter. Edgington had earlier

indicated that there is a negligible contribution to the input of plutonium from runoff via tributary rivers and streams. In this study the data suggest that there is a significant redistribution of sedimentary material which rapidly moves the radioactivity from its site of deposition on the surface of the sediments to a final site of deposition in the sediment layer. There are apparently large areas of the lake where no significant sedimentation occurs. In these areas there is a layer of floc, containing high concentrations of plutonium, overlying the glacial till or sand. Since no significant accumulations of sediment have occurred in these areas, it is likely that these one to two centimeter thick deposits are transitory and that the material is readily available for resuspension. The average sedimentation rate for Lake Michigan is quoted as about 0.1 cm per year with mixing depths typically 1 to 2 cm. In the chemical studies it was shown that most of the plutonium in the sediments is associated with the hydrous oxides of iron and manganese. Less than 7% of the total plutonium is associated with the sedimentary humic and fulvic acid fractions. In the few cores measured, the ^{241}Am ranged from 0.008 to 0.054 pCi per gram of dry sediment in the top four cm. Their studies of americium-241 with depth in the sediments showed that the americium is being formed in place and shows little or no preferential mobility over the plutonium-239-240.

Wahlgren et al.⁸ measured the plutonium, americium, and uranium in Lake Michigan biota. The results are summarized for americium and plutonium in Table H-VI.

TABLE H-VI
CR FOR ^{241}Am AND $^{239,240}\text{Pu}$ IN
LAKE MICHIGAN BIOTA

Biota	Concentration Ratios	
	^{241}Am	$^{239,240}\text{Pu}$
Phytoplankton	$\sim 1.6 \times 10^4$	1×10^4
Benthos	$\sim 6,300$	1,300
Zooplankton	~ 500	320
Planktivorous Fish	≤ 160	25
Piscivorous Fish	≤ 50	4

Bowen¹² found ^{239,240}Pu CR values for shoreline plants in Lake Ontario which were similar to those reported by Marshall et al.⁵ However, the plankton CR values ranged from 10 to 300 (vs 600-15 000 as reported by Marshall et al.⁵). Further, Marshall et al.⁵ reported CR values for Lake Ontario which ranged from 2 000 to 2 700. Observed differences in these data have not been resolved. Bowen's¹² data agree with those of Marshall et al. that benthic feeders accumulate plutonium to higher levels than limnetic feeders. Among predator species, some forms (i.e., largemouth bass, rock bass, white perch, coho salmon) deposit most of the plutonium in the bone while other forms (i.e., northern pike, yellow perch, freshwater drum) deposit plutonium primarily in the liver. These patterns have not been satisfactorily explained.

Yaguchi et al.¹⁰ have presented a reasonably comprehensive set of concentration factors for plutonium-239 in Lake Michigan as given in Table H-VII. It is noted that the fish were not dissected but were analyzed in their entirety. The relatively high concentration factors noted for the slimy sculpin and several others may be due to the higher access to the sediment by those species which live in close contact with it.

TABLE H-VII
²³⁹Pu CONCENTRATION RATIOS LAKE MICHIGAN
(wet weight basis)

<u>Sample</u>	<u>#Samples</u>	<u>Mean CR</u>	<u>Range</u>
Cladophora	16	3 800 ± 500	1 060 - 6 930
Mixed plankton	22	5 700 ± 800	620 - 15 300
Zooplankton	9	350 ± 60	122 - 653
Mysis	7	760 ± 60	587 - 989
Pontoporeia	2	1 600	1 450 - 1 830
Slimy sculpin	7	250 ± 60	128 - 560
Bloater	8	37 ± 3	21 - 50
Alewife	7	25 ± 2	17 - 30
Smelt	6	20 ± 4	6 - 33
Lake Whitefish	2	14	5 - 23
Coho Salmon	1	6.7	-
Chinook Salmon	1	4.0	-
Lake Trout	2	1.5	1.3 - 1.7

Adams et al.¹³ provided aquarium studies using plutonium microspheres. In the study each of three aquaria was spiked with one $^{238}\text{PuO}_2$ microsphere that was about 100 μm in diameter and was calculated to contain about 68 microcuries of ^{238}Pu . The microsphere was placed so that it was available to the circulating water but was not moved from its position by currents. It was found that in the first nine days the concentration in the aquarium water increased rapidly, with about 0.05% of the plutonium activity in the water of the aquarium. The activity then decreased over the next fifty to seventy days and finally leveled out to about 350 dis/min per liter after about 100 days. The concentration ratios for portions of goldfish and snails which were kept in the aquarium through the entire period but with exposures greater than 100 days are given in Table H-VIII.

TABLE H-VIII
CONCENTRATION RATIOS - ^{238}Pu FOR AQUARIUM

Time of Exposure Days	Concentration Ratios		
	Gut ^a	Flesh	Bone
Goldfish - wet weight			
104	720	30	< 200
181	110	50	< 870
183	1 300	30	< 340
184	2 100	16	< 1 300
185	2 100	23	< 550
Snails - dry weight			
		Flesh	Shell
185		2 300	1 100

^a + Contents

Dahlman et al.¹⁴ reported on the behavior of plutonium in the biotic components of White Oak Lake. Results from analyses of samples of various components of the lake system, as shown in Table H-IX, showed a general trend of decreased concentration of plutonium at higher trophic levels (shad vs large-mouth bass). Organisms living in or on the bottom of sediments of the lake had plutonium concentrations that were two or three orders of magnitude higher than for predatory fish, large-mouth bass, and bluegill. Filamentous algae associated with sediments in shallow areas of White Oak Lake had the greatest concentration of plutonium of any biotic component measured. Levels of plutonium observed in various fish species were related to their feeding habits.

Gastrointestinal contents of gold-fish, shad, and bluegill had plutonium concentrations higher than any biotic form in the system, indicating the source of intake was probably sediment. Further evidence of sediment ingestion by these fish was the fact that the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio of gut contents was similar to that measured in sediments.

TABLE H-IX
CONCENTRATIONS OF $^{239,240}\text{Pu}$ AND CONCENTRATION RATIOS
FOR FISH FROM WHITE OAK LAKE

Species	$^{239,240}\text{Pu}$ Content				Concentration	
	Carcass ^a		G.I. Tract		Ratio ^{c,d}	
	pCi/g	S.E. ^b	pCi/g	S.E.	C.F.	S.E.
Largemouth bass	2×10^{-4}	6×10^{-5}	6×10^{-3}	4×10^{-4}	0.4	0.2
Bluegill	1×10^{-3}	8×10^{-4}	4×10^{-2}	5×10^{-5}	3	2
Goldfish	1×10^{-3}	1×10^{-3}	8×10^{-2}	3×10^{-2}	3	3
Shad	2×10^{-3}	3×10^{-5}	4×10^{-1}	7×10^{-2}	4	0.1

^aTotal fish minus G.E. Tract.

^bStandard Error.

^cWater concentration of Pu used in calculation of CR values was 4×10^{-4} pCi/g.

^dCR values for carcass.

While ingestion of sediment appears responsible for the highest levels of plutonium (body burden) in fish, this mechanism apparently has not enhanced availability of Pu to biota because concentration ratios for biota in White Oak Lake were relatively low compared to those observed at other study sites.

Long-term chemically and/or biologically mediated transformations of plutonium compounds may be expected to occur in aquatic systems. These transformations may result in plutonium being complexed by naturally occurring chelating agents such as carboxylic acids (citrate), fulvic acid, or proteins.

Several laboratory experiments were carried out at Oak Ridge^{15,16,17} to determine the uptake, retention, and distribution of monomeric plutonium (IV) both in an aquatic vertebrate, the channel catfish Ictalurus punctatus, and in a littoral aquatic micro-ecosystem. A primary finding was that gastrointestinal intake by catfish was significantly higher than reported for mammals.

Chelation can either enhance or reduce the uptake of ingested plutonium relative to plutonium hydroxide (monomer) in channel catfish. The highest observed retention (whole body) at 63 days was 3.8% of ingested dose for ²³⁷Pu citrate, while retention of the fulvate was 0.6%. Reduced uptake of the fulvate complex is due either to its high molecular weight (> 10 000) or to its stability in metabolic systems. Increased uptake of plutonium-237 citrate is attributable to instability of the complex in metabolic systems.

Tissue distribution studies revealed that relatively little ($\leq 10\%$) of intracardially injected plutonium citrate was excreted. Blood clearance rates were similar to those found in small mammals, with the plutonium being primarily associated with the plasma protein transferrin. The fractional body burdens in bone, liver, and kidney 17 days after injection were 31%, 24%, and 9% of the injection dose, respectively. High kidney burdens relative to mammals are expected, since the kidney functions as the major site of homopoiesis in teleosts. Absence of significant excretion indicates that a short half-life component of elimination following gut clearance in gavage studies is due to plutonium labeling of the gut.

A distribution coefficient of 9×10^4 was observed for sediment in a year-old aquatic microcosm spiked with plutonium-237 nitrate. A materials balance at 90 days post-spike provided the following estimates: 0.001% in water, 0.04% in biota, and over 99.9% in sediments. Concentrations in whole animals including fish were surprisingly uniform (within a factor of 1.2-9.9% of mean sediment concentration.) This was related to gut loading of

sediments and/or surface contamination. The uptake by rooted macrophytes not exposed to surface contamination was quite small: 0.03 to 0.1%.

Based on this set of laboratory experiments, sorption of plutonium to plant surfaces, on gut walls, and on exoskeletons appears to dominate in submerged components of aquatic systems.

Marine Systems

Pillai, Smith, and Folsom¹⁶ are generally credited with making the first measurements which show significant quantities of plutonium in the ocean and significant concentration ratios for plutonium in biological systems. Their summary of the concentration ratios for ²³⁹Pu is given in Table H-X. These values are related to the average plutonium content of coastal waters since the samples of marine organisms were taken near the southern California coast. Plutonium concentrations ranged from 0.035 dpm/liter for the coastal samples to values of 0.194 and 0.303 dpm/liter for samples from the north Pacific far from the coast.

Templeton¹⁹ provided concentration ratios resulting from laboratory experiments in a fish (plaice) and a seaweed. These ratios are given in Table H-XI.

TABLE H-X
CONCENTRATION RATIOS - ²³⁹PU WET WEIGHT

<u>Sample</u>	<u>Concentration Ratio</u>
Red Tide	600
Green Algae	1 570
Giant Kelp	770
Plankton	2 500
Mussels (soft parts)	230
Mussels (shells)	290
^a Fish (bonito)	3

^aEdible portion and bones.

TABLE H-XI

CONCENTRATION RATIOS FOR PLUTONIUM IN MARINE
ORGANISMS FROM LABORATORY EXPERIMENTS.
(After Templeton, 1959)

Pleuronectes platessa tissue after
140 days exposure

Intestine	6.0
Liver	5.0
Kidneys	3.8
Skin	2.0
Bone	0.6
Muscle	0.77
Periphyra umbilicalis	1 000

Wong et al.²⁰ reported on fish and other specimens caught off Cape Hatteras, Cape Cod; plankton from nets towed on the transect from Long Island to Bermuda; and sargasso weed from the southern part of the Sargasso Sea. For concentration factors the "average north Atlantic sea water" derived from Bowen et al.²¹ was used as a reference. It was noted that the cartilaginous skeleton of the blue shark concentrated plutonium less than the better calcified skeletons of the bony fish. However, in the shark the liver concentration was higher than for any of the bony fish. The two bottom feeding fish, tautog and flounder, had high gut values and high liver concentrations. It was noted that the starfish were collected as they were feeding on mussel beds, and the starfish plutonium concentration was about four times that of the blue mussels on which it was feeding. This raised some question as to the decrease of plutonium content along the food chain. The concentration ratios reported in this paper are given in Table H-XII. However, it is noted in the later paper that some of these concentration ratios were changed due to a change in the calibration of the equipment used. The values in Table H-XII however, include some organisms which were not included in the later paper and the table is included for information.

Noshkin et al.²² have reported the previous data plus others. Interestingly, traces of thorium-228 were found in a number of the samples by

TABLE H-XII
MARINE CONCENTRATION RATIOS FROM
CAPE COD OR OPEN N. ATLANTIC OCEAN
(wet weight)

<u>Organism</u>	<u>Collection Site</u>	<u>Concentration Ratio</u>
Sargasso Weed	Atlantic	9 000
Plankton	Atlantic	2 300
Scalpa	Atlantic	900-2 400 (4)
Starfish	Cape Cod Canal	1 000
	Dyer's Dock	760
Soft shell clam	Hadley	380
Brown mussel	Hadley	290
Blue mussel	Cape Cod Canal	240- 260
	Woodneck	260- 290
	Dyer's Dock	160
Blue mussel shell	Woodneck	410

	<u>Muscle</u>	<u>Bone</u>	<u>Liver</u>	<u>Gut</u>	<u>Gill</u>
Blue shark	4	18	200	-	-
	-	25	150	-	-
	1	-	-	-	-
Blue fin tuna	2	90	-	-	-
Blue fish	5	570	26	-	-
Striped fish	4	160	14	36	14
Blackfish	-	510	64	730	-
Blackback flounder	1	50	57	1 060	-

analyzing the entire alpha spectrum. Concentration ratios for marine invertebrates as found by Moshkin are given in Table H-XIII.

In deriving the concentration ratios they again used the concentration of "average north Atlantic sea water" given by Bowen.²¹ It is of interest to note that Pillai measured considerably lower ²³⁹⁻²⁴⁰Pu concentrations in water along the Pacific coast than out in the Pacific Ocean.¹⁸ It is possible that this factor could effect the values given in Table H-XIII. It was noted here that the marine worm contained the highest concentration of plutonium found in the marine invertebrates. This worm is a non-selective, deposit feeder and ingests quantities of surface sediment. This is significant because the sediments which they inhabit are considerably higher in plutonium concentration than water (i.e., 61 dpm/kg vs 0.0019 dpm/L).

TABLE H-XIII
CONCENTRATION RATIOS OF ²³⁹Pu
FOR MARINE INVERTEBRATES

Organism	Tissue	Mean	Range
Blue mussel	Body	300(7) ^a	250-350
	Shell	490(3)	470-520
Brown mussel	Body	340(1)	
Soft-shell clam	Body	440(1)	
Oyster	Body	130(2)	100-160
Scallop	Adductor	24(2)	10-37
	Body	520(3)	410-690
	Shell	600(1)	
Whelk	Body	140(1)	
	Shell	300(1)	
Moon shell	Body	660(1)	
	Shell	690(1)	
Starfish	Body	1 020(2)	
Brittle star	Body	760(1)	
Marine worm	Body	4 100(1)	
Sponge	Body	2 100(1)	
Sargasso weed		2.1 x 10 ² (6)	0.3-10 x 10 ² ^a
Other seaweed		6.2 x 10 ² (6)	1-16 x 10 ²

^aNumber of samples averaged.

Hetherington et al.²³ have described in some detail the marine consequences of a controlled disposal of plutonium and americium from the British Nuclear Fuels Windscale Works to the Northeast Irish Sea. The quantities of such discharges by year are given in Table H-XIV. The liquid wastes are discharged through a pipeline extending out to sea 3.2 km in a depth of 20 m. The studies have indicated that a very large fraction of the plutonium, on the order of 96%, is removed to the sediments very soon after its release. Data on the resulting concentrations of transuranics in surface sediments are given in Table H-XV. It is of interest that the ratio between plutonium and americium does not vary greatly with distance from the outfall and from this it is concluded that americium discharged in this form behaves qualitatively similar to plutonium. These sediments were collected in July, 1974, and reflect the variation in the ratios of plutonium to americium in the discharged wastes shown in Table H-XIV. From core samples in the sediments it was deduced that the recent sedimentation rate in the area is approximately 2.3 cm per year. A summary of the concentrations found in the region within 10 km of the discharge during 1974 is given in Table H-XVI. Here it is of interest that the seaweed (*porphyra umbilicalis*) is gathered as an edible crop for people and surveys have shown a range of consumption rates up to 172 grams per day. In an earlier paper on this discharge, Dunster et al.²⁴ have calculated derived working levels based on the ICRP drinking water standards. The values for plutonium are given in Table H-XVII.

TABLE H-XIV

DISCHARGE OF TRANSURANICS INTO
THE NORTHEAST IRISH SEA (C_1/MO)

Year	Plutonium	^{241}Am
1968	69	48
1969	68	33
1970	78	45
1971	94	85
1972	129	181
1973	148	246
1974	104	266

TABLE H-IV
CONCENTRATIONS OF TRANSURANICS IN
SURFACE SEDIMENTS (pCi/g)

<u>Distance from Discharge</u>	<u>Plutonium</u>	<u>²⁴¹Am</u>
1	44 ± 4	56 ± 6
4.5	105 ± 7, 28 ± 2	113 ± 11, 49 ± 5
9	10 ± 1, 14.0 ± 0.5, 35 ± 2	15 ± 2, 17 ± 2, 47 ± 5
18.5	4.0 ± 4	5.2 ± 0.5
110	0.70 ± 0.06	0.7 ± 0.07

Hetherington has described in some detail the consequences of a controlled disposal of plutonium and americium from the Windscale Works to the marine environment. He concluded:

"In view of the speed with which this plutonium from Windscale disappears from the water, it is not considered likely that the mechanism for its removal is biological as proposed by Bowen et al.²¹ on the basis of their own work and that of Pillai et al.¹⁸ Rather it is assumed that the loss is due to the rapid hydrolysis and consequent precipitation of the plutonium present initially in the effluent in a soluble phase, either as a hydrous compound of the element itself or in association with suspended material already present in the seawater. Indeed, since the pH of the effluent is adjusted to achieve neutrality before discharge, and on the assumption that plutonium behaves at very low gravimetric concentrations in a manner which is consistent with its usual chemical properties, hydrolysis and colloid formation have most probably occurred before the effluent ever reaches the seawater. Alternatively, if colloid formation has not already taken place, then the element must appear in the seawater in a state showing great affinity for any solid material available. Whatever the mechanism, the measurements made on the

TABLE H-XVI
CONCENTRATION WITHIN 10 km OF DISCHARGE
DURING 1974 (pCi/g for a 1 Ci/da
DISCHARGE RATE

Material	Pu	Am
Sediment	10	5
Seaweed	1	1
Fish Flesh	0.004	0.003
Sea Water	0.0002	-

suspended solid fractions of the seawater samples have shown that the process leads to the removal of the plutonium from both suspension and solution, while the relatively high value for the fraction of the water-borne plutonium which passes a 0.22 μ m filter (72% averaged over 15 samples) suggests that the residue remaining in the water is present either as a colloidal suspension or in some other complexed form."

Concentration ratios derived from Heatherington²³ are given in Table H-XVIII.

Ward²⁵ studied the uptake of ²³⁹Pu by lobsters from aerated, filtered sea water. The lobsters were exposed in partitioned glass tanks in a room with a constant temperature of 9° to 12°. Every few days the lobsters were transferred to fresh tanks of sea water containing 6.5×10^{-2} μ Ci/L of plutonium over a period of about 230 days. Concentration ratios for flesh ranged up to three with, however, an indication that they may still have been increasing at the end of the experiment. The calcified shell appears to accumulate plutonium-239 at a rapid rate. A very high proportion (89.5%) of the total plutonium is in the skeleton which accounts for about 43% of the total weight of the lobster. Concentration factors for shells were on the order of 200 and gills were about 100. The flesh which, comprises about 28.7% of the total body weight, contains only 1.2% of the plutonium present in the entire body. The content of the cast shell is approximately twice that of the shell of the intermoult lobster. It is of interest that in this experiment the uptake of the tracer was only from the

TABLE E-XVII
DERIVED WORKING LEVELS - ^{239}Pu (pCi/g)

Fish (edible parts)	500
Seaweed (edible)	150

water, since the lobsters were fed on uncontaminated mussels. This could possibly indicate that higher concentration factors could be expected in animals that also obtained their food from the contaminated environment.

In a study of crayfish from the Great Miami River, Wayman et al.²⁶ reported that most of the plutonium was concentrated in soft tissues rather than in the sclerotized shell. Similar results were reported by Nelson and Noshkin²⁷ for the *Iridaea* clam and lobster from the Eniwetok Atoll. Noshkin²⁸ reported higher concentrations of $^{239-240}\text{Pu}$ in the shell than in the body of scallops, whelks, and moonshells collected off Cape Cod. Whether the differences reported are due to the chemical species of plutonium the organisms were exposed to at the various sites, or interspecific physiological differences, is open to question at this point. Recent studies have reported that plutonium isotopes can apparently behave differently in biological systems.^{3,29,30,31} In environmental studies, the discrepancies in behaviour can be explained by different physiochemical forms of the isotopes in the original source or to different sources of uptake. In laboratory experiments, differences in metabolic behavior can be attributed to differences in concentrations of the isotopes tested. Fowler et al.³² found that if the different plutonium isotopes (^{236}Pu , ^{237}Pu , ^{238}Pu , $^{239-240}\text{Pu}$) were present in the same physiochemical form, aquatic organisms were unable to discriminate between them, either in accumulation or excretion studies. They also concluded that the suggestion of Moghissi and Carter³³ and Ryman et al.¹⁵, that, for environmental studies, ^{237}Pu is most likely the best tracer for measuring plutonium kinetics in biological systems is correct.

Discussion

This review is not intended to be definitive; rather, we have attempted to synthesize the best available data. Although there appears to be a similarity among all environmental data, whether marine or freshwater, there are

TABLE H-XVIII
CONCENTRATION RATIOS FOR ^{239}Pu IN MARINE BIOTA
IN IRISH SEA WINDSCALE VICINITY - 1974
(Wet weight)

<u>Organism</u>	<u>Tissue</u>	<u>CR Value</u>
Seaweed	whole	1 000
<u>Vertebrates</u>		
Plaice	muscle	10
Dab	muscle	20
Whiting	muscle	6
<u>Mollusca</u>		
Mussel	soft tissue	2 000
Winkle	soft tissue	1 000
<u>Crustaceans</u>		
Crab	muscle	100

significant discrepancies. The most interesting common factor we were able to extract from a wide range of studies, both laboratory and field, is the similarity in the observed distribution coefficient for plutonium given in Table H-XIX. Primary conflicts involve differences in concentration ratios for freshwater algae and the tissue distribution observed in both freshwater and marine studies. These discrepancies are probably explainable based on differences in physiochemical form to which organisms are exposed at different sites.

However, in reviewing the data available for both fresh water and marine environments, we can see little real difference in the patterns of accumulation or in the actual values of concentration factors. The data from the waste pond at Hanford, while obtained under very unusual conditions, do serve, however, to indicate that there may be variations in specific situations well beyond those studied in this review.

An additional difficulty we encounter is the lack of any definitive information on the other two elements of interest - Am and Np. One study of neptunium⁶ in the Columbia River involved an isotope with a half-life of only 2.3 days so that the radioactive decay may well have limited the quantities of

TABLE H-XIX
DISTRIBUTION COEFFICIENTS FOR PLUTONIUM
ISOTOPES IN FRESHWATER AND MARINE SYSTEMS

K_d	Isotope	Environment	Reference
9.0×10^4	237	L,F	Trabalna & Eymann, 1976 ¹⁷
$1.3-9.4 \times 10^4$	237	L,M	Duursma & Parsi, 1974 ³⁴
$1.2-7.9 \times 10^4$	239	L,F	Trabalna & Frank, 1976 ³⁵
4.8×10^4			
1.3×10^5	239	M	Pillai & Mathew, 1975 ³⁶
5×10^4	239	F	Bowen, 1974 ¹²
3×10^5	239	F	Wahlgren et al., 1975 ⁸
5×10^4	239	M	Hetherington et al., 1975 ²³

*F = Freshwater M = Marine L = Laboratory

this isotope in the organism. Data from Lake Michigan indicates increased concentration factors for ^{241}Am over plutonium in the lower trophic levels by factors of 1.5 to 5.⁸ However, the values for fish are indeterminate with maximum values of increase over plutonium of 6-10. Data from the discharge of waste from Windscale²³ indicate little difference between ^{239}Pu and ^{241}Am in concentrations within 10 km of the discharge for equal discharge rates.

In selecting concentration factors for application, it is necessary to consider all possible uses by man. This is well illustrated by the edible seaweed in the Irish Sea. The use of shark liver for its oils as well as eating by some groups is another example that is of interest, particularly since the liver appears to be more effective in concentrating plutonium than the muscle. The future potential use of other products from the sea in various forms could also be of importance.

For purposes of calculation we have chosen concentration ratios for the three nuclides. These are not the highest measured nor are they the average, but, rather, values that appear reasonable for general use in light of present knowledge.

For Am and Np (^{241}Am , ^{237}Np) we have applied an arbitrary factor of 10 over the plutonium values. These values were chosen because these isotopes have a greater availability than plutonium in terrestrial systems and it is suspected that when data are available the same may be true in aquatic systems. Limited data from Lake Michigan and Windscale indicate the factor for ^{241}Am may be less than 10. However, due to the incomplete nature of the data, we have chosen 10 as the factor for biota.

Concentration factors chosen for use in both freshwater and marine environments are given in Table H-XX.

The very high affinity of plutonium for particulate matter in aquatic ecosystems (distribution coefficient $\sim 10^5$) makes it difficult to use the traditional expression of concentration ratio (CR) as a measure of the tendency of biota to accumulate this element in tissue. Rather, we feel the observed concentrations of plutonium in aquatic biota should be related to the primary abiotic source in the system, sediment (both suspended and bottom). In order to express this relationship, the term Trophic Transfer Factor (TTF) has been used by various researchers (Lipke 1971;³⁷ Trabalka and Ryman;¹⁷ Elwood and Hildebrand.³⁸) This concept substitutes the concentrations of an element in sediment and suspended particulate matter for the concentration in water normally used in the calculation of a concentration ratio. The underlying expectation is that, due to the high K_d 's, observed element accumulation in

TABLE H-XX
CONCENTRATION RATIOS RECOMMENDED

<u>Material</u>	<u>Plutonium</u>	<u>Americium Curium Neptunium</u>
Sediment	100 000	100 000
Plankton	5 000	50 000
Benthic Algae and Macrophytes	5 000	50 000
Benthic Invertebrates	1 000	10 000
Fish		
Bottom feeders	250	2 500
Plankton feeders	25	250
Piscivorous (fish eaters)	5	50

tissues of higher trophic levels will be dominated by gut absorption rather than direct uptake from water. Again, we stress that external contamination with sedimentary particulate matter and gut loading are not considered to represent true uptake and should be considered separately. This term, then, serves as a realistic measure of plutonium discrimination in food chains.

Some of the variation in TIF values observed can be explained by the relative trophic position of the organisms analyzed. The number of intervening food chain transfers between the organism analyzed and the abiotic source of plutonium should be inversely related to the observed TIF value. Therefore, in assessing potential transfer of plutonium to man from aquatic ecosystems, it is important to concentrate on those food sources most closely linked to sediment as a measure of maximum plutonium in human food. These would represent a short, single trophic transfer food chain as opposed to the traditional concept of the grazer food chains. Examples of important groups include bottom-feeding fishes, shell fish, and rooted macrophytes such as rice. A major dietary component of a large segment of the world population is rice. Although we could find no data on accumulation of plutonium in rice, this information is important since it is representative of a single trophic transfer from sediment and/or water to man. Moshkin²⁸ pointed out that marine organisms associated with the sediment-water interface, (i.e., benthic invertebrates) contain one hundred times higher plutonium burdens than marine free-swimming vertebrates.

Future research on pathways of plutonium to man from aquatic ecosystems should concentrate on those food chains which have the smallest number of trophic transfers between abiotic sources in the system and man. Additionally, emphasis on the relationship between the chemical characteristics of plutonium in abiotic components of the system and availability for short food chain transport to man should be stressed.

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APPENDIX I

MODELING RADIONUCLIDE TRANSPORT THROUGH SOIL AND ROCKS BY SUBSURFACE WATER

John C. Rodgers

I. INTRODUCTION

Once nuclear wastes are introduced into the subsurface environment by shallow land burial, a possible release pathway from the burial site to man is the mobilization of radionuclides into flowing groundwater. Contaminated groundwater could be obtained for use by man near the source by wells, or transport over long distances to surface waters may be required before interaction with man's environment occurs. Several factors associated with the placement of wastes in the subsurface environment, and the process of transport of radionuclides as solutes in groundwater, such as dilution, dispersion, and retardation of movement by soil-solute interactions, serve to create a natural barrier between the waste and man. Because of the complexity of the solute transport process, and because of the lack of generality and inability of field data to support extrapolation over long times and large spaces, site evaluation and risk assessment of migration in groundwater has relied heavily on mathematical simulation. Following is a review of some of the salient features of several approaches to the problem in current literature, and an evaluation of the predictions of a simple one-dimensional mathematical simulation of groundwater transport.

II. UNSATURATED - SATURATED FLOW

Typically, shallow land burial of radioactive waste involves placement of the wastes well above the top of the water table.¹ This means that most often release and transport of radionuclides by groundwater involves a phase where the wastes are in contact with soil moisture moving as unsaturated flow above the water table. However, release may occur directly into the zone of saturation, as for example, in the case where water mounding below a trench intersects the waste. The predominant direction of unsaturated flow is vertically downward until the flow reaches the zone of saturation. Within this zone, flow is principally lateral. The processes that control the transport of dissolved radionuclides in a hydrogeologic system include: 1) hydrodynamic dispersion combining the effects of mechanical dispersion and molecular diffusion. 2

convection of the solute by moving water, 3) physical-chemical reactions between solutes and the soil matrix (eg., ion-exchange), and between solutes eg., precipitation or coagulation), and 4) radioactive decay. Mathematical simulation of saturated - unsaturated flow and material transport requires the solution of the set of coupled differential equations for the movement of gas and water, and the differential mass transport equation. It also requires formulation of appropriate mathematical relationships describing the physical - chemical reactions mentioned in 3) above, a task not easily and satisfactorily done.

III. NUMERICAL MODELS

Until rather recently only separate portions of this overall simulation problem could be solved by various analytic and numerical techniques. The most powerful mathematical approach has been the use of numerical techniques. An important example is the work of Robinson and others,² who used the method of characteristics to describe the two-dimensional transport of radionuclides in the Snake River Plain aquifer (saturated flow). The model includes the effects of convection transport, flow divergence, two-dimensional hydraulic dispersion, radioactive decay, and reversible linear sorption (instantaneous equilibrium). The hydraulic phase of the model uses the iterative, alternating direction, implicit finite - difference scheme to solve the groundwater flow equations, while the waste transport phase uses a modified method of characteristics to solve the solute transport equations.

Reportedly,³ such numerical methods as this suffer from numerical dispersion effects and other limitations. Another numerical technique which virtually eliminates numerical dispersion is the Galerlein finite-element method for solving partial differential equations.³ An example of this approach is the two-dimensional material transport model developed at ORNL by Duguid and Reeves.⁴ It is designed to consider a combination of saturated and unsaturated flow. The water transport equation in this code is solved for specified boundary conditions by numerical methods using Galerlein finite-element methods. The region of interest (e.g., soil column under a waste trench) is represented by a two dimensional grid of subdomains requiring specified boundary conditions at boundary nodal points. One portion of the code solves the moisture transport equation, requiring such inputs as precipitation, moisture content as a function of pressure, and conductivity as a function of pressure. Also soil properties

such as compressibility, bulk density, and effective porosity must be specified. Another portion of the code uses the fluxes generated in the first portion, coefficients such as soil dispersivity, a retardation factor for solute movement based on specific radionuclide distribution coefficients, and radioactive decay rate for solution of the mass transport equation. This part of the code then provides the quantities of material in solid and liquid phases. The code is not easily implemented, due to the considerable detail required in the input deck.

IV. ANALYTIC TRANSPORT MODELS

While not as powerful as numerical methods in terms of such applications as precise simulation of heterogeneous field conditions at a particular site, analytical solutions of the mass transport equation (assuming saturated flow and certain types of generalized boundary conditions) have a useful role in identifying critical variables and processes, magnitude of effects, etc.

An example of this type of simulation is a set of related one-dimensional codes based on analytical solutions to the mass transport equation for a decaying chain of radionuclides moving through a geologic medium developed by Burkholder and others.^{5,6,7} The approach is to model transport of a chain of radionuclides assuming specifically: impulse or band release, water velocity, distance to breakthrough point (receiving water body), down-gradient (axial) dispersion coefficient, equilibrium adsorption constant, and soil bulk density and porosity. The codes can be applied to heterogeneous soil columns by assuming appropriate weighted average values of model input parameters, and to faulted, monolithic media provided the adsorption equilibrium data were obtained properly.⁶ Some of the predictions of interest of these models include, 1) the difference in adsorption characteristics among the nuclides in the decay chain, the migration path length, the leach rate and the half-lives of the nuclides can, in some circumstances create a condition where the predicted maximum (but not average) nuclide discharge to the biosphere is greater for certain nuclides than their corresponding value for direct discharge without transport through soil or rock⁷, 2) in other cases these conditions can cause the inventory of a given nuclide to exit the soil column over the entire period between the times its precursors exist and the time the initial inventory of that nuclide exists thereby greatly reducing the discharge rate at any one time⁵, and 3) axial dispersion significantly reduces the maximum discharge rate of the first chain member and the slowest decay chain member, but only slightly reduces the

discharge rates of other chain members with significantly faster decay rates.⁵ The data needs of these codes are difficult to meet, especially equilibrium sorption coefficients for members of the several TRU decay chains in the wide range of soil and groundwater conditions, particularly at some depth.

Another sort of one-dimensional transport model being developed by Serne and others⁸, illustrates the flexibility of the analytic approach. It is designed to closely model the soil-leachate chemical interactions as they affect sorption. Thus diffusion and dispersion are neglected and a Lagrangian coordinate system (following a fixed mass of fluid as it moves through space) is utilized. The model is a discontinuous equilibrium mass-balance model in which solution concentrations of ions present in both macro-amounts (chemicals) and trace amounts (radionuclides) are accounted for at all times. Empirical equations relating trace solute sorption to equilibrium macroion concentrations, derived by regression from data on soils of interest, are used in the code to calculate trace solute concentration. Macroion reactions considered include ion-exchange, and carbonate, sulphate, and solution ion interaction equilibria for Na, K, Ca, and Mg. The code has been verified for the movement of ⁸⁹Sr and ¹³⁷Cs through two types of Hanford soils. Application to the migration of TRU contaminated leachate through soils is hampered by deficiencies in present understanding of environmental actinide soil chemistry and the composition of TRU waste leach solutions.

V. DISCUSSION

The single parameter which most significantly affects the predicted movement of radionuclides through soil columns in any of these models is the equilibrium distribution coefficient, K_d . It is imperative to be aware of the limitations of this parameter, and the means for its measurement when applying any of these models. K_d is a lumped parameter, representing many (but perhaps not all) sorptive and retardation processes occurring between mobile radionuclides and soil. It is a function of many soil and leachate parameters including pH, redox potential, solute chemistry, radionuclide concentration, mineralogy of the soil, horizon through which transport occurs, and, in the unsaturated zone, moisture content. For a given mineralogy it is a function of the surface area of the porous medium. Hence, applicability of laboratory measurements of K_d may have limited (or worse, indeterminate) applicability to field conditions being modeled. Also extrapolation of K_d values from one area

or soil type to another can be extremely problematic. Since sorptive processes are strongly dependent on surface area and contact time, sorption is considerably less effective in fractured materials where flow occurs with maximal velocity and, minimal contact with surfaces. In such cases the predictive value of K_d based on core samples and laboratory tests (batch tests) is marginal.

An example of a two-dimensional model of the release and transport of TRU from a burial site will be presented here to illustrate many of the points raised in the foregoing review.

In most general terms, the two-dimensional model of ground-water transport of TRU contaminants from a shallow earth burial site will be represented by the simplified concept of an extremely large reservoir of contaminant being released at a constant, continuous rate through a finite plane surface at right-angles to a homogeneous, isotropic aquifer of finite depth, but otherwise unconfined extent. Movement of water and contaminant occurs in the X-Y plane, X being the down-gradient dimension. The mathematical boundary conditions are:

$$X=0, t \geq 0, C = C_0$$

$$X > 0, t=0, C = 0$$

(1)

$$X \rightarrow \infty, t > 0, C = 0,$$

where C = concentration of TRU activity in water, C_0 = concentration in the water directly under the release point ($X=0$).

The boundary conditions will be applied in two steps: first the source term will be computed, then the down-gradient transport will be described by a solution of the diffusion-transport equation.

For the source term, assume as an approximation that the total TRU activity of the site, Q , is being leached, with a constant fraction f , lost each year for a very long time. Then $Q_1 = Qf$ (Ci/yr) is the total rate of contaminant input to the aquifer. The model assumes that the release occurs into a plane of width equal to the width of the site and depth equal to the depth of the aquifer. Thus the holdup and dispersion occurring in whatever soil column intervenes

between the pit and the top of the aquifer is ignored for simplicity. The elemental volume in which the released activity is dispersed initially is this plane area times an incremental down-gradient distance ΔX . The total concentration in the contaminated frontal volume of soil and water is thus the product of the release rate per elemental volumes and the time required for a given water volume to traverse the incremental down-gradient distance:

$$C_t = \left(\frac{QI}{A\Delta X} \right) \cdot \left(\frac{\Delta X}{v} \right) = \frac{QI}{Av} \left(\frac{C_1}{\text{cm}^3} \right) \quad (2)$$

where A = plane area, and v = water velocity.

It will be assumed that the water velocity is low enough that equilibrium can be fully established between the leachate and soil. All aspects of the distribution of contaminant between soil and water will be described by the distribution coefficient K_d . The total activity is divided between soil and water, with water occupying a volume determined by soil porosity. Hence, for a soil with density ρ and porosity ϵ , the activity concentration on the contaminated front is distributed as:

$$C_t = C_s \rho + C_w \epsilon = K_d C_w \rho + C_w \epsilon \quad (3)$$

Solving for the activity in the water phase,

$$C_w = \frac{C_t}{\rho K_d + \epsilon} \quad (4)$$

Substituting from equation (2),

$$C_w = \frac{QI}{Av(\rho K_d + \epsilon)} \quad (5)$$

When the sorptive capacity of the soil at a particular down-gradient location becomes exhausted by continuous input of contaminants from the site, "breakthrough" occurs, and the concentration of TRU in soil water becomes:

$$C_w = \frac{Q_f}{Avc} . \quad (6)$$

Equation 6 is the simplest, most conservative model of impact of the release at any given location since no estimation of retardation and dispersion over large distances or long times is required. Of course, the source activity will be reduced by leaching and radioactive decay. This decrease will not be included in the following derivations for simplicity. The direct expression of the simultaneous effects of dispersion and retardation in the down-gradient aquifer on this initial contaminant front is modeled by solving the hydrodynamic dispersion and transport equation for the given boundary conditions. In two-dimensions, the equation for decaying contaminant concentration C is:

$$R_d \frac{\partial C}{\partial t} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} - v \frac{\partial C}{\partial x} - \lambda R_d C , \quad (7)$$

where

- D_x, D_y = x and y dispersion coefficients, respectively,
- R_d = retardation factor defined as, $R_d = (1 + \rho K_d / \epsilon)$,
- v = down-gradient water velocity,
- λ = radioactive decay constant,
- C = concentration in water.

The instantaneous solution of equation (7) at time t' for boundary conditions given in equation (1), and assuming an area source perpendicular to the direction of flow as described, with constant concentration C_0 , and with a width L centered at the origin is:

$$C(x,y,t;t') = \frac{C_0 v/R_d}{\sqrt{4\pi E_x(t-t')}} \exp \left\{ \frac{[x-(v/R_d)(t-t')]^2}{4E_x(t-t')} + \lambda(t-t') \right\} \left\{ \operatorname{erf} \left(\frac{y+L/2}{2\sqrt{E_y(t-t')}} \right) - \operatorname{erf} \left(\frac{y-L/2}{2\sqrt{E_y(t-t')}} \right) \right\} \quad (8)$$

where $E_x, E_y = D_x/R_d$ and D_y/R_d , respectively and $\operatorname{erf}(x)$ equals

$$\frac{2}{\pi} \int_0^x e^{-t^2} dt.$$

The effect of a continuous release from start to any time t is the time integral of equation (8):

$$C(x,y,t) = \int_{t'=0}^{t'=t} C(x,y,t;t') dt' \quad (9)$$

Figure 1 presents the results of a computer evaluation of integral (9) with water velocity $v = 30$ cm/day, $K_d = 100$, dispersivity in the y -direction of 2 ft²/day, and a variety of dispersivities downgradient.

The effects of increasing dispersion are seen to be twofold. First, the contaminant appears sooner at a given place than would be expected assuming little or no dispersion; and second, there is a considerable delay in attaining levels of concentration at the fully saturated value. For example, with relatively high dispersion ($D_x = 604$ m²/day), centerline water concentrations 16 meters downgradient are 10% of their ultimate value over 100 years earlier than would be predicted under conditions of extremely low dispersion. However, under these conditions the water concentration has not reached 50% of the saturated value in 1500 years.

The introduction of cross-gradient dispersion does not appear to alter the predicted centerline concentration greatly.

Fig. 1. Effects of Increasing Dispersion.

These computations illustrate how the details of the moving front of contaminant are modeled by a solution to the hydrodynamic transport equation. At the same time they emphasize the conservatism of the simple, saturated model.

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APPENDIX J

TRU WASTE ASSAY INSTRUMENTATION

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There is strong evidence that transuranic (TRU) waste assay is vital to the overall waste management programs of the DOE. In the first place there can be a significant cost/benefit relationship by reducing the volume of wastes that are classified as TRU wastes and that require expensive retrievable storage. Secondly, for waste repositories, such as the proposed WIPP, there is a need for assay of all or a representative sample of incoming containers. This provides quality assurance for the operating and health and safety aspects of the plant. (There have been too many examples in the past of waste storage facilities accepting the stated contents of the container, when, in reality, the container held vastly greater amounts and, hence, presented potentially large safety and other problems. There must always be a receiver assay capability to minimize mistakes and avoid later surprises.) In the discussion that follows, a brief description will be given of the various waste types and the applicable assay instrumentation. Emphasis will be placed on detection limits and assay accuracies, equipment and operational costs, and evaluation of the operational characteristics and assay limitations.

Basically, DOE, or "defense", wastes can be divided into two major categories of low and high density. On occasion, these wastes can be contaminated (besides the TRU material) with small amounts of fission products. Wastes from the commercial nuclear power industry, on the other hand, are by definition heavily contaminated with fission products. At the present time, and except for some wastes from D/D (decontamination/decommissioning) projects, only the low density portion, the "combustibles", of the DOE wastes can be assayed at the 10 nCi/g level. These wastes make up approximately half of the DOE wastes by volume. The other half of the DOE wastes, the high density wastes, and all of the commercial wastes are presently inaccessible to nondestructive assay. Development Programs now underway or planned for these areas are discussed below.

DOE Combustible Waste

For low density wastes that are essentially free of interfering fission

products, several nondestructive assay systems have been built. The first MEGAS¹ (Multi-Energy Gamma Assay System) design consisted of a single large area NaI detector, with a thin beryllium entrance window, and a mechanical turn table that moved the waste package past the stationary gamma and x-ray detector. Four of this model have been constructed and are in operational use at plutonium facilities. The system assays 2 ft³ cardboard containers holding from 5-10 kg of combustible wastes. The system automatically weighs each package and performs the TRU analysis in 200 sec counting times. The analysis is read out on a computer terminal in units of total nCi/g, with separate answers for plutonium and americium. Detection limits are well below 1 nCi/g with assay accuracies in the $\pm 40\%$ range. Instrumentation costs are approximately \$35K with operational costs (the operator) approximately \$20K per year. Such costs can be quickly recouped, however, when much of the wastes are shown to be less than 10 nCi/g. For example, at Los Alamos in 1977 over \$200K was saved through the use of the MEGAS in assaying the room wastes generated in the plutonium processing facility. These savings were determined by calculating the difference between handling all the wastes as retrievable TRU wastes (at \$50/ft³) and handling only that waste measured to be above 10 nCi/g (8% of the total) as retrievable. The other 92% of the waste was buried nonretrievably with the burial estimated to cost, conservatively, \$5/ft.³. A single instrument can easily assay all of the combustible wastes generated in a large plutonium facility. The system is easy to operate by moderately skilled technicians and is reliable. However, the system can be fooled by two things. The first is high density wastes where the TRU material inside the package is shielded from the detectors by the waste itself. The second problem with the system is the interference from any beta/gamma emitting fission products. Above fission product contaminations of the order of 100 μ Ci/package², the system is no longer capable of 10 nCi/g detection. A new design of the MEGAS will alleviate this problem and is discussed below.

The MEGAS II² has a similar NaI detector as the original MEGAS. Added are two additional detection systems: a high energy resolution hyperpure germanium (HpGe) detector and four ³He neutron detector banks.³ The HpGe detector will tolerate more fission products than the NaI detector, as will the neutron banks which are very insensitive to gamma rays. The neutron banks will allow the assay of higher density wastes, not at the 10 nCi/g level, but at much higher levels, perhaps nearer to several thousand nCi/g. The importance of the neutron

system on the MEGAS II is that significant amounts, gram fractions, of plutonium cannot be taken out in the waste, even in the presence of high density materials and/or large amounts of fission products. The MEGAS II costs approximately \$60K with similar operational costs as the original MEGAS. Because the MEGAS II has not been installed in a facility as yet, its operational characteristics have not been determined at this time.

DOE Decontamination/Decommissioning Waste

A special category of high density wastes is that generated from D/D programs as the DOE begins decommissioning outdated plutonium facilities including the exhumation of old waste burial grounds. For some types of waste generated in these operations, there are nondestructive assay techniques that are currently being used with success. For surface contamination such as on the interior surfaces of plutonium gloveboxes, two new instruments have been developed.² The first is a high range alpha probe (air ionization counter, 60 cm² surface area) that is placed on the surface to be measured. The electronics are capable of counting rates up to 200 million cpm. A second, and complimentary method, is based on a thin NaI detector that is held above the surface to be measured. Here, the low energy x-rays and gamma-rays emitted by the TRU material following alpha decay are measured. For the glovebox measurement mentioned above, the NaI detector is placed into a clean non-leaded glove from the outside and the measurement performed through the glove wall. The direct alpha measurement, on the other hand, must be made from the inside of the box due to the very limited range of the alpha particles. Both instruments have a very large low bias if lumps of TRU material are present or TRU material is hidden behind dense objects such as gaskets and seals. Precluding this, detection limits are well below 10 nCi/g. Costs of the equipment are less than \$10K for both instruments. Measurement times are short, with entire gloveboxes being assayed in less than 60 minutes. Medium-skilled, technician operators are required.

In the exhumation of outdated waste grounds, several nondestructive assay techniques have been developed for field survey of soils at less than 10 nCi/g activity levels. One method is the portable phoswich system^{4,5,6} that is carried in a backpack by a man. The phoswich detector is a combination NaI-CsI sandwich, photon scintillator coupled to a single phototube. Special electronics reduce the detector background to approximately three times lower

than the conventional field FIDLER⁷ detector. Detection limits are 1 nCi/g for plutonium and 100 pCi/g for americium in a real-time scan mode. System costs are approximately \$10K. Medium skilled technician operators are required. The systems have been field tested for several years with good success in all types of weather conditions. The presence of other radioactive materials, such as fission products, will fool the system, however, and material buried more than several cm below the surface may not be seen.

A second soil assay system is a portable ZnS detector⁶ that is placed directly over a dish of raw soil that has been dried on the surface. The system counts alpha particles directly from the very top layer of soil in the dish. Accuracies, compared to wet chemistry, are generally in the $\pm 90\%$ region. Detection limits are near 25 pCi/g for any alpha emitter. Equipment costs are less than \$5K, with very low operator costs with medium-skilled technicians. Assay times are in the 5 minute range. The system will not see "hot" particles located under the top surface of the soil in the sample dish, so that sampling errors can be very significant as with wet chemistry results. The system is not bothered to any extent by the presence of fission products. Note that both the field phoswich and the ZnS systems have application² to the assay of large amounts of soil passing by fixed detector heads as part of large exhumation projects of waste burial grounds.

DOE High Density Wastes

At the present time, there is no satisfactory method to assay high density TRU wastes in bulk form at the 10 nCi/g level. A program² is presently underway to develop active interrogation methods (at or near the 10 nCi/g level) using external sources of high energy photons and neutrons to induce fissions in the TRU material. Initially, a 30 MeV electron linear accelerator (LINAC) is being used to generate the necessary photon beam and, later, to produce neutrons for preliminary studies. The LINAC is presently being used because of its availability and capability for such studies. Accelerators eventually used for this purpose will be of smaller energy, size, and cost. Sealed tube and Cockcroft-Walton neutron generators are likely candidates. It is worth noting that small LINACs and Cockcroft-Walton generators have already been mounted in truck trailers and moved around the United States for the purpose of assay of much larger quantities of TRU materials (well above 10 nCi/g levels) in support of nuclear safeguard programs. Also developed in the safeguards program are

passive spontaneous fission neutron counters⁸ for the assay of gram fraction quantities of plutonium contained in 55 gal. drums. The systems are aimed at scrap generated at a plutonium facility rather than the much lower level wastes of interest here.

Commercial Nuclear Power Industry Wastes

The active interrogation methods discussed above for use on high density DOE TRU wastes has potential application to TRU wastes heavily contaminated with fission products from the commercial sector. It is possible that a single assay technique might be used for both DOE high density wastes and commercial wastes of any density. The active interrogation program of Ref. 2 is addressing this.

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APPENDIX K

NATURAL ENVIRONMENTAL SOURCES OF ALPHA-EMITTING RADIONUCLIDES

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I. INTRODUCTION

There are considerable variations in levels of exposure to natural radiation around the world. But man has not systematically avoided regions of elevated natural radioactivity; on the contrary, many thermal springs containing high concentrations of natural radioactivity in water, sands, and air have been sought out and developed as health resorts for centuries. Spas became particularly popular in the 19th century some years before the discovery of radioactivity. Subsequent to the discovery of radioactivity and the association between elevated natural radioactivity and some mineral or thermal springs, a belief grew that there is some beneficial effect of radioactivity on health.¹ This belief persists in some people today.²

The purpose of the following discussion is to review the range of levels of natural alpha-emitting radionuclides in soils and rocks in different localities around the world in order to develop a more quantitative perspective on certain kinds of natural radiation environments in which man lives.

II. UNITS

The quantities of natural radioactive materials are most frequently given in parts per million (ppm, or $\mu\text{g/g}$). But for purposes of discussion and comparison with transuranium elements in wastes, it is helpful to convert the weight fraction to the radioactivity of the materials of interest. This requires some assumptions and conversion factors for the two principal natural decay series of interest here, the uranium (^{238}U) and the thorium (^{232}Th) series.

Tables B-IV to B-VII in Appendix B give the details of the decay products and the half-lives of the various members of the uranium and thorium series. Table K-I lists the major alpha emitting daughters along with the activity associated with each while in equilibrium with the parent of the chain.

There are, however, influences in nature that will cause some disequilibrium to occur among the daughter nuclides. Thus, if water passes

TABLE K-I

ALPHA-ACTIVITY OF URANIUM AND THORIUM DECAY CHAINS
AT EQUILIBRIUM

<u>Quantity</u>	<u>Isotopes</u>	<u>Alpha-Activity</u>
1 g of U-natural	^{238}U	3.33×10^{-7} Ci
(in secular equil-	^{234}U	"
ibrium with all	^{230}Th	"
daughters)	^{226}Ra	"
	^{220}Rn	"
	^{218}Po	"
	^{214}Po	"
	^{210}Po	"
	Total	2.7×10^{-6} Ci

1 ppm U = 1 $\mu\text{g/g}$ = 2.7 pCi/g alpha activity

1 g of Th-natural	^{232}Th	1.09×10^{-7} Ci
(in secular equili-	^{228}Th	"
brium with all	^{224}Ra	"
daughters)	^{220}Rn	"
	^{216}Po	"
	^{212}Bi	0.38×10^{-7}
	^{212}Po	0.72×10^{-7}
	Total	6.5×10^{-7}

1 ppm Th = 1 $\mu\text{g/g}$ = 0.65 pCi/g alpha activity

through the soil, the more soluble long-lived daughters may be leached from the site of formation and deposited elsewhere. The presence of the daughter of ^{226}Ra , ^{222}Rn , a noble gas with a half-life of about 3.8 days that is long enough to permit migration away from its point of formation, means that under conditions where the gas can diffuse away from the parent, this decay product and its daughters will be in quantities lower than equilibrium. The loss of 50% of these daughters for the top meter of the soil has been given as a typical value.³ In deeper ore bodies, particularly those of a large size, such movement should be less significant. If one considers the 50% to be typical, then 1 ppm of uranium would have an activity of 2 pCi/g of alpha activity rather than the 2.7 pCi/g at equilibrium. For the thorium chain, the half-life of the radon daughter is small enough that a similar phenomenon is not expected.

III. DISTRIBUTION OF URANIUM AND THORIUM

Uranium and thorium are naturally most abundant in granitic, intrusive rock and volcanic rock. Uranium and thorium concentrations in the U.S. continental crust range from 0.2-7.0 ppm for uranium and 0.2 to 36 ppm for thorium. The averages weighted by areal abundance are 2.5 ppm (5-7 pCi/g) for uranium and 10 ppm (6.5 pCi/g) for thorium.⁴ But in some areas the concentrations are considerably higher. For the 7 000 square-mile Colorado Front Range the averages are 4.6 ppm (9.2-12 pCi/g) uranium and 22.7 ppm (15 pCi/g) thorium, with some relatively small areas having rock containing as much as 100 ppm (200-270 pCi/g) uranium, and 400 ppm (260 pCi/g) thorium.⁴ The Conway granite in New Hampshire has been known for some time to have elevated natural radioactivity. Values ranging from 4.3 to 25.5 ppm (12-69 pCi/g) uranium, and 30 to 77 ppm (20-50 pCi/g) thorium have been reported in some of these granites.⁵ Another source of elevated natural alpha-activity in the U.S.A. is the phosphorite rock and "black sand" of Florida. The phosphorite contains uranium ranging from 135-350 ppm (365-945 pCi/g) and the "black sand" contains thorium ranging from 250-1000 ppm (162-650 pCi/g).⁶

There are other regions of the world where even greater concentrations of natural alpha-activity are found, most notably in Brazil and India. Along the Atlantic coast of Brazil (Rio de Janeiro and Espirito Santo states) there are extensive deposits of monazite sand, as much as 300 miles along the shores of Espirito Santo. The monazite proper consists of 70 percent rare earths, 20 percent thorium oxide, and 0.15-0.25 percent uranium oxide. The monazite

concentration in layered patches of sand ranges from 5-20 percent, but even over a considerable extent of ordinary sand, it may reach 0.5 percent. At the high end of these ranges, the alpha-activity concentration approaches 8 nCi/g from thorium, and 14 nCi/g from uranium content. The total alpha-activity concentration is, therefore, on the order of 22 nCi/g in the more concentrated sands. Other regions in Brazil with remarkably high naturally radioactive soils and rock are Pocos de Caldas and the area around Araxá and Tapira.

At one site within Pocos de Caldas (the Morro do Ferro) there are extensive deposits of thorium ore. Values of three percent thorium are not uncommon (20 nCi/g).⁷ At Araxá and Tapira the radioactivity is associated with a magmatic rock called apatite (complex fluorophosphate of calcium) which is profusely mineralized with materials including pyrochlore. Pyrochlore has a high content of thorium, along with rare earth oxides, and only relatively minor amounts of uranium (thorium oxide, 7 percent, or 60 nCi/g; uranium oxide, 0.22 percent, or 6 nCi/g, for a total of about 66 nCi/g).⁷ Of course the pyrochlore amounts to only a small percentage of the total rock (about six percent or about 4 nCi/g), except in certain rich layers.

In the Kerala and Tamil Nadu States on the west coast of India there are extensive deposits of monazite sand that contain relatively high concentrations of thorium. Monazite is also found in soils in these regions in smaller amounts.

Kerala beach sand contains 5-9 percent monazite.⁸ Kerala monazite is approximately 10 percent thorium oxide and 0.4 percent uranium oxide.⁶ The alpha activity of such sand is then approximately 5 nCi/g thorium and 830 pCi/g uranium. Soil values are in the ranges of 0.4-0.5 pCi/g uranium and 7-25 pCi/g thorium.⁸

It appears then that the bulk of the naturally radioactive rock and soil around the world contains on the order of 10 pCi/g of alpha-activity from the uranium and thorium series. There are regions in Brazil and India where populations live in areas containing significant amounts of alpha emitters in the soil.

IV. EFFECTS OF HUMAN POPULATIONS

Some studies of possible effects on human populations living in areas of elevated background radiation from naturally radioactive materials in the soil have been done. From the perspective of the transuranic elements these have the

added feature of high external gamma radiation, up to two rems per year in the highest areas, and the high levels of radon isotopes in the atmosphere. Thus, any discernible effect will have been associated largely with these radiation sources and separating any effect from the alpha emitters other than ^{220}Rn will be difficult. The following kinds of information are available.

1. A very extensive demographic and gamma radiation dosimetric survey of a portion of the population in the monazite sands region of Kerala State, India, has been reported.⁹ On the basis of the TLD dosimetric measurements it is expected that, as a first approximation, of the 70 000 individuals living in the region covered by the survey, about 16 600 are likely to be receiving a dose exceeding 500 mr/yr, and several hundred might be receiving exposures greater than 2 r/yr (57 out of 8513 individuals surveyed out of this population received a dose greater than 2 r/yr.)⁹ Data on pregnancies and their termination were extensively reviewed to determine if such vital statistics reflect the elevated exposure. The number of pregnancies per couple (Fertility Index) was not different between groups receiving widely different levels of background radiation (the categories for this and other statistics are 100-500 mr/yr, 600-1000 mr/yr, 1100-2000 mr/yr, and >2000 mr/yr).

No clear-cut patterns emerged from a study of the sex-ratio among the off-spring. Infant mortality was high among all groups studied. Infant mortality is a sensitive indicator of the living standards of a community, and primitive medical facilities, inadequate sanitation, and the like are characteristic of the region. Infant mortality rates as high as those found in this region were reported in other parts of India having normal background. However, it was noted that the highest mortality rate (309/1000 live births) was recorded in the group of nine couples living in Thekkunghagum receiving an exposure from gamma radiation alone greater than 2 r/yr. The frequency of abortion abnormalities and multiple births was not significantly different among the different categories. However, the frequency of still births in the group receiving greater than 2 r/yr was significantly higher than in other groups (6.14% vs 3.6% in the lowest category of exposure). The conclusion of the study was that although differences were recorded between the highest and lowest exposure groups, the parameters analyzed were not sensitive enough to reveal statistically significant differences.

2. A number of studies of the population in the Brazilian areas of high natural radioactivity have been made.¹⁰ One physiological manifestation attributed to the radiation environment of Guarapari, Brazil, is an elevated rate of chromosome aberration in the peripheral lymphocytes. The lack of reliable public health records, or routine medical examination and the generally hard conditions of life endemic to these people have not provided a base for a statistical study of macroscopic somatic damage. A cytogenetic survey, in which the chromosomes of the radiosensitive, peripheral, blood lymphocytes are microscopically analyzed for aberration rates, revealed a type and frequency which cannot be attributed solely to an external radiation source, and which suggests the presence of an additional exposure to a high LET radiation.¹⁰ The source is thought to be a temporary body burden of thoron and its daughters inhaled while inside Guarapari buildings. Although direct evidence concerning the source has not been compiled, a dosimetric model has been developed utilizing a small population of workers in a monazite-sand processing mill in a Rio de Janeiro gas mantle (ThO_2) factory.¹⁰ This model confirms the cytogenetic response to transient elevated levels of thoron and daughters.

3. Another study of a selected population group among the inhabitants of the monazite belt in Kerala, India, focused on certain dermatoglyphic parameters (finger ridge counts) thought to be sensitive indicators of genetic change in man.¹¹ Total finger ridge count is one of the more thoroughly studied quantitative characters in man. The familial correlations for this trait are in remarkable agreement with the theoretical values for perfectly additive genes.¹¹ In the case of a quantitative trait, an increase in mutation followed by recombination might be expected to add to its variance, and radiation exposures with high acute doses have confirmed such an expectation. However, when dermatoglyphic data from 74 males native to the Kerala area were compared with those from 72 ethnically comparable males native to another area exposed to near normal levels of radiation, all six quantitative characters studied showed lower variance among the higher exposed population compared to the controls. Two possible interpretations for this somewhat puzzling finding have been proposed: a) the exposed population may have originated from a small group of possibly related individuals, or b) the effects of chronic low-level radiation may not be the same as those of high, acute doses. The former may sometimes decrease rather than increase variance of quantitative traits. The authors suggest that

it is conceivable that chronic low-level radiation has a much greater phenotypic rather than genetic effect on development, and that a generally poor environment may reduce variance, as has been observed with respect to variance of I.Q., in individuals living in poor environments.¹¹

4. A cytogenetic investigation of the population exposed to the elevated radiation environment of Badgastein, Austria, was initiated in 1968. The major source of exposure is the radon and daughters in the thermal baths (^{222}Rn content of the water is $\sim 15\ 000\ \text{pCi/L}$, in the air of the baths, $\sim 90\ \text{pCi/L}$).² The calculated alpha dose rates to various organs (principally lungs and blood) range from $0.07 - 0.15\ \text{rem/yr}$ (lungs) in inhabitants of the surrounding area, to $50 - 100\ \text{rems/yr}$ (lungs) in thermal gallery miners and trainleaders.² The upturn point in the cytogenetic effect vs alpha blood dose curves lies somewhere between 0.3 and $1\ \text{rem/yr}$. An investigation has begun on the age and cause of death of the people living in the area of Badgastein and control populations in non-radioactive spas with similar geographical and ecological conditions. To date the findings are that the longevity of the Badgastein population is certainly not below the control places, and the cancer mortality is certainly not higher.²

A very large number of studies of possible effects on human populations from exposure to natural radioactivity at different levels and under different conditions have been made, only a few of which have been reviewed here. The evidence of well over ten years of study is inconclusive. On the one hand, certain effects that could be attributed to exposure to ionizing radiation have been observed,^{9, 11} but on the other hand, the search for severe adverse health effects (life-shortening, cancer) has not been at all conclusive.^{9, 10} The reasons for the inconclusiveness regarding effects of elevated natural radiation include such problems as,

1. The population size under study is not large enough to exhibit epidemiological manifestations;¹⁰
2. An adequate control population cannot be found;⁹
3. A physiological effect which has been observed,^{2, 9} chromosome aberrations, not only presents the difficulty of establishing an adequate control since several other environmental agents can cause chromosomal breaks, but also it is extremely difficult to

- make an estimate of the degree of damage associated with such aberrations since they are actively selected against;¹² and
- 4) The results of many animal studies¹³ indicate that as the levels of exposure to internal radiation from such sources as ²²⁶Ra and ²³⁹Pu are successively reduced to very low levels, the survival curve (percent survival vs time) more closely approximates the natural survival curve that results from aging processes. Hence, below some level the effects of internal irradiation may not contribute significantly to the natural consequences of a host of aging effects.

To conclude: human populations are known to have lived for thousands of years in regions of the world where surface deposits of natural alpha activity on the order of several nanocuries per gram occur without effects that are either so severe or different in kind from natural ones that these regions were set aside as not fit for habitation or growing food.

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APPENDIX L

TECHNICAL DEVELOPMENTS IN WASTE MANAGEMENT

During the development of this report, the Task Group identified a number of suggestions that could lead to improved control of wastes containing transuranium elements. Since the improvement of technology was not a purpose of this study, the suggestions were not pursued further. However, they are described briefly in this appendix for possible future study by those involved in waste management.

1. While not a technical point, the need to resolve the question of the proper approach to protecting possible future intruders became apparent. As was discussed in the body of the report, the Task Group could not reach agreement as to whether such actions should be included in the pathways. The Task Group also noted that they were not constituted to grapple with this question. Instead, it was believed that the question involved legal, ethical and social expertise not represented on the Task Group.

We feel that it would be useful to have a properly constituted group study this question with a view to providing a policy to be used by technical people.

2. While some success has been had in minimizing waste volumes, we believe that further efforts are needed to reduce not only volume but the quantity of contained transuranium elements. This could involve studies of the processes used to reduce scrap and make the scrap and waste forms more tractable for management or for recovery. At present, the difference between waste and scrap is defined by the cost of recovery. Perhaps processes more amenable to recovery at a lower cost could result in smaller quantities of the transuranium elements going to waste. We think immediately of the acid-digestion possibility for combustibles which may result in a stream easier to handle than incinerator ash for recovery, but there may be other methods. Our philosophy is that every gram that can be recycled to the main stream is a gram that does not go to waste.

We are not certain of the responsibility for funding such an effort, since it could be considered as process improvement, but we believe that it is vital that waste management considerations extend to every step of any process.

3. Another item for consideration arose in our discussion of the use of plastics for containing wastes. Our studies indicate that such containment in a non-degradable plastic would be useful if deep burial is attempted, but the practice can be undesirable in shallow burial because of the tendency to preserve the combustibles. This, of course, only applies if intrusion is considered a viable pathway.

If shallow burial continues, we believe that some effort should be devoted to obtaining degradable plastics that will provide the same protection to waste handlers as the present plastic.

4. The possibility of using additives to the waste at the time of emplacement to control possible migration could be investigated. These additives could change chemical form or the conditions in the waste mass. The migration and reconcentration of various metals from ore bodies in nature could provide clues to the type of additive that would be most effective.

5. Another possibility for controlling migration would be to line the burial pit with a layer of absorbent material that would remove the transuranics from the water passing through. This could possibly be most effective if used in conjunction with an additive that would aid in controlling the chemical form and pH of the transuranics in the water.

6. The use of a rock covering for the burial ground to control erosion should be considered. This would also discourage burrowing animals and, possibly, minimize transfer to the surface by plant uptake. In an article by Judson¹ there is a picture of a Roman road built of basalt blocks in 200 B.C.. The road is now a low ridge because the less resistant lithified volcanic ash of the hillside upon which the road was built has been eroded away. Such erosion control would need serious investigation to find the types of situations for which it is useful and the sizes and shapes of rocks that would provide the best long-term stability. Obviously if the rock is too small it will, itself, erode away. However, if it is too large it may serve as a source of future building material and be removed.

7. In connection with the above, it was suggested the burial ground be marked with permanent markers in a universal language so that future generations could replace the covering as needed.

8. Criteria for acceptable areas for shallow earth burial are needed to permit decisions on continuing in present areas and in looking for possible future areas. We have concluded, elsewhere in this report, that burial in impermeable formations, particularly if fractured, should be avoided in areas of moderate to heavy rainfall because of the tendency for the pits to fill with water and overflow or possibly provide seepage paths through the fractures. Data available indicate lower leaching if the water from rainfall passes through the waste to the aquifer without accumulation in the pit. Such data should be studied and definite site criteria established to allow avoidance of unfavorable sites.

9. The present approach to radioactive waste disposal appears to call for an either-or approach. Either the waste goes to shallow earth burial or it goes to a repository at a significant cost. As has been seen, a major portion of the problem with shallow earth burial arises from the possibility of direct exposure of people to the waste mass following uncovering by erosion or other means or from intrusion into the wastes. This raises the possibility of providing a third alternative for those wastes which are too high in contamination for the relatively exposed shallow earth burial but are low enough to be placed in an areas less secure than the repository. Dry caverns, abandoned mines, or deliberately dug caverns in dry formations were suggested as possibilities. The lower limits for artifacts that are proposed in this report also suggest that such a location could also be used for items such as these if the area were properly secured by collapsing any openings.

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