CONF-7810208

313
2-21-80

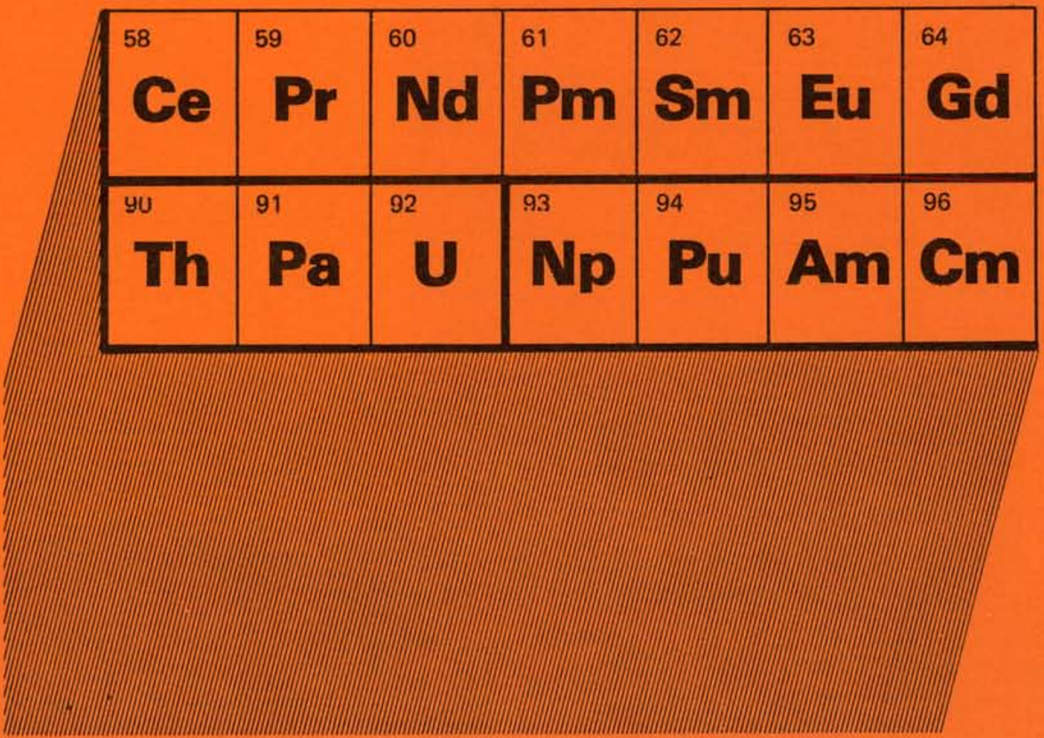
DR. 712

Workshop On Environmental Research For Actinide Elements

Proceedings Of The Workshop
October 10-12, 1978
Airlie House
Airlie, Virginia

MASTER

Department of Energy
Office of Health and Environmental Research



58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd
90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm

DISCLAIMER

CONF-7810208
UC-70

Workshop On Environmental Research For Actinide Elements

Proceedings Of The Workshop
October 10-12, 1978
Airlie House
Airlie, Virginia

Department of Energy
Office of Health and Environmental Research
Washington, DC 20545

Edited by: Robert L. Watters

58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd
90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Available from:

National Technical Information Service (NTIS)
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161

Price:	Printed Copy:	$ 5.25
	Microfiche:	$ 3.00

TABLE OF CONTENTS

INTRODUCTION

This workshop was the fourth in which the Department of Energy and its predecessors, the Energy Research and Development Administration and the Atomic Energy Commission have brought together the scientists who have worked on environmental research related to the transuranic elements and other actinides. Representatives from the regulatory agencies have also been invited to participate in these workshops. The objectives have been to provide a forum for the discussion of the latest research results and develop guidance for future research direction.

The topics of discussion in this fourth workshop centered on the adequacy of plutonium information, the needs and direction of future transuranium research and the research necessary for an appraisal of the potential hazards associated with thorium and high specific activity uranium isotopes. These topics were discussed in four panels which have prepared reports that make up the body of these proceedings. The four panels and their chairmen were:

Ecosystem Research	T. E. Hakonson Los Alamos Scientific Laboratory
Plant/Soils Research	C. E. Francis Oak Ridge National Laboratory
Marine and Freshwater Research	A. B. Joseph Office of Health and Environmental Research, Department of Energy
Statistics and Modeling	R. O. Gilbert Battelle Pacific Northwest Laboratory

In addition to the appraisal and planning studies, a report was given on an intercomparison of analytical quality control. This exercise was conducted by the Environmental Measurements Laboratory (EML) of the Department of Energy under the direction of H. Volchok and was a voluntary activity for the research laboratories involved in environmental research with the transuranic elements. The EML report with additional responses since the workshop presentation is included at the end of the proceedings.

The OHER staff wishes to thank all of the participants for their cooperation and thoughtful contributions. Special thanks are extended to the panel chairmen for the time and effort which they expended in planning, conducting and reporting the panel discussions.

ECOSYSTEMS RESEARCH PANEL

The Problem

Environmental and health issues concerning the transuranic elements stem from the potential for mobilization and transport of these elements within terrestrial ecosystems. The schematic in Fig. 1 illustrates the concept of environmental plutonium distribution and transport and indicates potential transport pathways through a simplified ecosystem under the assumption that the soils compartment is the source of the plutonium.

Chemical, physical, and biological processes control movement of plutonium within ecosystems. Physical and biological transport processes are illustrated by the arrows on the left and right side of Fig. 1 and represent processes that are driven by wind, water, biological, and mechanical activity. For example, wind, water, or mechanical (i.e., tilling soil) resuspension of soil may result in contamination of plant and animal surfaces with redepositing soil plutonium. The diet of consumer organisms may also contain this surficially deposited plutonium. Examples of biological transport include the movement of plutonium associated with a grazing animal and the subsequent redistribution of this material through defecation and/or death as well as burrowing and grooming activities that result in contamination of the animal.

Chemical transport is shown by the passage of soluble plutonium from soil through plant roots and across physiological membranes (i.e., lungs, gut) of consumer organisms. Leaching of soluble plutonium through the soil column is another form of chemical transport.

The ability to predict plutonium behavior in the environment requires information on the following topics:

1. The ecological relationships in contaminated ecosystems including the content and size of the compartments and the exchange of materials between compartments.

2. The pathways and mechanisms of plutonium transport through the ecosystem and the relative importance of these pathways as sources of contamination to receptor compartments, including man.

3. The rates of transport of plutonium between ecosystem compartments.

4. Differences in plutonium transport as a function of source characteristics, regional ecology, and time.

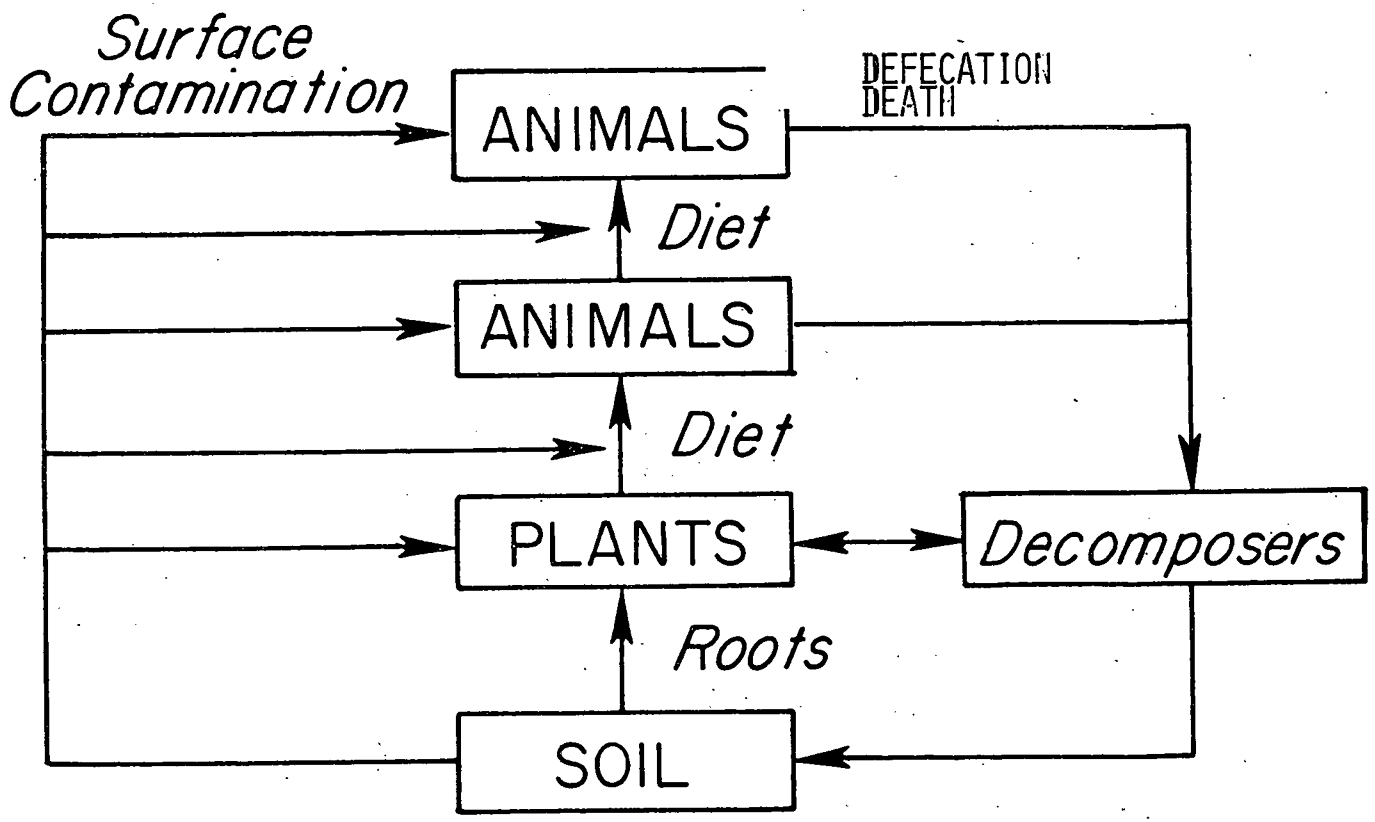


FIG. 1 CONCEPTUAL MODEL OF TRU TRANSPORT IN TERRESTRIAL ECOSYSTEMS

In order to address these topics, we are highly dependent on data obtained from direct sampling of existing contaminated sites, site contamination histories, plutonium sources and ecological relationships in the various study locales. This information, in combination with the chemical data obtained from laboratory studies and from a limited number of field studies on plutonium transport, provided the basis for the discussion in the Ecosystems Research Panel.

Panel Objectives

The broad directive given for this workshop was to integrate current knowledge on actinides in the environment and attempt to formulate useful generalizations. In keeping with this mission, the Ecosystems Research Panel set the following as specific objectives:

1. Identify commonalities and provide generalities about the distribution and transport of transuranic elements in ecosystems.
2. Evaluate the potential for changes in concentrations of transuranic elements in ecosystem components over contemporary and extended time scales.
3. Determine whether transuranic elements become concentrated as they are transported through food webs.
4. Identify information needs, particularly for other actinides.
5. Identify potential environmental problems associated with nonproliferative fuel cycles.

To address these issues, the panel posed a series of questions that were formulated to draw out discussion on general transuranic element dynamics in ecosystems. The format for the panel's discussion was to pose the question, discuss applicable information and then attempt to formulate a general statement with appropriate caveats summarizing our answer.

Discussion Topics and Panel Response

The following list of topics was addressed by the panel. The consensus response to the ensuing discussion is given:

1. Generalizations about distribution of transuranic elements in terrestrial and aquatic ecosystems as a function of time.

   (a) Where are transuranic elements in presently contaminated ecosystems?

Response: Soil and sediments contain at least 99 percent of the ecosystem inventory of Pu although a few percent less than this amount may appear in soil and sediment under chronic input conditions due to interception of airborne particles by plant surfaces. However, after cessation of chronic input, virtually all of the Pu will be transferred to the soil which serves as an effective sink for transuranic elements in ecosystems.

Although little is known about transuranic elements other than Pu, it is generally believed that these elements will be distributed very nearly like Pu. There is at least one major exception to this generalization. In the Arctic only about 30 percent of the ecosystem inventory of fallout Pu is in soil with lichens containing most of the remainder. However, there is a continual loss of Pu from lichens to soil so that even in this case, soils will be the ultimate repository for Pu. While small, the amount in green plants can vary over several orders of magnitude (10^{-5} to 10^{-3} of the amount in soil). Attached material, e.g., resuspended particles, can amount to several percent. In freshwater aquatic situations greater than 99 percent of the Pu will be found in the sediments. This is true for Pu translocated from soil by surface runoff and Pu entering through industrial process effluents.

   (b) Are there major differences in the distribution of transuranic elements as a result of site and transuranic element source factors?

Response: Site/source factors are comparatively unimportant for approximate estimates of distribution in terrestrial ecosystems including ponds. However, the small mobile fraction in air or on plants may be significantly affected by source characteristics such as particle size, chemical species and physical-chemical form.

(c) What changes in transuranic element distribution will occur with time?

Response: For a one-time deposition in soil, dilution will occur in the long term as a consequence of Pu migration downwards, e.g., of the order of tenths of a centimeter (or less) per year. Short term redistribution will occur as a consequence of Pu bound to soil particles being translocated by rainfall/runoff processes. The rate and direction are highly dependent on water movement and soil type. Activity of soil biota, plants and animals, can be expected to redistribute Pu in both directions. The net effect nevertheless will be a reduced surface concentration available for resuspension to air and surface biota. In many locations, the time frame for this phenomenon is not well established.

(d) Can we relate the results of studies on the environmental distribution of transuranic elements to the potential for human exposure?

Response: Current predictive models indicate that redistribution can alter the relative importance of pathways of human exposure, e.g., for the LMFBR estimates, a plant/soil concentration ratio of less than 10^{-3} indicated a predominant radiation dose from the inhalational route of entry whereas for concentration ratios greater than 10^{-3} the predominant radiation dose was from food chain transfer.

(e) Are further field studies needed on the distribution of transuranic elements? (Minimum and optimum efforts.)

Response: At contaminated sites, Pu distribution in soil and plants is fairly well characterized. Considering surface contamination of foliage (in field situations) the field data agree reasonably with experimental data from the laboratory for plant uptake. Some ecosystems such as Arctic tundra and tropical rain forest, could be better characterized but may not be important from the standpoint of nuclear development. There is greater need to look at Cm and Am distributions in ecosystems because of their increasing concentration with time relative to Pu in radiation waste management. In the context of long lived waste materials there is a need to examine the distribution of Np because of the greater tendency for mobility and lower soil sorption. There is a need to study applicability of present data to the new nuclear technologies, e.g., alternative cycles (uranium and thorium isotopes).

2. Generalizations about transport in terrestrial ecosystems.

(a) What is the relative importance of chemical, physical and biological processes in mobilizing transuranic elements in the environment?

Response: Physical processes predominate in transporting transuranics in the environment when bulk movement is considered. While only small fractions of the total inventory are involved in chemical and biochemical transport their significance is great because biological availability is related to solubility (chemical mobilization) and incorporation into biological tissues normally requires membrane transport (biochemical mobilization).

(1) Importance of physical processes: can be seen in the resuspension experiments at Hanford (1), the Nevada Test Site (2-6), and Savannah River (7); and the transport by water erosion in studies of gulley erosion at Los Alamos (8-9), large and small midwestern watersheds in studies at Argonne (10-11), and the Savannah River (12).

(2) Importance of chemical-biochemical processes can be seen in studies at Oak Ridge on root uptake (13) and studies at Battelle on gastrointestinal absorption of biologically incorporated plutonium (14).

Fewer data are available for Am and Cm than for Pu and virtually no field data for Np exist.

(b) Concentrations of Pu do not increase in higher trophic levels of ecosystems. There is no biomagnification. Transport of "soluble" forms across membranes leads to Pu concentrations of the receptor which are 10^3 to 10^4 less than the concentration of the donor organism.

Response: Relative to a donor component, the Pu concentration of the receptor is always less, i.e., the ratio of receptor/donor is less than 1.

Inventory distributions of Pu follow the order:

soils & sediment > vegetation > primary consumer > secondary consumer

which also reflects trophic transfer processes, the inverse of biomagnification.

Supporting data sets for these relationships are available for:

Grassland ecosystems--Rocky Flats
Forest ecosystems--Oak Ridge
Canyon ecosystems--Los Alamos
Old Field ecosystems--Savannah River
Desert ecosystems--NTS

It is important to be certain about donors to a receptor organism. Several special cases are:

Receptor	Donor
Native/domestic animals	soil ingestion
Aquatic animals	sediment

Biomagnification is generally defined in terms of concentration increases in successive trophic levels expressed in terms of the quantity of the element present per unit of mass. This definition does not permit a comparison of plutonium concentration in plant mass in relation to plutonium concentration in soil solution.

Although passage across biological membranes represents a small fraction of Pu movement in the ecosystems, this small quantity can be very important because it can accumulate in critical organs. Time dependent increases in Pu concentration can occur in selected tissues, but life-times of organisms do not allow biomagnification.

(c) Can we predict transuranic element behavior in ecosystems?

Response: The short-term behavior of plutonium can be predicted to a limited extent in a few well-studied ecosystems.

Models have been published which describe the transport of plutonium in terrestrial ecosystems at Oak Ridge National Laboratory, Nevada Test Site, and Savannah River. Conceptual and computational aspects of these models are applicable to other terrestrial systems, but the parameter values applicable to most other systems have not been well developed. The parameter values for the ORNL and NTS models are derived from experiments or field data. Some attempts at sensitivity analysis have been made.

There is not sufficient information to extend existing models to extended time periods (beyond 100 years). However, transuranic element analogues such as Th and Nd can be used for such projections.

There is not sufficient information to incorporate time-variable rate parameters or stochastic processes into existing models. Hence error estimates on predictions are not readily available.

Although Pu transport models are under development for ecosystems at Rocky Flats and Savannah River, parameter values for many other ecosystems are not available.

Parameter values for transuranic elements other than plutonium are generally lacking.

(d) Are further field studies required on transuranic element transport?

<u>Response</u>: Transport processes are reasonably well understood, but applications for hazard assessments are generally imprecise. Therefore, selective field studies are needed if we are to predict better the transport of transuranic elements in ecosystems. Time series data, which reveal much about transport pathways and rates, are generally lacking.

Experimental studies on interception and retention of transuranic elements in various physical-chemical forms by vegetation are badly needed since there are few data and since these are critical processes. Existing models need to be validated in a variety of ecosystems and under varied conditions.

More emphasis needs to be placed on actinides other than plutonium. Agricultural ecosystems have received little attention in the ecosystem context. Also, they are of primary concern with respect to human intake. Therefore, they deserve greater attention. Relative to other environmental stressors, the transuranics may be a non-problem.

3. Additional Research Needs

U/Th Fuel Cycle

The use of Th-232 to produce fissile U-233 adds higher specific activity U isotopes (U-232,-233,-234) to the biosphere inventory of man-made radioactivity than the U-235/Pu-239 cycles. The primary radiation hazard in terms of environmental transport to man should accrue from the U-232 decay series. Radiation doses to bone from deposited U-232 (and daughters) will be about four times greater than for an equal Pu activity. Transuranic elements will also be produced by the fuel mixtures proposed, albeit in different proportions.

Potential Problems

1. Mining/milling and the consequences of accumulating the Th-232 decay series; i.e., the Ra-228/Th-228 problem.
2. Recycle and inevitable release of high-specific activity U isotopes to local environments.

3. Waste disposal.

4. Mixed Th/U-235 and Th/Pu-239 fuels will present similar transuranic element problems as Pu breeders. Proportion will differ.

Unique Environmental Issues

1. Compared to transuranic elements, U will be more mobile in ecosystems, including human food chains.

2. U-232 and U-233 decay chains have short-lived daughters compared to Pu, therefore, increased relative lifetime dose commitments are possible.

References

1. Sehmel, G. A. 1978. Transuranic and tracer simulant resuspension. Pacific Northwest Laboratories, BNWL-SA-6236, 75 pp.

2. Anspaugh, L. R., P. L. Phelps, N. C. Kennedy, J. H. Shinn, and J. M. Reichman. 1974. Experimental studies on the resuspension of plutonium from the aged sources at the Nevada Test Site. In Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants. Report CONF-74092. pp. 727-743.

3. Anspaugh, L. R., P. L. Phelps, N. C. Kennedy, H. G. Booth, R. W. Goluba, J. M. Reichman, and J. S. Koval. 1974a. Resuspension element status report. In The Dynamics of Plutonium in Desert Environments. P. B. Dunaway and M. G. White (eds.). Report NVO-142 pp. 221-297.

4. Anspaugh, L. R., J. H. Shinn, and D. W. Wilson. 1974b. Evaluation of the resuspension pathway toward protective guidelines for soil contamination with radioactivity. In Population Dose Evaluation and Standards for Man and His Environment. IAEA Vienna. pp. 513-524.

5. Anspaugh, L. R., J. H. Shinn, and P. L. Phelps. 1974c. Resuspension and redistribution of plutonium in soils. In Proc. Second Annual Life Sciences Symp., Plutonium--Health Implications for Man. May 22-24, 1974. Los Alamos, New Mexico.

6. Anspaugh, L. R., J. H. Shinn, and P. L. Phelps. 1975. Resuspension and distribution of plutonium in soils. Report UCRL-76419.

7. Milham, R. C., J. F. Shubert, J. R. Watts, A. L. Boni, and J. C. Corey. 1976. Measured plutonium resuspension and resulting dose from agricultural operations on an old field at Savannah River Plant in Southeastern United States of America. In Proc. Symp. Transuranium Nuclides in the Environment. IAEA-199-SM-99, pp. 409-420.

8. Nyhan, J. W., F. R. Miera, Jr., and R. J. Peters. 1976. Distribution of plutonium in soil particle size fractions in liquid effluent-receiving areas at Los Alamos. J. Environ. Qual. 5:50-56.

9. Hakonson, T. E., J. W. Nyhan, and W. D. Purtymun. 1976. Accumulation and transport of soil plutonium in liquid waste disposal areas at Los Alamos. In Transuranium Nuclides in the Environment. IAEA/ERDA Symp., IAEA-SM-199/99, Vienna, pp. 175-189.

References Cont.

10. Sprugel, D. G. and G. E. Bartelt. 1978. Erosional removal of fallout plutonium from a large midwestern watershed. J. Environ. Qual. 7(2):175-177.

11. Muller, R. N., D. G. Sprugel, and B. Kohn. 1978. Erosional transport and deposition of plutonium and cesium in two small midwestern watersheds. J. Environ. Qual. 7(2):171-174.

12. Hayes, D. W. and J. H. Horton, Jr., Plutonium and americium behavior in the Savannah River marine environment. In Transuranic Elements in the Environment. TID 22800, USDOE.

13. Dahlman, R. C., E. A. Bondietti, and L. D. Eyman. 1976. Biological pathways and chemical behavior of plutonium and other actinides in the environment. In Actinides in the Environment, ACS symposium series No. 35, pp. 47-80.

14. Sullivan, M. F. and T. R. Garland. 1977. Gastrointestinal absorption of alfalfa-bound plutonium 238 by rats and guinea pigs. In Pacific Northwest Laboratory Annual Report for 1976 BNWL-2100 NTIS.

Ecosystems Research Panel

E. Bondietti
L. Cadwell
R. C. Dahlman
T. E. Hakonson (Chairman)
W. C. Hanson
J. C. Laul
K. McLeod
J. K. Miettinen
N. Singh
J. R. Trabalka
B. E. Vaughan
H. E. Walburg
R. L. Watters
M. Werkema
F. W. Whicker
R. Wildung

SUMMARY OF THE SOIL/PLANT PANEL

Introduction:

This report is not intended to summarize any individual or group research effort that was presented to the panel, but is confined to the overall conclusions and recommendation on direction of further research needs on actinides in the environment.

I. Plutonium in Soil and Its Availability to Plants:

The panel agreed that individual soil properties such as pH, organic matter content, cation exchange complex, redox potential, and texture, all influence sorption of plutonium on soil and the subsequent availability to plants. However, the interaction between soil properties makes it difficult to evaluate independently the influence of each. Two exceptions noted were pH and clay content. The majority of the panel agreed that increasing soil pH results in decreased plant uptake, either as a result of greater sorption to soil particle surfaces, or as a result of complex formation; increasing clay content had a similar effect. It was the opinion of the panel that there is little need to investigate further the effect of soil properties on sorption of plutonium on soils and its uptake by plants. A notable exception was the lack of understanding of the influence of Eh* on sorption of Pu to soil. The panel felt that Pu sorption decreases as Eh is increased; however, this depends on the stability of insoluble hydrolyzed Pu(IV) oxidation state and that of the more soluble species Pu(V) and Pu(VI) in soil. On the other hand, the panel agreed that Eh would have little influence on plant uptake of plutonium because of the reducing influence of plant roots, i.e., Pu(V) and Pu(VI) will be reduced to the more unavailable Pu(IV) by metabolically active roots.

Phosphate minerals were thought to decrease plutonium sorption to soil and decrease plant uptake; however, no specific data were presented to support the conclusions.

---

*Redox potential, the potential of a half-cell measured against the standard hydrogen half-cell.

The members of the panel felt that complexation of plutonium by soluble organic ligands (those derived naturally from soil organic matter or from soil microorganisms and especially added as synthetic chelates) strongly influenced and was possibly a controlling factor in the availability of plutonium to plants (Appendix A). Sources of organic ligands include compounds derived from soil organic matter by natural processes, soil microorganisms and their metabolic products, and especially synthetic chelating agents which are sometimes used in agriculture and often found in nuclear wastes. Low molecular weight organic ligands appear to compete more successfully in the forming of coordinate bonds with plutonium than do the inorganic ligands commonly found in soil solution, such as OH^- (from hydrolysis of water), HCO_3^-, and CO_3^{-2} depending on pH and CO_2 partial pressure. To date the evidence for such a mechanism is indirect; however, there is some direct evidence of plutonium complexation with microbial metabolites and dispersed soluble soil organic matter. At the same time, it is appropriate to point out that the significance of the natural organic-Pu-complex in plant uptake has not been domonstrated with respect to its impact on dose to man via the ingestion pathway. Continued cropping of plutonium contaminated soil in the greenhouse has shown significant increases of plutonium uptake by plants in the first few years, implying that plutonium in the roots of previous croppings is more plant available than soluble inorganic plutonium added to the soil. However, members of the panel noted that this enhanced plutonium uptake leveled off after the first few years. The panel concluded that future plutonium plant uptake studies be directed at plutonium source term evaluation.

Other conclusions of the panel regarding plutonium-soil-plant interactions were:

- The dominant form of plutonium in soils is a polymeric form.
- K_d's* ranged from 10 to 10^4.
- Concentration ratios (CR's)+ ranged from 10^{-3} to 10^{-5} for whole plants and 10^{-5} to 10^{-7} for the grain of plants.
- The outstanding characteristics noted about plutonium in soils were its low solubility, multiple oxidation states, ability to form polymers and ability to complex with inorganic and short chain organic ligands.

---

*Concentration radionuclide per gram of solid/concentration radionuclide per ml of solution.

+Concentration radionuclide per gram dry weight of plant tissues/concentration radionuclide per gram dry weight of soil.

The panel felt that plutonium in soil was a non-problem with regard to radiation dose to man via plant uptake route over the short-term (less than 100 years); however, no upper level of soil plutonium concentration was considered.

It was the unanimous opinion of the panel that the range in CR's introduced a major uncertainty in plutonium dose to man via soil-plant food chains; however, there was concern among some of the members that dose assessment scientists might use CR's that are inappropriate for certain soils. Members realized the difficulty of characterizing soils with ranges in CR's in view of the inability to agree on the effects of individual soil properties on behavior of plutonium in soil. At the same time, members of the panel felt that extremes in soil pH and texture could be considered key factors with regard to anticipated CR's. The following is only a guide and should be used only as a "rule of thumb" where specific site data are not available.

Suggested CR's for Pu in Soils of Varying pH and Clay Content

	CR's		Soil Properties	
	Grain	Whole Plant	pH	Clay Content (%)
Maximum:	10^{-3}	10^{-2}	<5.5	<10
	10^{-4}	10^{-3}	>5.5	<10
Minimum:	10^{-5}	10^{-4}	>5.5	>10

Panel members also felt that the above CR's are strictly those in which foliar contamination with plutonium contaminated soil is not included. Such contamination can easily overshadow that amount of plutonium which is taken up via the root pathway. Observed field concentration ratios are known to vary inversely with rainfall. This is considered to be due to the tendency for rainfall to remove foliar contamination.

II. Soil Sampling Procedures for CR's:

The concept of concentration ratios has been developed and uniformly defined to characterize the transfer of a radionuclide from the soil to the plant via plant uptake after a condition near steady-state has been reached. Obviously this ratio only has significance if the concentration of the radionuclide in the soil is that for soil with which the plant roots come in contact or soil in close proximity to the plant roots. There is no reason to expect a relationship between Pu concentration in soil outside the root zone and Pu uptake. Use of CR's are more appropriate to agricultural systems where the soils are mixed to a certain soil depth (e.g., the plow layer of 0-20 cm). Thus, CR's determined in the greenhouse with homogeneous soil-plutonium mixtures may not be appropriate for certain natural ecosystems where the contamination is in the top 0-3 cm, or at specific waste management sites where radioactive contaminated soils have been buried by uncontaminated soils. The panel felt that the preferred method for soil sampling for CR's under such conditions would be sampling the soil in the feeding root zone of the plant for which the CR is desired.

III. Pathways for Foliar Uptake of Actinides:

Wind and mechanical resuspension* is known to occur from surficial contaminated soils. However, the resuspension rates required for predicting downward airborne concentrations cannot be confidently predicted (experimental values range up to 13 orders of magnitude). Data presented to the panel by George Sehmel (Appendix B) questioned the validity of earlier data which showed that airborne plutonium concentrations decreased with a half-life of approximately 40 days. Other investigators have also observed that such an approximation of 40 day half-life may be in error; in fact, some could not detect significant differences in airborne concentrations over time. Since George Sehmel was the only scientist present who has dealt extensively in the airborne measurements of resuspended particulate matter, the panel felt that they could not make any conclusion or recommendations regarding his observations. His views are expressed in Appendix B. In the same vein, T. Tamura emphasized the problem that there are no data available on the influence of soil aggregate stability, or methods developed to determine soil aggregate stability which will subsequently influence resuspension.

---

*Resuspension refers to that material in soil which becomes airborne. It was originally used with regard to radioactive fallout from weapons testing. It is now frequently used to describe the process of material becoming airborne whether or not the contaminant was originally deposited from the air.

D. Cataldo presented evidence that plutonium assimilated following deposition on plant leaves is more mobile in the plant than plutonium entering via root uptake. The general consensus of the panel was that it was indeed an interesting observation, but they were indecisive regarding its importance in the movement of plutonium to man via the plant-man foodchain. Regardless, in arid climates, foliar uptake or contamination appear to be just as important as the root uptake route in estimating dose-to-man via foodchains. The magnitude of the foliar route is enhanced in arid climates relative to humid climates because of the greater tendency for wind resuspension and generally higher soil pH values than for soils of the humid climates.

IV. Conclusions Regarding the Use of K_d:

The distribution coefficient (K_d) is a conventional method used to designate the partitioning of a chemical species between the solid phase and the liquid phase of a suspension. Theoretical considerations limit its use to concentrations where the measurement characterizes the exchange reaction and there are no other competing reactions of significance, e.g., precipitation. When trace concentrations are used, this is generally not a problem, i.e., the mass of the radionuclide being studied is small compared to the total sorption capacity of the matrix. Its inception in soils and sediment work was with relative short half-life radionuclides (namely Sr and Cs), and activity per se was used to express or imply mass concentration. G. Beall illustrated how mass effects can produce erroneous conclusions when radionuclides with a long half-life are used. The following is an example: suppose 250 ml of a solution containing 2500 dpm of Np-237/ml (approximately 1 nCi/ml or 1.6×10^{-6} g Np/ml) was used to determine the K_d of 0.1 g sample of soil having a CEC of 10 meq/100 g. In this case, sufficient Np-237 is present to saturate approximately 20 percent of the exchange sites. Some preliminary data presented by G. Beall also indicated that the solubility constant for Am-carbonates may be exceeded at pH values greater than 7; tentative calculations suggest that 10^{-41} is the K_{sp} for $Am_2(CO_3)_3$. In some cases reported in the literature precipitation rather than sorption may account for the observed results. Thus, when using long-lived radioisotopes, the concentration of the radionuclide in solution and on the solid phase should be given, preferably in mass/volume or mass/mass units. Additional information, such as, pH, ionic strength, and the concentration of the counter ions, should be reported. If this is done, it will give investigators an opportunity to compare results under the various conditions that affect K_d measurements and thus provide confidence that the K_d determined does characterize an exchange reaction.

V. Conclusions Regarding Plutonium Availability to Plants Over the Long Term (In Excess of 1000 Years):

As a result of the extensive soil-plant studies conducted at various locations within the USA, the plant availability of plutonium can be estimated, although the mechanisms for its availability are not clearly defined.

If synthetic chelating agents are present, they will control the chemistry, mobility and plant availability of plutonium in soil, i.e., the soil characteristics become insignificant. As mentioned above, they are frequently present in nuclear waste and are used in some agricultural areas. The effect of their presence on plutonium availability over a very long time, e.g., >1000 years, is unknown.

Studies to date have provided the necessary information to approximate the transport of plutonium to man via the ingestion pathway. It is anticipated that this information will be valid for at least 100 years. We anticipate that these short term studies are likely to be a fair approximation of the long-term availability to plants, and plutonium availability will either remain constant or possibly decrease. These conclusions are based on the knowledge of availability of stable Th from plant uptake studies, the physical and chemical behavior of mineral soil properties, and knowledge of soil weathering processes.

VI. Future Research Needs on Actinides in Soil/Plant Systems:

The increased emphasis on alternate nuclear fuel cycles such as the thorium-uranium (Th/^{233}U) fuel cycle has created interest in the movement of ^{232}U and its daughters in the environment. Interest has been developed because of the high specific activity of ^{232}U; thus, the hazard to man will be more from radiological effects rather than chemical effects, as in the case for the long-lived uranium isotopes (^{238}U and ^{235}U). The panel agreed that CR's for uranium were 10 to maybe 100 times more than those for plutonium, and that the near future research efforts should be directed at clarifying the range of CR's for uranium. It was anticipated by the panel that the CR's for uranium will vary as a function of soil property more than that observed for plutonium. Although considerable is known about the geochemistry of uranium and thorium, there is relatively little known about the ecosystem dynamics of natural uranium and thorium. The environmental chemistries of uranium, thorium, and plutonium in the IV oxidation state are quite similar. However, the most stable oxidation state of uranium in the environment exposed to the atmosphere is U(VI), which is significantly more soluble than Th(IV) or Pu(IV). Its greater solubility and its complexation with carbonates makes movement and uptake by plants in alkaline soils significantly greater than thorium and plutonium.

Additional research should be directed at determining the influence of the oxidation state of Np(IV and V) sorption on soil and uptake in plants. Soil-plant research should also be continued with respect to Am and Cm. Greater emphasis should be placed on research methods to predict the long-term behavior of the actinides (particularly plutonium) in soils.

APPENDIX A

Organic Matter Complexation of Actinides

The importance of hydrolysis in governing Pu behavior, including the soluble fraction, has been demonstrated. However, the small fraction available for plant uptake from the soil solution must be truly in solution as an ionic species for some finite period of time.. Several possible mechanisms exist for providing a source of the Pu ions available for plant root uptake, including ion pairs of Pu(V)-Pu(VI) and HCO_3^{-1}, $-CO_3^{-2}$, or complexes of Pu(IV) with organic ligands. Organic ligands form the most stable complexes with Pu(IV), but both low molecular weight ligands and HCO_3^{-1} and CO_3^{-2} levels are directly related to microbial activity and the decomposition of organic materials. There is direct evidence of Pu complexation with microbial metabolites and considerable presumptive evidence supporting a microbial influence on Pu solubility in soil.

The importance of defining the mechanisms responsible for maintaining a pool of biologically available Pu is critical for the prediction of the behavior of Pu and other actinides in the soil-plant systems over the long term and in environments not specifically studied.

Dominic A. Cataldo
Pacific Northwest Laboratory

APPENDIX B

Resuspension of Actinides from Soil

Wind and mechanical resuspension is known to occur from surficial contaminated areas. However, neither the resuspension rates required for predicting downwind airborne concentrations, nor the airborne concentration at the contaminated site, can be confidently predicted. The uncertainties or range of experimental values range up to 13 orders of magnitude. Continuing research is needed to reduce this uncertainty to a tolerable level in order to develop predictive transport models which are validated.

We know resuspension is occurring from controlled sites, but we suspect resuspension and atmospheric transport is also occurring from off-site low-level-surficial contamination caused by prior deposition. The relative extent and quantity of this off-site transport needs to be established, the rate of spreading, and its source determined by fingerprinting by isotopic Pu analysis.

We now know Pu is being transported in the air on both respirable and non-respirable host particles. Both particle diameters are suspect of being either an immediate or a subsequent respirable concern. When large host soil particles impact on a surface, many smaller soil particles are released. It is reasonable to assume that Pu particles attached to the host soil particle could also be released. Thus, the rate of surficial contamination spread needs to be determined.

There is little data for linking airborne Pu concentrations (dpm/g) to surficial concentration (dpm/g). The range of $(\mathrm{dpm/g})_{\mathrm{airborne}}/(\mathrm{dpm/g})_{\mathrm{surface}}$ is not unity, but can range seven orders of magnitude from 10^{-4} to 10^{3}. This uncertainty is caused in part by collecting airborne material which was transported from upwind (rather than at the surface below the air sampler) the non-homogeneous variation of the surface contamination, as well as too few surface sample results. Data are needed to link airborne concentrations to surface properties including - contamination level, distribution on the soil particles, size distribution, ground cover, as well as wind stress and mechanical stress resuspension variables.

Data are needed to resolve and/or determine the change in resuspension airborne concentration with time. Early data indicated airborne concentrations at NTS decreased with a half-life of ~40 days. Thus, credit was taken in dose assessment calculations for reduced airborne concentrations for succeeding time. However, airborne data were subsequently collected at the same NTS site by a different investigator. Subsequently, the early and latter airborne concentrations were compared by a third investigator. This comparison showed that airborne concentrations were essentially the same (within one order of magnitude) for both time periods. Thus, credit cannot be taken for reduced resuspension with time. Consequently, several resuspension experiments under different controlled conditions are still needed to resolve the change in resuspension, and hence, airborne concentrations with time.

George Sehmel
Pacific Northwest Laboratory

APPENDIX C

Panel Members

C. W. Francis, Chairman
Oak Ridge National Laboratory

D. C. Adriano, Secretary
Savannah River Ecology Laboratory

G. W. Beall
Oak Ridge National Laboratory

J. C. Corey
Savannah River Laboratory

D. A. Cataldo
Pacific Northwest Laboratory

R. Franklin
Department of Energy

T. R. Garland
Pacific Northwest Laboratory

S. Y. Lee
Oak Ridge National Laboratory

S. G. Metcalf
Rockwell Handford Operations

E. H. Essington
Los Alamos Scientific Laboratory

H. Nishita
University of California, Los Angeles

E. M. Romney
University of California, Los Angeles

R. G. Schreckhise
Pacific Northwest Laboratory

G. A. Sehmel
Pacific Northwest Laboratory

J. C. Wheppard
Washington State University

T. Tamura
Oak Ridge National Laboratory

A. Wallace
University of California, Los Angeles

W. Weimer
Pacific Northwest Laboratory

D. Wester
Argonne National Laboratory

MARINE AND FRESHWATER RESEARCH PANEL

INTRODUCTION

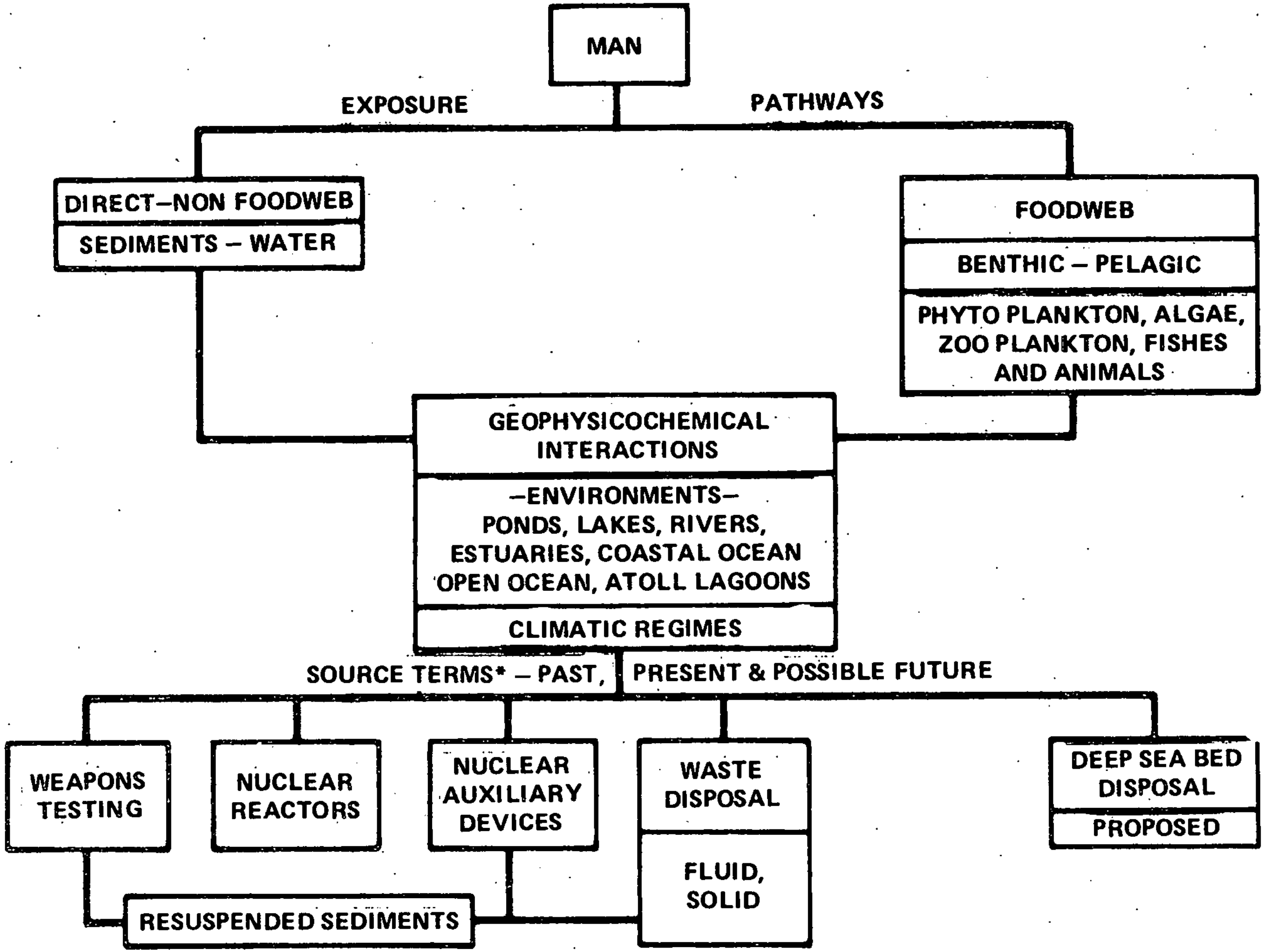
Figure 1 presents an abbreviated schematic outline of the concerns of the Panel. As the figure indicates, our ultimate concern is to be able to predict the exposure of man. The diagram hints of the complexity of source terms, environments and pathways--both food web and non-food web--that comprise the research problems of predicting the exposure of man. It should be kept in mind that this research area is superimposed on the natural behavior of these environments and biota, which also is incompletely understood.

Most of the transuranic elements, Pu, Am and Cm, in the surface waters are from nuclear weapons testing rather than the direct discharge from nuclear industries. The principal one from nuclear weapons tests is Pu. About 120 KCi of Pu are now deposited on the land and about 210 KCi are estimated to be in the oceans. Americium and curium inventories from this source are small. A buildup of ^{241}Am will occur due to decay of ^{241}Pu; however, at most it will represent about 30 percent of the Pu activity.

Nuclear industries usually discharge directly to the environment, e.g., Windscale (U.K.), Trombay (India), and the Miami River (Ohio). In some cases the Am activity exceeds the Pu activity (Windscale). Other sources are nuclear weapons tests on ocean islands and plane accidents involving nuclear devices. Most studies in surface waters have been based on Pu from fallout.

Plutonium in the open ocean is derived from atmospheric deposition whereas in smaller water systems such as rivers and streams the mobilization of the transuranic elements from watersheds may be important. In coastal waters and large lakes, a combination of the two processes is important in the budget of transuranic elements. Most observations are based on fallout, therefore, we are observing the system's average response to fairly uniform distributions of Pu. The use of many of these observations to site specific problems should be done with caution. However, one important point is that in all aquatic systems the sediment or sedimentary particles are the primary repository for the transuranics.

At present, transuranic element concentrations measured in the open aquatic environment do not pose a hazard to man and in fact, are insignificant when compared to the natural radiation environment. The open aquatic environment is taken to mean that which is accessible to members of the public.



*RADIOACTIVE ACTINIDES OF CONCERN – $^{237, 239}$Np, $^{238, 239, 240}$Pu, ^{241}Am, $^{242, 244}$Cm, $^{232, 233, 234, 235, 238}$U AND DAUGHTERS, AND ^{232}Th AND DAUGHTERS.

FIGURE 1. – SCHEMATIC OF CONCERNS OF THE MARINE & FRESHWATER RESEARCH PANEL.

DESCRIPTION OF RESEARCH RESULTS AND NEEDS

Transport Processes

Several considerations guided the deliberations of this group charged with identifying priority research needs concerned with describing and predicting the behavior of the movement of actinides about the earth's surface.

First of all, there is recognition that we should be dealing with uranium and thorium isotopes as well as the transuranics inasmuch as the former may well be involved in future energy systems.

Secondly, problems are usually site specific. Although it may be possible to generalize about the behavior of actinides during their movement about the earth's crust, the unique situation will usually prevail. Thus, priorities should be given to areas where nuclear fuel is processed.

Finally, the group was uncomfortable about certain transport problems when expertise was unavailable. For example, is air transport of particles from high level storage areas well researched? Are there leaks of actinides to the atmosphere at nuclear fuel facilities?

The following schematic diagram, Figure 2, is proposed as a common basis for discussion of physical transport processes moving actinides in different aqueous environments. Plutonium is used in this example, however, similar diagrams could be drawn for each actinide and there would certainly be a large degree of commonality.

The four equilibria identified are all amenable to study both in the laboratory and in the field. Equilibrium 1 roughly defines the basic chemical forms of the plutonium (actinide element) present in solution. The relative importance of equilibria 2 and 3 depends upon equilibrium 1, and the important chemical parameters which affect this equilibrium will, therefore, need to be identified and measured. The relative magnitudes of the distribution coefficients in equilibria 2 and 3 will in general differ and one or the other will dominate.

Any understanding of transport processes of the actinides between the water in which they are soluble and the solids on which they are sorbed requires firstly, knowledge of the equilibria that will ultimately be established if no changes in the system occur, and secondly knowledge of the rates at which these equilibria are approached. Information

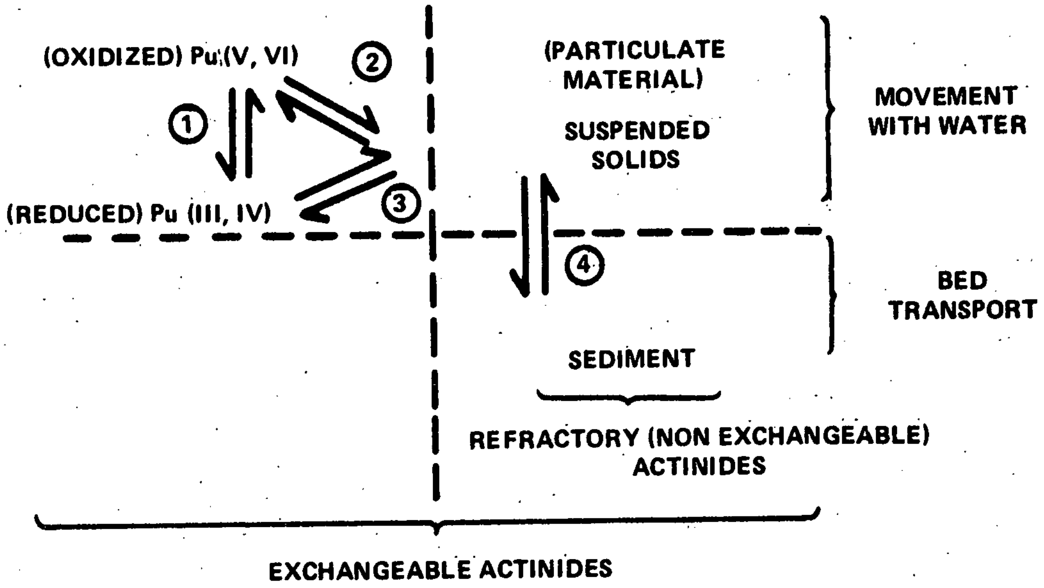


Figure 2. Schematic Diagram of Physical Transport Processes in Aquatic Environments

about the equilibria includes the association constants between each of the ions and those ligands which strongly complex the ions, the association constants of the ligands with the hydronium ion, the redox potentials of actinides with multiple oxidation states, and the distribution ratios of the actinide and/or actinide-ligand ions between the aqueous media and the solids which strongly sorb them. Of these, the association constants of the ligands with the hydronium ion are the only ones for which reliable values may exist. For the association constants of the ions with ligands, present knowledge strongly suggests that complexing is important but there is little or no information about the actual values of the constants. For the distribution ratios of the actinide and/or actinide-ligand ions between the aqueous media and solids, there are approximate (factor of 10) values for the distribution ratios in certain natural systems and strong indications of what the principal adsorbant solids may be, but no information as to which of these solids dominates. (It should be stressed that the principal adsorbant for one actinide is not the same as that for another. This applies even to different oxidation states of the same element, i.e., Pu(IV) and Pu(VI) may be adsorbed onto different solid phases.)

The oxidation-reduction regime is particularly important as it controls the form of elements such as uranium and plutonium. There are no reliable values for the redox potentials of the actinide complexes in near neutral media. Furthermore, this lack extends to the important natural constituents such as iron and manganese that are multivalent in the environment. The chemical behavior of such actinides as plutonium and uranium in oxic systems such as the Great Lakes and the open ocean can be predicted with assurance to be vastly different from those that exist in anoxic systems such as organic-rich sediments.

There is a need for both *in vitro* and *in vivo* investigations and combinations of both. In the absence of such studies, it will not be possible to develop models to predict the behavior of the actinides in natural systems.

Rate measurements are urgently needed. There is little or no information about the rates at which the multivalent actinides equilibrate with atmospheric oxygen in the oxic systems nor the rates at which the oxidized forms are reduced when the system goes from oxic to anoxic. Although it has been generally assumed that the Pu ion-ligand reactions (association and disassociation) are rapid, there is no real evidence that this is true at the very low concentrations involved. There is uncertainty about a similar situation for the rates at which the actinide and/or actinide-ligand ions adsorb and desorb from solids.

Ground Water

The transport of actinides by underground water has received insufficient attention. The problems that should be studied include the potential methods of injection of the actinides into various types of aquifers, the effect of the dissolved and solid phases in the water on the chemical nature of the injected actinides, and the effect of intervening geological formations during the transport of the contaminated water on its way to the aquifer.

Rivers

Rivers receive their transuranic input primarily from mobilization of transuranic elements off watersheds. Estimates of watershed Pu loss rates to rivers range from 0.05 percent/year to 0.005 percent of the estimated Pu inventory in the watershed. The higher value is for a heavily cultivated watershed in Ohio versus the heavily forested Savannah River watershed which is reservoir regulated. These loss rates result in Pu concentrations of ∿.1 to ∿2 fCi/l in the rivers. Similar estimates of Am and Cm watershed loss rates are not available because of sparsity of river concentration data and because the watershed inventories of these nuclides have not been made. However, based on ratios of Pu/Am concentrations one would expect Am removal values to be similar to Pu. On-going research on other types of river systems will provide additional watershed loss rate values. Literature on Th and U concentrations are available for rivers; however, a review of the literature to determine its usefulness for determining Th and U transport processes has not been made.

Transport of actinide elements by river systems is a subject of major concern in assessment of the potential hazards to man resulting from the handling of reactor wastes.

The behavior of actinides in rivers may be strongly influenced by the chemistry prevailing in the specific drainage basins. For example, rivers draining arid lands are typically lower in both suspended and dissolved organic carbon and higher in dissolved carbonate ion than rivers in more humid climates. Rivers draining swamp areas contain extremely high loads of humic acids and other dissolved organic compounds. Differences have been observed in the behavior of trace metals in rivers as a function of these parameters. There is little information concerning the distribution of actinide elements between the dissolved and suspended load of sediments and also concerning the influence of ligands, sediment mineralogy, and sediment chemistry on the distribution. The lack of information includes the transuranium elements in general, but also extends to uranium and thorium and their daughter products in rivers.

Both field and laboratory research are required to make possible predictive models for the behavior of actinide elements in these systems. Studies concerning adsorption properties of actinides and also U-Th series nuclides on a variety of surfaces need to be supplemented with observations in nature of the parameters which control the behavior of these elements. The contamination of the earth's surface with fallout nuclides and releases from nuclear facilities provide adequate amounts of these elements for studies in natural systems. It may be fruitful to pursue studies of chemical analogs in natural systems, especially U and Th as analogs for Pu^{+6} and Pu^{+4}. U and Th are additionally important because of the probable future emphasis on U-Th fuel cycles rather than breeder reactors.

The chemical interactions of actinides in rivers will control the form in which these elements are introduced into estuaries. The understanding of the chemistry of the input source is necessary to a description of this next step in transport of materials to the ocean.

Estuaries

Estuaries receive water containing transuranic elements from terrestrial sources as well as the ocean. At this interface between the oceans and rivers, many interactions occur. The principal interactions are the deposition of suspended sediment from terrestrial sources by salt water flocculation and the ensuing sorption-desorption reactions. Sediment deposition patterns vary from estuary to estuary, and are subject to changing with storms and river flow. Dredging to maintain shipping channels is an important translocator of sediment in many estuaries. The dredged material is sometimes used as landfill, with the filled areas used as farmland or for other activities.

Plutonium concentrations in estuarine waters are about the same as for rivers, ∿.1 to 3 fCi/l, the higher concentrations are usually associated with higher suspended sediment loads. Probably half of the riverine concentration is deposited in the estuary and the other half is transported to the ocean. However, the transuranic input from the ocean has not been properly evaluated.

Plutonium concentrations in estuarine sediments range from <1 fCi/gm to 200 fCi/gm (dry wt) which is in the same range as for other aquatic environments. From Pu data it is obvious that deposition patterns are nonuniform and most sediments show a decrease in Pu concentration downward. Many times no significant difference in Pu concentrations were found in sediments from the headwaters to the mouth of the estuary.

Large estuaries can support a substantial fishery (i.e., Chesapeake Bay); however, Pu concentrations in fish from estuaries are about the same as for rivers and coastal areas (<10 fCi/gm dry wt) which results in doses ∿0.001 percent of the dose from natural radioactivity. However, estuaries are sites of major shell fisheries and shellfish normally have concentration ratios (concentration in organisms/concentration in seawater) for transuranic elements that can be as high as 1000. Due to lower consumption of shellfish and still relatively low Pu concentration (∿10 fCi/gm dry wt) the radiation dose to the human population is still insignificant when compared to the natural radiation dose.

A variety of estuarine areas, characterized by their physics, biology and chemistry, are under investigation but these have not yet produced the amount of information required for generalizations about actinide transport.

Cycling of Actinides in Aquatic Ecosystems

Observations have been made in the last decade of actinide behavior in freshwater and marine aquatic environments around the world. The locations studied include areas which have received inputs of transuranic elements from stratospheric fallout alone--such as the Great Lakes, coastal zones, estuaries and the open oceans and the areas where nuclear weapons debris fell locally such as Bikini and Enewetok Atolls. Coastal areas including the English Channel, Irish Sea, and Trombay, India, and rivers such as the Miami in Ohio have had releases from fuel fabrication or reprocessing plants. Results of these studies were examined in terms of four general prevalent concepts:

1. Transuranic elements are insoluble and immobile in the aquatic environment;
2. transuranic elements in marine systems behave differently than in freshwater systems;
3. the behavior of the transuranic elements in the environment is dependent upon source term; and
4. transuranic elements do not increase in concentration with higher trophic level.

Geochemical Interactions

The experimental data for freshwater systems so far indicate that two of the general hypotheses in relation to the transuranic elements, viz. (1) that they are insoluble and immobile in the aquatic environment, and (2) that their behavior is dependent upon source term are not confirmed. While it may be argued that any element which seeks the sediments as strongly as plutonium is insoluble, when the solubility can vary by two orders of magnitude the element cannot be considered to be completely insoluble. However, there are complex interrelationships between the tendency for complexation and the various oxidation states.

We could find no evidence to suggest that differing sources have any effect on the chemical properties of the transuranic elements in aquatic systems when moderately well dispersed in the system. The results indicate that the transuranic elements are soluble to some extent in both freshwater and marine systems and therefore available for direct active physiological transfer across biological membranes. From the evidence available plutonium appears to behave chemically and biologically alike in the oceans and the Great Lakes. Since these systems are oligotrophic and their chemical properties are largely controlled by their respective carbonate cycles, the similarity in the values of K_D and the ratio of Pu(V,VI)/Pu(IV) should be expected. However, the behavior in the oceans may not be as simple as it would appear.

Nelson reported that, while all the plutonium remaining in solution immediately after discharge from the Windscale Plant will pass through an ultrafilter and is predominantly Pu(VI), samples collected in the North Sea which contain a far lower concentration of plutonium have a higher fraction of their content as Pu(IV).

In alkaline, oligotrophic lake systems soluble plutonium appears to be in equilibrium in solution with the major fraction present as Pu(VI) or Pu(V) (probably as a carbonate complex), but Pu(IV) is primarily taken up by particulate material. In other lakes a majority of the soluble plutonium appears to be present as Pu(III) or (IV).

These studies show the danger of attempting to extrapolate the results of laboratory experiments carried out with moderately concentrated solutions ($\geq 10^{-6}$M) to predict the behavior of plutonium in the environment, where the maximum concentration observed has not exceeded $\sim 10^{-13}$M. It must be recognized, however, that somewhere within the concentration range 10^{-13} -10^{-6}M, plutonium ceases to exhibit the properties of simple ions and the formation of polymeric species cannot be ignored.

Biological Accumulation

The major result common to all biological studies to date is that the transuranic elements do not increase in concentration with higher trophic level in aquatic organisms, thus bearing up the previously held concept. While bioaccumulation of plutonium and americium occurs in all organisms, biomagnification, in general, does not appear to occur at higher trophic levels. However, at present we do not know what fraction of the element in the environment is biologically available to man relative to a calculated concentration factor in aquatic organisms.

It is clear that we do not yet understand the complexities of the transfer of the transuranic elements through aquatic food chains. However, there is no evidence so far to suggest that these elements behave like mercury in the food chain, in that high concentrations are not found in edible fish species. The experiments so far confirm the difficulties in understanding of the biological cycling of these elements in aquatic systems. The variability in the values of the concentration ratios reflects not only biological variation but also variations in the concentration or chemical form of plutonium or americium in the water column, particularly for free-moving species.

In terms of transfer to humans, it is clear that the transuranic elements are not significantly enriched in the fresh edible fish. However, little is known about the mechanisms of transfer through the food chain.

Marine geochemical cycles of the naturally-occurring actinides and their daughters are well known, owing in part to their usefulness for studies of ocean dynamics. Tetravalent and trivalent forms such as those of thorium or actinium are least soluble and their transport tends to be primarily with particulate matter. Biological uptake of tetravalent and trivalent actinides is also, therefore, relatively slight compared to other members of the series. On the other hand, hexavalent uranium and divalent radium are more soluble, more mobile and of greater biological availability in the marine environment. Radium-226 and ^{228}Ra both are released into seawater from sediments. The former appears to be returned to sediments in zones of high biological productivity.

It is well established that ^{210}Po, a member of the ^{238}U series below the highly mobile inert gas ^{222}Rn, is a major contributor to the natural radiation dose for some marine organisms. In the ^{232}Th series it appears that ^{228}Ra is the most mobile member but relatively little is known of its ecological dynamics. It will be important to determine the transfer routes and coefficients whereby ^{228}Ra and its ^{228}Th daughter may move from coastal waters to man. By coincidence, the natural introduction of relatively soluble ^{228}Ra into coastal water and its subsequent decay to ^{228}Th mimics the potential inadvertent release of relatively soluble ^{232}U (a by-product of the thorium fuel cycle) and its decay to ^{228}Th. The series of short-lived, alpha-emitting daughters of ^{228}Th make its specific biological dose rate among the greatest of all radionuclides. It is therefore important to determine its fate in marine systems both for the estimation of natural dose rates and, also, for assessment of potential hazard from by-products of the thorium fuel cycle.

Sediment Uptake and Redistribution

Experimental studies in the Great Lakes, Buzzards Bay, Irish Sea, and Trombay Harbor have shown that in these comparatively shallow bodies of water greater than 95 percent of the total plutonium introduced as a result of fallout and discharges from fuel reprocessing plants is rapidly transferred to sediments.

Where the rate of sedimentation is comparatively rapid, the sediments provide a record of not only the history of deposition but also evidence of redistribution of the sediments due to physical, chemical and biological processes.

Interface Processes Section

While several interfaces of biogeochemical importance may be defined for the transuranic elements in natural waters, the interface of major importance is that between man and the environment. In recent years, a considerable effort has gone into the study of the transport of plutonium across this interface. The results of this effort have afforded a general understanding of the possible impact of plutonium on man. However, similar knowledge is lacking for the other transuranium elements, i.e., Np, Am, Cm, as well as uranium and thorium and their respective radiologic daughters. This information will be important in proposed nuclear fuel cycles.

It has been stressed that on a long time scale (hundreds to thousands of years) the oceans will be an extremely important component of the biogeochemical cycle of actinide elements. Hence, a quantitative understanding of the basic physical constants of the oceanic system must be forthcoming, including deep water circulation rates, thermal coefficients of deep sea oozes and plasticity of sediments. However, in considering the interface to man, shallow water systems are far more important than the deep ocean. In fact, on a short time scale (days to hundreds of years) freshwater and estuarine systems will be of prime importance because of their proximity to man's activities.

The problems of source term, chemical form, storage and transport in various compartments of the interfaces to man (freshwater, estuaries, groundwater and atmosphere) have been addressed by the other discussion groups. The general conclusion in every case is that the available information is, by necessity, site specific. In addition, the data which do exist are primarily for plutonium and are barely adequate. Information on americium, uranium, thorium and their radiologic daughters is limited and certainly not adequate, while information on neptunium and curium is essentially non existent.

The final compartment in the interface of environment with man that was discussed by this group was diet. It is generally accepted that there is no biomagnification of plutonium in large aquatic food webs that eventually transfer to man via the ingestion of piscivorous fish. The data base for the other actinides, especially in marine systems, does not allow us to make the same statement for these elements. However, little is known, even for plutonium, about transfer of material to man via shortened food webs where individuals directly consume organisms from the initial trophic level which are known concentrators of plutonium. Examples of this behavior may be found in any of a number of cultures which regularly eat seaweeds or seaweed products (e.g., algin). Finally, what is the relative importance in these food webs of dissolved vs. particulate forms of all the actinides on accumulation by organisms and relative availability to man?

It was the conclusion of this discussion group that very little information exists on the chemical forms, transport, and availability to man of the actinide elements and uranium and thorium daughters in freshwaters, groundwaters and estuaries that would allow the construction of a predictive model of these elements in the environment. Of particular concern, is the paucity of data for elements of the uranium and thorium series in food webs of fresh and nearshore surface waters and the chemical changes that these elements may undergo in groundwater environments.

Transcending the entire discussion of the environmental interface with man is the primary observation of the site specific or regionality of the problems that we are asked to address. It is difficult to imagine that so few investigators, facing the limited number of environments that each is capable of studying, will arrive at solutions on a global scale. Emphasis should be placed on achieving predictability within regions having similar problems. Whether those regions be defined by locale, climate or social customs is not important. What is important is that the practical boundaries and magnitude of the interfaces with man be quantitatively defined within a region. These conditions have barely been met for plutonium and, for the most part, have not been addressed for the other elements under discussion.

Research Needs

Programmatic Problem Areas Releated to the Nuclear Alternatives

1. Evaluation of parameters related to the release of radioactive wastes in the oceans, particularly in regard to sea-bed emplacement.

2. The evaluation of the critical parameters related to the calculation of dose to man--concentration factors and gut transfer for:

   a. the transuranic elements,
   b. alpha-active products from alternative fuel cycles, and
   c. natural alpha-emitters.

3. The need to develop comprehensive conceptual and then predictive models for the chemical speciation, biological uptake and transport of these radionuclides in all watersheds coupled to the oceans which will interface with the models used by ICRP to predict dose to man.

Research Topics of Critical Importance Related to These Problem Areas Which Have Not Been Adequately Explored

1. Characterization of source terms in relation to labile and nonlabile forms of transuranic elements. Are there any sources at present in the environment which are strictly nonlabile, i.e., like high-fired oxides?

2. The determination of chemical form (speciation) in water bodies likely to contribute significantly to the dose to man:

    a. oxidation state

    b. complex formation with major inorganic ligands and with characterized natural organic ligands (see c below)

    c. characterize organic complexing agents responsible for preferential solubility of Pu(IV) in certain freshwater systems

    d. as affected by microbiological reactions

3. The determination of concentration ratios--is the uptake by organisms dependent upon chemical form and/or environment? Concentrate on species and environments leading directly to man. Validate utility of the use of trophic transfer factors.

4. Make a careful evaluation of distribution coefficients (K_D's) with sediments: Is equilibrium really attained?

    a. Are K_D's dependent upon sediment type, water chemistry (complexing, etc.), mineralogy, and surface adsorption.

    b. Do precipitates of the transuranic elements occur in the environment, i.e., is the solubility product concept valid?

    c. What happens in passage from fresh to saline conditions?

5. Determine the transfer of radioactivity from terrestrial to aquatic environments and thus to the oceans due to wind and water erosion. Integrate the transuranic element data in relation to our general knowledge of the transport of sediments in all coastal environments, from land, to rivers, to oceans.

6. Determine the loss of plutonium and americium from temporary sinks (0-100 years) and transfer to permanent sinks in sediments.

a. What is the rate of transfer of sediments from the continental shelf to the deep oceans?

b. Evaluate the residence time of radionuclides in the surface layers of sediments with respect to sedimentation rate, bioturbation, catastrophic events in critical areas and the transfer to the biotic community.

7. Are there any possible mechanisms which can return radionuclides from the deep ocean to the continental shelf?

8. Continue to use radionuclides as tracers for physical, biological and chemical processes. The processes will be the same for all other energy-related pollutants whether organic or inorganic. The sinks will be the same. The major advantage in using for example Pu(IV) and Pu(VI) as tracers for Th(IV) and U(VI), respectively, is that the Pu is not in equilibrium and therefore we can measure the rate of change.

9. Investigate the effects of pollutants from other technologies on the (re)mobilization of transuranic elements in the environment. What will be the effect of acid rain for example on the concentration of plutonium in soft water lakes and reservoirs?

10. Invasion of actinides into groundwater.

11. Kinetics of exchange of introduced isotopes of Th and U with their natural isotopes--differentiation between surface adsorption of pollutants and incorporation into mineral lattices.

REFERENCES

1. Goldberg, E. D. and K. Bruland, 1974. In "The Sea" (E. D. Goldberg, Ed.) Vol. 5, pp. 451-489. Wiley-Interscience, New York.

2. International Atomic Energy Agency, 1976. "Effects of Ionizing Radiation on Aquatic Organisms and Ecosystems." International Atomic Energy Agency Technical Document 172. IAEA, Vienna.

3. Kaufman, A., R. M. Trier, W. S. Broecker and H. W. Feely, 1973. J. Geoph. Res. 78:8827-8848.

Airlie House Meeting
Actinides in the Environment
October 10-12, 1978
Marine & Freshwater Panels

Dr. James Alberts
Savannah River Ecology Laboratory
Drawer "E"
Aiken, SC 29801

Dr. Larry K. Benninger
Department of Geology
University of North Carolina
Chapel Hill, NC 27514

Dr. Vaughan T. Bowen
Department of Chemistry
Woods Hole Oceanographic Institution
Woods Hole, MA 02543

Dr. D. Glenn Boyer
Environmental Control Technology Division
Mail Stop E-201, EV-131
U.S. Department of Energy, GTN
Washington, DC 20545

Dr. Jerome Bucher
Materials and Molecular Research Division
Lawrence Berkeley Laboratory
Berkeley, CA 94720

Dr. John L. Burnett
Office of Energy Research
Mail Stop J-309, ER-14
U.S. Department of Energy, GTN
Washington, DC 20545

Professor Gregory Choppin
Department of Chemistry
Florida State University
Tallahassee, FL 32306

Dr. Norman H. Cutshall
Oak Ridge National Laboratory
P. O. Box X
Oak Ridge, TN 37830

Dr. David N. Edgington
Radiological & Environmental Physics Division
Argonne National Laboratory
Argonne, IL 60439

Dr. Dick Emery
Pacific Northwest Laboratory
Battelle Blvd.
P. O. Box 999
Richland, WA 99352

Dr. L. D. Eyman
Environmental Sciences Division
Oak Ridge National Laboratory
Building 2001
P. O. Box X
Oak Ridge, TN 37830

Dr. William O. Forster
International Atomic Energy Agency
Karntner Ring 11
P. O. Box 645
A-1011
Vienna, Austria

Dr. Richard O. Gilbert
Pacific Northwest Laboratory
Battelle Blvd.
P. O. Box 999
Richland, WA 99352

Dr. Edward D. Goldberg
Geological Research Division, A-020
Scripps Institute of Oceanography
University of California
La Jolla, CA 92093

Dr. Leo S. Gomez
Sandia Laboratories
P. O. Box 5800
Albuquerque, NM 87185

Dr. Peter J. Hanson
National Marine Fisheries Service
Beaufort Laboratory
Beaufort, NC 28516

Dr. Florence L. Harrison
University of California
Lawrence Livermore Laboratory
P. O. Box 808
Livermore, CA 94550

Dr. David Hayes
Savannah River Laboratory
Aiken, SC 29801

Dr. J. M. R. Hutchinson
Coordinator-Radioactivity Section
Center for Radiation Research
National Bureau of Standards
Washington, DC 20234

Dr. Arnold B. Joseph
Office of Environment
Mail Stop E-201, EV-33
U.S. Department of Energy, GTN
Washington, DC 20545

Dr. Robert Larsen
Radiological & Environmental Research
Argonne National Laboratory
Argonne, IL 60439

Dr. W. E. Martin
Battelle - Columbus Laboratories
505 King Avenue
Columbus, OH 43201

Dr. George Matlack
Los Alamos Scientific Laboratory
P. O. Box 1663
Los Alamos, NM 87545

Dr. Helen M. McCammon, Director
Ecological Research Division (EV-33)
Office of Environment
Mail Stop E-201
U.S. Department of Energy, GTN
Washington, DC 20545

Dr. Don Nelson
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439

Dr. G. D. O'Kelley
Chemistry Division
Oak Ridge National Laboratory
Oak Ridge, TN 37830

Professor Robert Pain
Department of Chemistry
University of New Mexico
Albuquerque, NM 87131

Dr. R. J. Pentreath
Ministry of Agriculture, Fisheries
and Food
Fisheries Radiobiological Laboratory
Hamilton Dock, Lowestoft
Suffolk, England

Professor William R. Schell
College of Fisheries
Laboratory of Radiation Biology
University of Washington
Seattle, WA 98195

Dr. Martha R. Scott
Department of Oceanography
Texas A&M University
College Station, TX 77843

Dr. H. J. Simpson
Department of Geology
Lamont-Doherty Geological
Osbservatory
Columbia University
Palisades, NY 10964

Dr. William Templeton
Pacific Northwest Laboratory
Battelle Blvd.
P. O. Box 999
Richland, WA 99352

Dr. Robert Trier
Lamont-Doherty Geological
Observatory
Columbia University
Palisades, NY 10964

Dr. Herbert Volchok
Environmental Measurements
Laboratory
U.S. Department of Energy
376 Hudson Street
New York, NY 10014

STATISTICS AND MODELING PANEL

Panel Objectives:

The original goals of the Statistics and Modeling Panel were to identify:

- The type and quality of environmental data needed to make generalizations concerning environmental transport of actinides to man.
- Statistical design and analysis needs.
- Modeling needs.

We began by attending other panel sessions to gather information, be available for discussions, and keep abreast of possible future directions of research. The remaining time we met together as a panel to discuss issues and prepare a rough draft of our final report. This report begins with the identification of perceived problem areas and a list of specific recommendations to help solve these problems. This is followed by a summary of discussions that occurred during our panel meetings. We conclude with some summary remarks.

Identification of problem areas:

At an early stage of the workshop we came to the general consensus that several problem areas in statistics and modeling exist:

- Modeling and statistics are not being utilized to their full advantage by researchers.
- A need exists for increased communication between modelers, statisticians, and researchers.
- The efforts of the modeler, statistician, and researcher are often uncoordinated.
- Modelers are frequently charged with providing a prediction for decision making purposes with inadequate data.

The net effect of these influences is to slow down both the rate at which research objectives will be achieved and the quality of the modeling exercise. The panel feels researchers need to have a greater awareness of the role that modelers and statisticians can plan in the conceptualization, design, execution, analysis, and report preparation phases of a research effort.

Ideally, interaction between the statistician, modeler, and researcher should follow this pattern: An integrated, tentative model for the process under study is developed in concert with the researcher, making full use of his prior knowledge and of any hypotheses he may have regarding processes. Close interaction at this conceptualization phase helps the researcher define his study objectives to a finer degree and should result in a more efficient study design. Effective communication exposes aspects of the intended research that had not been previously recognized. Preliminary analyses or computations are performed to identify sensitive factors and as a direction to aid research. Pilot studies are conducted to collect information required to design an efficient research effort. Once data have been obtained and statistically analyzed, they are used to evaluate the original tentative model, obtain a better understanding of the process, determine if significant parameters have been identified, perform model sensitivity analyses, identify model weaknesses, and attempt a validation of the model. These analyses form a basis for designing future studies which, in turn, result in additional data for input models.

In essence, we are describing the scientific method and its application to environmental studies. In reality, this ideal is seldom accomplished. The Statistics and Modeling Panel believes the process is not working optimally due, at least in part, to a lack of communication between modelers, statisticians, and researchers.

Recommendations:

In light of the above discussion, this panel makes the following general recommendations:

- OHER should consider steps it can take to increase communication between researchers to help ensure that research goals are met with increased efficiency.
- OHER should take a more active role in defining research objectives and coordinating research efforts between laboratories.

One approach to implementing these recommendations might consist of establishing a modeling-statistics-researcher group (or person) within OHER to help direct and coordinate research, and to make sure that the right questions are being asked and that studies are properly planned and executed to answer these questions.

This panel has also identified a number of other, more specific, modeling and statistical needs. Specific recommendations directed at increasing communication and improving the design and analysis of environmental actinide studies are:

1. Increased attention to precise definition of study objectives. Precise study objectives help ensure an easier transition to more efficient study designs and more powerful statistical analyses. A close matching of sampling methods with study objectives is essential. The formulation of objectives and methods requires that we ask ourselves the right kinds of questions. Two examples are: How will study results be used, and is the study designed to provide results compatible with objectives?

2. A closer working relationship between modelers, statisticians, and researchers. Modelers and statisticians should be involved in the design of studies to help ensure that appropriate data are obtained for modeling purposes, and to gain a better understanding of the quality of data that is possible to obtain under field conditions. The researcher needs to gain a better appreciation of the data requirements of the statistician and modeler, and to improve his understanding of their role at various stages of the study.

3. The greater use of pilot studies to obtain information for designing more efficient field studies. Questions concerning number and spacing of field samples, components of variation, and other design considerations cannot be answered with any assurance unless pertinent prior information are available. Even if only a few data or preliminary estimates of means and variances are available, this information can be very useful in the design of studies to maximize output for fixed cost.

4. Increased communication between researchers at different laboratories. Although we realize that differences in source terms and ecosystem structure can create problems in the comparison of data between sites, we believe that increased communication could promote (a) an understanding of the similarities and differences among modeling approaches, (b) studies with similar objectives to allow evaluations of differences in distribution and transport processes at different sites, and (c) the development of comparable sampling, analytical, and statistical techniques.

5. Design of replicated field studies and experiments. Efforts should be made to develop and implement experimental field study designs replicated at different locations, climates, and times. The design of such studies should be a joint effort by a research team familiar with the characteristics of each study site across the country.

6. Greater awareness of dangers in the inappropriate use of estimators such as ratios and arithmetic means. Concentration ratios and arithmetic means are examples of estimators whose values are easily changed by outlying data points. Estimates of ratios are in general highly variable and are often statistically biased when few data are available. The frequency distribution of ratios is usually skewed, which complicates the statistical analysis. In addition, there are several different ways of estimating an average ratio, where each may give a different result. Finally, all too often, we fail to evaluate whether the numerator (y) and denominator (x) are related by $y = Rx$, i.e., as a linear regression through the origin, where R is the ratio. The absence of this relationship implies R depends on the value of x rather than being constant for all values of x.

7. Increased awareness of unresolved problems in data analysis regarding data transformations and < detectable numbers. It is clear that differences in methodologies exist on a number of subjects--especially data transformations and the handling of < detectable numbers. These problems should be studied and recommendations made to the OHER community.

8. Minimum standards for data reporting should be considered. As a very minimum, the arithmetic mean, standard deviation, and number of samples should always be reported. Probably no single thing could be as informative to other investigators, modelers, and statisticians as being consistent in always reporting the mean, standard deviation and sample size. We emphasize that these are minimum requirements, i.e., in most, if not all, situations, other summary statistics and data analyses are highly desirable.

9. Efforts should be made to assess variation in model prediction. Statistical and modeling techniques should be developed for estimating realistic confidence intervals or "ranges of uncertainty" about estimated parameter values such as transfer coefficients. This "stochastic" approach is recommended over the "deterministic" procedure of computing a single predicted

value unaccompanied by any estimate of precision or accuracy. The stochastic approach is recommended even when simplifying assumptions must be made regarding the shape of frequency distributions for estimates of model parameters. Additional insight might be gained by intercomparison studies of selected models developed by different laboratories or research groups. Predictions of terrestrial, atmospheric, or aquatic models could be intercompared starting from common base conditions and hypothetical source terms or releases. Whenever possible, predictions should be further compared to existing applicable field data.

Panel Discussion:

The above recommendations grew out of discussions within our panel concerning the role of models, types of models, modeling techniques, and the influence of data quality on modeling and statistics. In this section we summarize some of that discussion.

Modeling serves the purpose of numerically describing our current hypotheses about a process or set of processes. Once developed it has the advantage of inexpensively providing a method of examining the sensitivity of predictions to the parameters involved. The model is, accordingly, a valuable tool in designing and/or selecting future research projects to obtain additional information about important parameters. Modeling can also provide a holistic analysis. That is, OHER research is generally directed at individual elements of the environment, e.g., definitions of chemical behavior in aqueous environments, measurement of resuspension factors, etc. However, the end analysis must incorporate and integrate each of these elements.

We can consider models to be of two basic types: (1) those used to guide investigators into the structure and function of some system (termed "research" or "process" models), and (2) those used to provide information to decision makers, regulators, or assessors (termed "predictive" models). Although we term the second type predictive models, it is the intent of both types to estimate the magnitude of some phenomenon given certain measured values.

The process model is intended to describe the behavior and interrelationship of parameters for purposes of understanding the process. The requirements of the process model are that sufficient data points be obtained to identify the significant parameters and the form of their influence on the particular system under investigation. An iterative procedure for modeling and experimentation is typically utilized in the process model until the model duplicates the observation at some minimum level of acceptability. However, a process model is never finalized in the sense that it is always possible to extend it to greater levels of complexity. For example, an aspect of the model which treats an animal as a single unit may be further decomposed into organ systems, organ, tissue, cellular, and biochemical levels.

The process model typically forms the basis for prediction models. The prediction model may be used as a best estimator based on present knowledge for decision making purpose or as a means of demonstrating compliance with regulation. We may use large and involved predictive models that rival process models in their complexity and attention to detail, but this does not ensure their accuracy or validity. The predictive model is a user-oriented product whose success will be determined by minimizing the amount of site specific information required and the sophistication of the calculations to be employed while maintaining an "acceptable" level of accuracy. The simple model $P_1V_1 = P_2V_2$ is sufficient to predict the behavior of gases and does not require Einstein's thermodynamic explanation to work!

The major functional difference between process and predictive models occurs in what happens after they have been found to produce inaccurate predictions. The theory (hypotheses) underlying a process model must be rethought and restructured to incorporate additional phenomena so that the revised model more accurately predicts the real world. The revision of a predictive model may or may not require restructuring of theory. It is more likely that revision will involve some simple mathematical change such as substituting a non-linear function for a linear function.

The quality of data required for predictive models may be characterized in at least two ways: <u>accuracy</u> (data are unbiased and are measuring what we really need to meet study objectives) and <u>precision</u> (variance of estimated parameters is sufficiently small to meet our needs). Modelers are at the mercy of investigators with respect to the accuracy of estimates. This fact is not often understood by investigators. Modelers are often forced into using insufficient data or equally insufficient approximations. It should be hoped that when insufficient data are employed in model construction, the modeler appreciates the weakness of his model. Furthermore, the modeler should understand that an investigator may hold a model in ill reput if he finds his data misapplied.

Precision of data is a modeler's ratsbane. A transfer that is estimated with a 95 percent confidence interval whose width is only 10 percent of the estimate might be considered a good estimate of the "true" value. In spite of this, the prediction of the model could be altered by an order of magnitude by using either the lower or upper bound of the 95 percent confidence interval. The impacts of potential errors in parameter estimation upon model prediction is a current area of theoretical interest. Until more is known about the effects of inaccurate parameter estimation, we will have to work on the assumption that an estimate with a small confidence interval is, on the average, better than an estimate with a large confidence interval. This is particularly true for parameters which have a large controlling function on the behavior of the model.

This panel did not arrive at a comprehensive answer to whether or not presently available data are "adequate" for prediction of actinide movement in the environment. A central problem is that standards of adequacy have not been established, i.e., how accurate and precise do model predictions have to be? The answer to this question depends on the particular process under study and the consequences of an inaccurate prediction. Indeed, the establishment of acceptable error limits on model predictions for individual research projects or models would be most helpful in the design of environmental studies. Without specific guidance from management on this matter, decisions made by the statistician concerning the type, number and spacing of samples over the study site effectively determines the limits of accuracy and precision that can be obtained in model predictions.

It is our opinion that presently available data are probably sufficient to construct reasonably accurate predictive models for transport of plutonium to man. We say "probably" because we cannot be sure until we use the existing data to attempt to construct models or modify existing models, and decide what level of accuracy these models should obtain. At present, the data are probably insufficient to develop predictive models for Am, Th, U, Np, and Cm.

Conclusions:

The research goals of the OHER environmental actinide research program can be achieved with greatest efficiency when clear communication on research objectives, methods, and results exists between modelers, statisticians, researchers, and OHER management. We believe that modeling and statistics are not being fully utilized, that the efforts of the modeler, statistician, and researcher are often uncoordinated, and that the modeler is frequently provided with inadequate data for purposes of prediction. In the opinion of this panel, OHER should continue and expand efforts to improve communication and should consider establishing a modeling-statistics-researcher group (or person) within OHER to help direct and coordinate individual research projects.

Participants*

P. M. Altamere
C. T. Garten
R. O. Gilbert (Chairman)
C. A. Little
W. E. Martin
J. E. Pinder
R. G. Schreckhise
G. C. White

---

*The following members of the Statistics and Modeling Panel were not able to attend the Workshop: Dr. B. G. Bennett, Environmental Measurements Laboratory; Dr. Stephen V. Kaye, Oak Ridge National Laboratory, and Dr. L. L. Eberhardt, Pacific Northwest Laboratory.

AN ANALYTICAL LABORATORY INTERCOMPARISON EXERCISE

To assure a continuing knowledge of the accuracy and precision of analytical data and to sustain a high degree of scientific credibility, laboratories must maintain a systematic quality control (QC)-quality assurance (QA) program(1). Good QC-QA programs generally involve "blind" analyses of knowns, duplicates and blanks. The programs must be repeated often enough to enable perception of a change in quality. "Blind" means that to the extent possible, the QC-QA samples resemble real unknowns and that the analysts not be aware that they are in any way special.

Another very useful aspect of QC-QA is participation in interlaboratory intercomparisons. The main value in these efforts is in detecting systematic biases which may gradually and often imperceptibly occur. These efforts should utilize samples containing known concentrations of radionuclides in natural matrices(2) where practical.

In support of the freshwater and marine programs of the Office of Health and Environmental Research, DOE, the Environmental Measurements Laboratory (EML) organized and carried out an analytical laboratory intercomparison. This exercise focused on radionuclides of concern in the research activities of the major contractors funded by the Division of Ecological Research. For convenience, the exercise was dubbed BERLI-1 for the first Biological and Environmental Research Laboratory Intercomparison.

Seven samples were offered to the participants, on a voluntary basis. Each contractor chose the samples he/she wanted and analyzed for the radionuclides involved in their particular research. The samples offered were:

1. Fresh Water (FW)--about 2 liters of distilled water spiked with known amounts of Cs-137, Eu-152, Pu-238, Pu-239, and Am-241.

2. Sea Water (SW)--about 2 liters of sea water spiked as the fresh water.

3. River Sediment (RS)--50 - 100 g aliquots of National Bureau of Standards' "homogeneous river sediment" Research Material 45a.

4. Marine Sediment (MS)--50 - 100 g aliquots of the International Atomic Energy Agency's intercomparison sediment No. SD-B-3.

5. Saltwater Fish (SF)--50 - 100 g aliquots of a large freeze dried, homogenized sample of saltwater fish, spiked with algae containing elevated radionuclide concentrations.

6. Vegetation Meal (VM)--50 - 100 g aliquots of a large freeze dried, homogenized sample of carrots, spiked as the fish sample.

7. Soil (SO)--50 - 100 g aliquots of a large pulverized, homogenized soil sample containing elevated transuranic concentrations.

The samples were shipped in May and June of 1978. Table 1 lists the participating contractor laboratories and principal investigators. The numbers in parentheses in Table 1 denote the order in which data were received from the contractor laboratories. These numbers are used in subsequent tables as identifiers. Laboratories with no identifying numbers had not submitted analytical results by January 1, 1979.

One non-contractor laboratory, the Bhabha Atomic Research Centre, Bombay, India (Dr. K. C. Pillai), requested, was sent samples and has transmitted results. However, since the purpose of this report is to intercompare the contractors, the Pillai data are not included.

Results:

For this intercomparison, it was decided that the concentrations of the radionuclides of interest should be high enough to override the uncertainties encountered in low level radioassay. The natural samples were chosen and the spiked samples were prepared accordingly. The results were successful in this aspect as only a small number of data were reported as "less than," "not detectable" or with errors of greater than ±50 percent.

Tables 2 through 9 list all of the analyses received through November 15, 1978. Where it appeared useful, the median value was computed and wherever possible an "expected" concentration is listed on the final line in each table. These fall into four categories, more or less in order of decreasing confidence in our knowledge of the actual concentrations.

1. The fresh and seawater samples which were quantitatively spiked with known amounts of Cs-137, Ce-144, Eu-152, Pu-238, Pu-239+240 and Am-241 (henceforth in this report Pu-239+240 will be referred to as Pu-239).

2. The river sediment sample RM 45a was drawn from the same batch of wet blended material as was the NBS Standard Research Material No. 4350. The data strongly support the notion that for the radionuclides of interest in this intercomparison the concentrations in 45a and 4350 are essentially identical. The listed expected values are therefore those quoted (decay corrected where necessary) in Noyce _et al_.(3)

3. The marine sediment samples were part of a fairly large number of aliquots of IAEA intercomparison sample SD-B-3(4). In all, about 40 laboratories analyzed the sample for one or more radionuclides. The values quoted for Co-60 and the Cs isotopes are the averages obtained after rejecting outliers by Chauvenet's criterion, a method commonly used in IAEA intercomparisons. The IAEA report did not apply this criterion to the transuranic analyses because of suspected inhomogeneity. The expected values shown in Tables 6, 7 and 8 are the medians of all reported data in reference 4.

4. The expected values for the final three samples, salt-water fish, vegetation meal and soil are median concentrations of three or more analyses carried out at the Environmental Measurements Laboratory. The number of analyses are noted in parentheses.

No tabulations were made for nuclides reported by less than two participants. Hence a small number of submitted data for Sr-90, Sb-125, Ru-106, and Ce-144 do not appear in this report.

REFERENCES

1. Harley, J. H., Volchok, H. L. and Bowen, V. T., Quality Control in Radiochemical Analysis, in Reference Methods for Marine Radioactivity Studies ii, p. 63, International Atomic Energy Agency, Vienna, 1975.

2. Bowen, V. T., Natural Matrix Standards, Environment International, 1, 35-39, 1978.

3. Noyce, J. R., Hutchinson, J. M. R., Mann, W. B. and Mullen, P. A., Development of a National Bureau of Standards Environmental Radioactivity Standard: River Sediment, Institute of Electrical and Electronics Engineers, Inc., Annals No. 75CH1003-I 19-5, 1976.

4. Intercalibration of Analytical Methods on Marine Environmental Samples, International Laboratory of Marine Radioactivity, IAEA, Progress Report No. 17, July 1978.

TABLE 1
PARTICIPANTS IN BERLI-1

	Laboratory	Principal Investigator
(1)	Argonne National Laboratory	Dr. David N. Edgington
	Colorado State University	Dr. F. Ward Whicker
(10)	Lamont-Doherty Geological Observatory	Dr. H. James Simpson
(12)	Lawrence Livermore Laboratory	Dr. Florence Harrison
(2)	" " "	Dr. Victor E. Noshkin
(5)	Oak Ridge National Laboratory	Dr. E. A. Bondietti
(4)	" " "	Dr. Norman Cutshall
(8)	Oregon State University	Dr. Thomas Beasley
	Pacific Northwest Laboratory	Dr. Wallace Weimer
(3)	Scripps Institution of Oceanography	Dr. Edward Goldberg
	Savannah River Ecology Laboratory	Dr. James Alberts
(14)	Savannah River Laboratory (E.I. duPont)	Dr. Albert L. Boni
	" " "	Dr. David Hayes
(11)	Texas A & M University	Dr. Martha Scott
(9)	University of Washington	Dr. William R. Schell
	Washington State University	Dr. John C. Sheppard
(7)	Woods Hole Oceanographic Institution	Dr. Vaughan T. Bowen
(13)	Yale University	Dr. Karl K. Turekian

( ) Indicates order in which data were reported through November 15, 1978.

TABLE 2

K-40 RESULTS IN BERLI-1
(pCi/kg)

Laboratory No.	Sample Designation SW-1	RS-1	MS-1	SF-1	VM-1	SO-1
2	-	-	1.1×10^4	1.7×10^4	2.4×10^4	-
4	277	1.4×10^4	1.2×10^4	1.6×10^4	2.4×10^4	2.1×10^4
8	-	1.4×10^6	1.2×10^6	2.0×10^4	-	-
12	-	1.3×10^4	1.0×10^4	1.5×10^4	-	-
Median		1.4×10^4	1.2×10^4	1.7×10^4		
Expected		1.5×10^4				2.1×10^4 (6)

TABLE 3

Co-60 RESULTS IN BERLI-1
(pCi/kg)

Laboratory	Sample Designation						
No.	FW-1	SW-1	RS-1	MS-1	SF-1	VM-1	SO-1
2	-	N.D.	-	N.D.	1040	2620	-
4	-	-	2530	N.D.	1030	2500	-
7	-	-	2310	111	917	2550	*
8	-	-	2480	86	1149	-	-
9	<2	*	2460	146	900	2300	<75
10	-	-	2580	105	-	-	-
12	-	-	2430	112	1036	-	-
14	<2	<2	3370	125	760	3100	<.2
Median			2480	112	1036	2550	
Expected			2470	90(5)			

N.D. = not detectable or trace

\- = no data reported

* = error reported ≥ 50%

( ) = No. of analyses reported.

TABLE 4

Cs-137 RESULTS IN BERLI-1
(pCi/kg)

Laboratory No.	Sample Designation FW-1	SW-1	RS-1	MS-1	SF-1	VM-1	SO-1
1	6.3	6.7	2590	1.77×10^5	2330	6025	600
2	-	N.D.	-	1.83×10^5	2880	7000	-
3	5.5	-	1690	1.67×10^5	1830	-	-
4	8.1	9.2	2480	1.79×10^5	2900	6650	530
7	6.1	5.6	2180	1.71×10^5	2540	6130	398
8	8.4	8.7	2570	2.34×10^5	3150	-	-
9	4.7	5.1	2490	1.62×10^5	2780	6570	449
10	-	-	2440	1.96×10^5	-	-	-
12	.54	.95	2207	1.79×10^5	2702	-	-
14	17.9	6.9	2970	1.99×10^5	2260	7550	410
Median	6.2	6.7	2480	1.79×10^5	2780	6610	449
Expected	5.9	5.9	2480	1.81×10^5 (32)	3050 (12)	6490 (9)	480 (6)

N.D. = not detectable or trace

- = no data reported

( ) = No. of analyses reported.

TABLE 5

Eu-152 RESULTS IN BERLI-1
(pCi/kg)

Laboratory No.	Sample Designation FW-1	SW-1	RS-1	MS-1	SF-1	VM-1	SO-1
2	-	5.8	-	N.D.	N.D.	N.D.	-
4	-	-	4950	-	-	-	-
7	-	-	4610	<1000	<1000	<1000	11,200
8	-	-	4840	194	-	-	-
9	6.5	5.0	5330	*	<80	<100	<70
12	-	-	450	-	-	-	-
14	<3	<.5	2870	<.03	<.03	<.06	<.02
Median		5.0	4780				
Expected	5.9	5.9	4850				

N.D. = not detectable or trace

- = no data reported

TABLE 6

Pu-238 RESULTS IN BERLI-1
(pCi/kg)

Laboratory No.	Sample Designation FW-1	SW-1	RS-1	MS-1	SF-1	VM-1	SO-1
1	.026	.029	1.4	21.6	1.5	4.5	57.8
2	-	.03	-	20.	1.7	4.1	-
3	.024	.027	1.1	17.3	1.9	-	-
5	-	-	-	-	-	4.3	58.
7	.021	.023	1.34	19.4	1.38	2.9	50.9
8	.030	.023	1.53	23.0	1.44	-	-
9	.053	.101	7.	14.	6.	18.	32.
10	-	-	1.4	19.8	-	-	-
11	.029	.027	1.4	20.6	-	-	52.8
13	-	-	-	17.8	-	-	-
14	.013	.027	1.4	15.4	1.51	3.5	44.2
Median	.026	.027	1.4	20.	1.5	4.3	52.8
Expected	.024	.024	1.45	21. (38)	1.58 (4)	3.9 (4)	-

- = no data reported

( ) = No. of analyses reported.

TABLE 7

Pu-239 RESULTS IN BERLI-1
(pCi/kg)

Laboratory No.	Sample Designation						
	FW-1	SW-1	RS-1	MS-1	SF-1	VM-1	SO-1
1	.097	.101	44.4	604	20.2	47.6	2680
2	-	.107	-	570	19.6	43.	-
3	.10	.10	40.0	541	17.4	-	-
5	-	-	-	-	-	43.	3170
7	.094	.094	35.7	560	17.8	35.6	2750
8	.13	.11	37.8	626	18.9	-	-
9	.035	.114	21.	369	13.	26.	1700
10	-	-	34.5	530	-	-	-
11	.063	.090	34.9	586	-	-	2940
13	-	-	32.3	508	-	-	2103
14	.060	.066	36.0	430	17.2	37.6	2350
Median	.096	.101	35.3	560	18.4	43.	2720
Expected	.098	.098	37.8	606(39)	19.1(4)	42.6(4)	2750(5)

- = no data reported

( )=No. of analyses reported

TABLE 8

Am-241 RESULTS IN BERLI-1
(pCi/kg)

Laboratory No.	Sample Designation FW-1	SW-1	RS-1	MS-1	SF-1	VM-1	SO-1
1	.027	.026	7.0	142	4.0	6.2	245
3	-	.025	10.0	194	4.7	-	-
5	-	-	-	-	-	13.	392
7	.037	.037	8.8	188	6.1	13.1	349
8	.040	.039	9.9	162	6.0	-	-
9	*	-	-	-	-	-	392
14	<2.	<2.	<120.	<230.	<160.	<110.	<310.
Median	.037	.031	9.4	175	5.4	13.	371
Expected	.035	.035	8.5	158(26)	4.1(6)	11.7(3)	350(3)

- = no data reported

* = error reported ≥ 50%

( ) = No. of analyses reported

TABLE 9

OTHER RADIONUCLIDES IN BERLI-1 (pCi/kg)

Laboratory No.	MS*	RS*		SF*
	Cs-134	Eu-154	Eu-155	Cm-244
4	900	1300	-	-
7	-	-	-	13.7
8	1081	1140	198	17.6
9	-	-	116	-
12	1036	86	-	-
14	985			
Median	1010	1140		
Expected	1500(18)	1400		

*Sample designation

☆U.S. GOVERNMENT PRINTING OFFICE:1980 310-912/189 1-3