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Workshop On Environmental Research For Actinide Elements

Proceedings Of The Workshop
March 17-19, 1981
Lawrence Livermore National Laboratory
Livermore, California

September 1981

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

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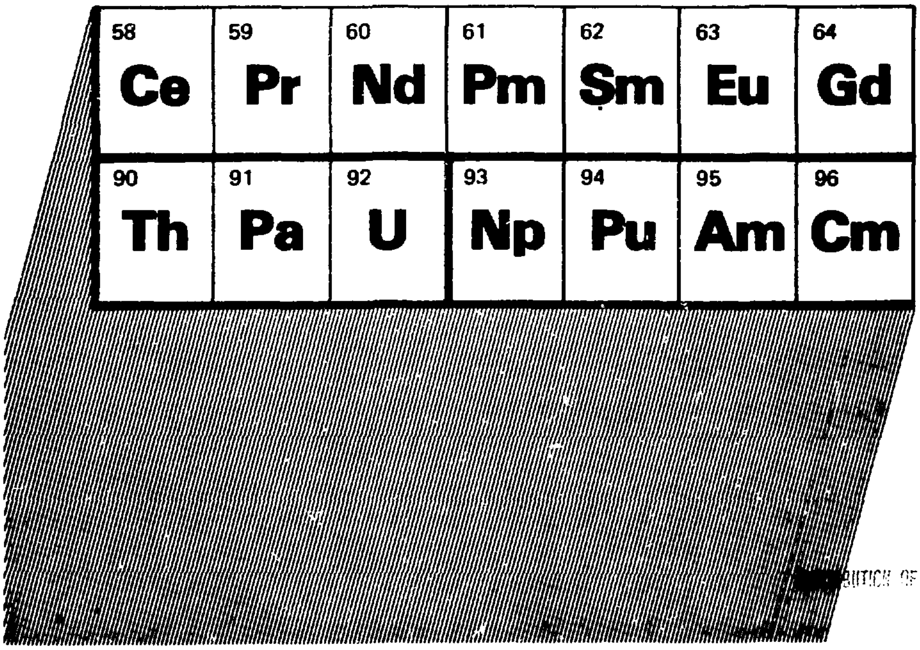


INTRODUCTION

This workshop was the fifth which the Department of Energy and its predecessors have sponsored to produce a forum for discussions about research on actinides in the environment. The workshops bring together research scientists and specialists in radiological hazards evaluation to discuss informally the latest research results, research problems, and what information is needed for radiological evaluations.

The discussions in this workshop were directed to the advances which have been made in the environmental chemistry of plutonium and to the feasibility and worth of developing environmental transport models which might serve as predictive tools for long term behavior and as guides for future research needs. Two models of the soil/plant pathway were presented for critique and as examples of possible approaches. These produced considerable interest and both positive and negative responses in the four discussion panels.

These panels and their chairman were:

Model Development	R. G. Schreckhise
Marine	D. M. Nelson
Terrestrial	W. C. Hanson
Freshwater/Groundwater	J. J. Alberts

The Office of Health and Environmental Research staff wishes to thank all of the participants for their cooperation and thoughtful contributions. Special thanks are extended to the panel chairmen for their time and effort in planning, conducting, and reporting the panel discussions.

MODEL DEVELOPMENT - PANEL SUMMARY

WORKSHOP ON ENVIRONMENTAL RESEARCH FOR ACTINIDE ELEMENTS

March 17-19, 1981
Lawrence Livermore Laboratory

Panel Participants:

Chairman - Gene Schreckhise

S. Bard
B. Bartrum
D. Cataldo
C. Cowan
P. Durbin
C. Garten
D. Gilbert
E. Jenne
D. Kocher
B. Napier
J. Pinder
J. Thomas
B. Watters
G. White

ABSTRACT. The model development panel consisted of participants with varied backgrounds and opinions. There appeared to be a consensus on the following points: 1) the Cowan-Jenne plant submodel appears logical and well behaved with respect to what is known about Pu dynamics in plants, 2) there exists too little rate data (kinetic data) which is needed to model actinides in the environment, 3) the purpose of the two present Pu models (Pinder and Cowan-Jenne models) is not to predict radiological dose but rather to understand the process of Pu uptake by plants and Pu immobilization in soil, 4) we have a possible range of simple to complex models for actinides in the environment; all can help suggest research and all can help researchers conceptualize their thinking about the environmental behavior of actinides, 5) model predictions of the long-term behavior of Pu and other actinides in the environment are characterized by large uncertainty.

MINUTES

March 17

The panel convened after the plenary morning session at which Pinder and Jenne both spoke about their versions of a soil-plant model for plutonium. First item of business was informal introduction of panel participants and their work interests in the panel. A list of panel participants precedes these minutes.

Schreckhise then briefly summarized the evolution of the current modeling effort on plutonium. Beginning at the 1978 workshop at Airlie House with the Statistics and Modeling Panel there was a perceived need to summarize and draw together various research work on actinides. In May 1979, a followup workshop in Aiken, S.C., brought together model users (dose assessment people) with experimentalists and field researchers for discussions on the development of a model for long-term forecasts of Pu in the terrestrial environment. A modeling committee (Schreckhise, Pinder, Thomas, Garten, and Watters) first met in Washington in August, 1979, and conceptualized the model "Dirt" which was later summarized in a written draft document by Pinder. Jenne joined the modeling committee at its meeting in Seattle in July 1980. Since then Pinder (SREL) and Jenne and Cowan (PNL) have been developing independent but not necessarily alternative models for Pu in soil-plant systems. (These were summarized in draft documents sent to panel participants prior to this meeting.) Schreckhise set forth objectives for the current panel: critique the existing models and arrive at a concensus as to future modeling needs, types of data needed for modeling, and the best future direction of the modeling project.

The previous written report of the Statistics and Modeling Panel at Airlie House in 1978 was briefly reviewed with attention to recommendations of the panel at that time and actions taken since that time to fulfill the panel recommendations. Progress has been made on many of the previous panel's suggestions with notable examples being increased discussion and communication via workshops, the modeling committee, and publication of Trans-Stat from PNL.

For the panel, Cowan presented a more detailed discussion and analysis of the Jenne-Cowan plant submodel for plant uptake of Pu from soil. The results, from simulations of translocation of Pu into a plant were shown and discussed. There was reasonably good agreement between laboratory data and time dependent behavior of the model predictions. The model was calibrated on few data points because of a lack of kinetic data for Pu in soil and plants. There appeared to be unanimous agreement that very little rate data of the type needed for modeling actinides is available from laboratory or field research. Data showing the time dependent behavior rate data of actinides in the environment are needed for present and future modeling efforts.

Cowan pointed out that more work needed to be done on the model equations and simulations of the plant submodel. The model appears to be well behaved and logical with respect to what is known about Pu dynamics in plants. The model predictions might be validated with hydroponic experiments. Cowan summarized with several conclusions: 1) the amount of Pu in leaves depends on the initial soil concentration, 2) plant storage of Pu is important, 3) transpiration may be a driving force for Pu translocation, and 4) within limits the time at which the Pu was added to the hydroponic solution did not affect the final amount in the leaves.

Panel discussion of the model followed Cowan's presentation. Several questions were raised. How does such a model apply to long-term dose calculations? How important is the long term if the growing cycle is only one year? Isn't the soil system of greatest importance for long-term predictions? What are the critical experiments and data needs for the Jenne-Cowan model? How was the model parameterized? How should the variability in rate parameters be dealt with in simulations and validation?

Suggestions and comments from the panel included the following. Despite short growing seasons up to 80% of the plant may be left in the field following harvest. Soil is the more important model component for long-term forecasts. Critical data needs for the model at this time are growth rate data, transpiration rate data, and calibration data on Pu kinetics in plants (i.e. storage and uptake). Some data on parameter variability can be obtained (between pot variation). The model might be first validated using elements other than Pu.

Next Pinder answered questions from the panel regarding his model. The exact solid phase compartments in the model are unknown, rather they are abstract and their existence is hypothesized based on empirical soil sorption-desorption rate data. Again there is little rate data on Pu sorption-desorption for different soil types.

— BREAK —

Following the break there was some general discussion of loss mechanisms from soil. A variety of opinions were voiced by participants. Loss rates are important but difficult to determine. Plowing results in soil disturbance with some downward migration of Pu. Leaching by water, physical movement, and biological turnover are all important. Biological transport is probably more significant than leaching. Soil erosion may remove Pu from soil faster than radioactive decay.

The purpose of the modelling exercise was next explored. Questions: 1) How does the current modelling focus fit in with predicting dose? 2) What about natural variability (soil-plant types) over different geographic regions? The purpose of the plant-soil models presented is to understand the process of Pu uptake by plants — the purpose is not to predict dose per se. Detailed models of the type being developed will help ascertain if the more general simplified dose models are "valid". The application of the detailed plant submodel in regional or environmental type assessments is a more distant consideration for the model builders at this time.

The modelling approach was discussed. If Pu transfers in the environment eventually come to steady state then a specific activity approach to predictions, using analog elements, might be feasible. There was uncertainty as to whether Pu ever approaches steady state in the environment. The current approach is to build a detailed soil-plant model with unknown parameter values rather than build a model based on available data. The latter approach is too constraining and discourages research. Agricultural processes, such as combining, should probably be included in the models. Simplification of the detailed models is always possible at a later time.

Gary White presented data on the importance of splash up at LANL garden plots. As much as 50% of the field CR value is due to splash up of small soil particles by rain onto plant surfaces. A fraction of the remaining 50% is due to wind resuspension of soil onto plants and a smaller fraction is due to uptake via plant roots. Some general discussion of foliar uptake followed. The foliar uptake of Pu in some environments can be many times more important than root uptake but it depends on the concentration of Pu in soil.

— ADJOURN —

March 18

Modelling panel participants attended other panel meetings.

March 19

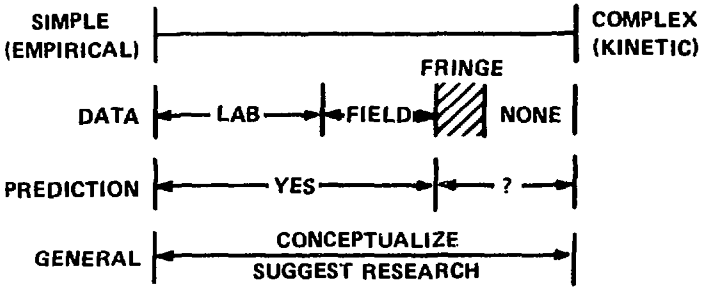
The panel reconvened for final discussions and preparation of the summary report for the afternoon plenary session.

Gary White discussed the use of nonlinear least squares parameter estimation (NLSPE) and its application to obtaining parameter estimates for the type of soil-plant models that have been developed for Pu. Based on empirical data the available computer programs for NLSPE can arrive at unbiased and minimum variance parameter values with confidence intervals. The differential equations for the model are required input to the computer programs. Two available computer programs are SIAM (from NIH) and BMDPAR. These programs can help one determine if the data and the model fit well together. They work well for monotonic functions but not so well for oscillatory data.

Figure 1 depicts White's concept of the spectrum of modelling problems the panel is dealing with at present. We have a range from simple to complex models all of which may suggest research and all can be conceptualized. Not all models can be used for prediction because some models have no data for parameterization. There was disagreement within the panel about whether we should predict something beyond our available data base.

Schreckhise summarized the state of our current knowledge about actinides. Radiation doses can be estimated for Pu and only with considerable uncertainty for other actinides. Predictions of the long-term behavior of actinides using mathematical models are characterized by large uncertainty.

Part of the problem in communication between modellers and researchers has been semantics. As a group we have given too little attention to definition of terms and perhaps created confusion among researchers about modelling purposes and techniques. Although no official poll of the workshop was taken there appeared to be pessimism and skepticism among many workshop participants as to the worth of a long-term model for actinides in the environment. Perhaps some cost-benefit analysis of the modelling effort would resolve differences between optimists and pessimists and settle the question of "Is the modelling effort worth continuing?"



MODELING SPECTRUM

FIG. 1

TERRESTRIAL ECOSYSTEMS - PANEL SUMMARY

WORKSHOP ON ENVIRONMENTAL RESEARCH FOR ACTINIDE ELEMENTS

March 17-19, 1981
Lawrence Livermore Laboratory

Panel Participants:

Chairman - W. C. Hanson

D. C. Adriano
E. A. Bondietti
L. L. Cadwell
D. A. Cataldo
P. B. Dunaway
T. R. Garland
T. E. Hakonson
D. K. Halford
T. Jaakkola

P. W. Krey
K. McLeod
H. Nishita
E. M. Romney
G. A. Sehmel
H. E. Walberg
W. C. Weimer
T. F. Winsor

Introduction

The panel wishes especially to thank our distinguished visitors, Drs. R. Fukai and T. Jaakkola, for presenting their research results on short notice. The marine behavior of Pu and Am in the Mediterranean Sea was of great interest to the terrestrial investigators because of its added dimension to understanding transport mechanisms. The preliminary results from actinides in Finnish Lapps was of great importance in providing initial data on food chain transport to upper trophic levels.

Modelling

The Terrestrial Panel discussions revolved around two basically different models, defined as simplistic and process models. The first type consists of linear models for which ranges of Concentration Ratios (CR's) are available and which are generally applicable for dose estimates. The second type is a process model of fine detail for which several experiments and inputs will be required that demand a degree of sophistication that is not now available.

Models should evolve from simplistic to complex only as reliable scientific data and sensitivity analyses drive the evolution. When data sets are extensive, quantitative models can be developed to describe the system mathematically. When data are not available or where ranges of parameters are small as compared to plutonium, models are less appropriate to risk estimates and are restricted to testing hypotheses advanced jointly by experimenters and modelers.

The panel concluded that our present limited understanding of actinide distribution and transport in terrestrial ecosystems requires separate model development approaches. Once a process model is in place and is used for predicting future actinide behavior, the assignment of parameter values cannot involve data more precise than existing experimental values. Foremost in this category are data derived from "real world" investigations which are reasonably based.

We recommend

1. Models should be no more complex than real world data allow.

2. Field data should be evaluated to establish priorities in model development including an evaluation of physical transport of the actinides.

3. Predictive models should not be complicated with processes that cannot reasonably be validated.

Environmental Chemistry

The panel concluded that the Concentration Ratios (CR's) for plutonium that were reported at the second workshop on Transuranic Elements in the Environment are still applicable and reinforced by subsequent research. Better understanding of these data and their application to specific environmental areas now provide reasonable basis for prediction of actinide transport. Research results suggest that extension of the CRs to actinides other than plutonium are as follows.

1. ^{241}Am is more mobile within biotic systems than Pu, often by an order of magnitude;
2. ^{244}Cm can be considered to behave similarly to ^{241}Am; and
3. ^{237}Np is often 10^3 times more mobile than Pu, with CR's (plant:soil) of 10^{-1} to 10^0.

We feel that research is needed to define more accurately the ecological behavior of ^{237}Np in particular. It is recommended that carefully considered studies in this area be supported. Such studies probably must be conducted at a few radioactive waste disposal sites or in carefully controlled laboratory or greenhouse environments.

Studies of secondary importance, in the view of the panel, were suggested for uranium. Its ubiquity in the environment has been generally taken for granted and little focused research on its environmental transport has been undertaken.

Finally, we recommend to all researchers that careful attention be given to the compendium of results embodied in the book Transuranic Elements in the Environment, TIC/DOE 22800.

FRESHWATER/GROUNDWATER - PANEL SUMMARY

WORKSHOP ON ENVIRONMENTAL RESEARCH FOR ACTINIDE ELEMENTS

March 17-19, 1981
Lawrence Livermore Laboratory

Panel Participants:

Chairman - James Alberts

N. Cohen
D. Hayes
H. Nishita
J. Trabalka
W. Weimer
W. Lei
J. Swanson
G. Kuzo
L. Brush
T. Sibley
L. Kirby
P. Linsalata

The Freshwater/Groundwater panel met in joint session with the Marine panel on two occasions to discuss the environmental chemistry of the actinide elements and the modelling efforts as currently defined by the two actinide transport models. The results of those efforts will be discussed under the appropriate section headings.

Environmental Chemistry

A considerable amount of time was spent in discussions centering on the oxidation states of plutonium in aquatic systems. Researchers at Argonne National Laboratory (Nelson and Larsen) have demonstrated that plutonium exists in the Pu(V) oxidation state in Lake Michigan waters. In addition, they have shown that leaching of freshwater sediments from the canals at Mound Laboratory with filtered Lake Michigan water rapidly produces a steady state plutonium concentration in the water and that the plutonium is in the (V) oxidation state under those conditions. Further evidence of the stability of the Pu(V) oxidation state in aquatic systems was presented by researchers from PNL (Swanson), who showed in laboratory experiments of the dissolution of plutonium oxides that the plutonium in solution was the (V) oxidation state at pH<4. The results of both ANL and PNL workers do not agree with the study at ORNL (Bondietti) which indicates a long term equilibrium of Pu(V) and Pu(IV) under laboratory conditions. This discrepancy is yet to be resolved.

It was demonstrated (Nelson) that the addition of natural organic matter, which was isolated by ultrafiltration of Okefenokee Swamp water, to Lake Michigan water caused a reduction in the oxidation state of Pu(V) to either Pu(IV) or Pu(III). These laboratory studies are in agreement with earlier field observations that the concentrations of reduced plutonium oxidation states (III and IV) are positively correlated with dissolved organic carbon concentrations. These results plus the observations that plutonium in the highly reducing groundwaters at Maxey Flats, Kentucky, is in reduced oxidation states, highly complexed and predominantly anionic (Kirby), point up the need to understand the chemistry of the solution before making predictions of the expected oxidation state of plutonium in aquatic systems. Discussions related to this point led to a reemphasis of caution in using stability field diagrams to predict the oxidation state distribution of plutonium in natural systems. Calculations of free energies of the plutonium oxidation states using formal oxidation-reduction potentials at various pH values (Alberts) showed the need to know the concentrations of the possible couple reactions in natural systems before full use of thermodynamic values may be employed. Furthermore, the question of kinetics was discussed relative to these reactions and it was emphasized that reactions are dependent on molar concentrations of reactants. This latter point may become important when reactants have concentrations between 10^{-8} to 10^{-17} M.

Research on other actinides was also discussed by the panel. Data was presented (Sibley) to show that the concentration ratios (concentration per unit mass sediment/concentration per unit volume) often reported as K_d values for actinides in sediment/water system are not independent of sediment/solution ratio, but rather increase with sediment weight. This increase is not linear. Furthermore, concentration ratios determined by both adsorption and desorption studies of the same sediments show that ratios in desorption experiments are higher by as much as one order of magnitude than the same ratio in an adsorption experiment. These findings were confirmed by observations made on deep ocean sediments (Brush). The explanation of these results are currently unknown, but it is apparent that processes are active in soil solution which need to be defined before predictive statements may be made about the solid/liquid distribution of actinides under changing environmental conditions. One example of this phenomenon was presented (Alberts and Sibley) for the dependence of Cm concentration ratios on pH and organic matter. Increasing pH above 7 results in a decrease in concentration ratio of Cm in highly organic sediments apparently as a result of stable Cm organic complexes forming and remaining in solution. This phenomenon did not occur with low organic concentration sediments.

Again, the importance of both organic matter and suspended particulate matter was emphasized (Hayes) in the transport of actinides in rivers. High organic concentrations in southeastern coastal rivers are correlated with higher plutonium concentrations in those rivers as compared to plutonium concentrations in Piedmont rivers where particulate loading is important.

General discussions in the panel pointed up the fact that considerable progress has been made in understanding the environmental chemistry of plutonium in the Laurentian Great Lakes and some other surface waters of similar chemistries. However, it was also apparent that our understanding of other actinides and other types of waters are lacking. The example of movement of transuranic elements in groundwaters at Maxey Flats, Kentucky, (Kirby) showed that plutonium was in an entirely different physico-chemical form than in surface waters and that Am concentrations were comparable to those of ^{238}Pu. Much of the difference can be attributed to the highly reducing environment of groundwaters, the high soil/water ratio which increases the number of reactive interfaces and the variable source term. Also, our knowledge of Th and other possible analog elements in the freshwater environment is small. Studies are being initiated to investigate some of these questions (Lei), however most of the work is yet to be undertaken.

Modelling of Actinide Movement

The joint panels discussed the current modelling effort at some length. In addition, further discussion was carried on by our panel after the joint sessions.

The two models which have been constructed to date deal with the transfer of actinides to crops primarily via root uptake. Relative to these two models, the panel felt that we could address only one small compartment of either model, that being soil solution chemistry. The panel felt that both modelling approaches had difficulties. The "DIRT" model requires knowledge of the kinetics of adsorption and desorption of actinides from various components of the solid phase. Little data exist which are applicable to this approach. The other model depends on thermodynamic data in an existing geochemical equilibrium model to predict the behavior of actinides in soil solution. Faults with this approach include: the validity of the existing thermodynamics data including the contribution of natural organic matter, the assumption of equilibrium, the lack of knowledge of the effect of microenvironments and microcontaminants on the chemistries of the actinide elements, and the general lack of knowledge of the chemistry of actinides other than Pu in the widely divergent physicochemical conditions encountered in nature. While both approaches have problems associated with them, the panel felt that neither had an inherent advantage and both should be pursued in an attempt to define the soil/crop processes.

The discussion of modelling expanded to include models for aquatic systems. It was felt that the basic assumptions of the current modelling efforts included an acute source term, predictions on a time scale in excess of 100 years and an assessment of impact on man. These basic assumptions may need modification or expansion in aquatic environments.

When considering source term, the energy regime of the environment into which the source is introduced must be considered. In high energy environments, the material is most likely to be transported into a low energy environment which will act as a "sink." For long term prediction, the high energy environment may not exist at the future time scale being considered. This phenomenon is purely a transport mechanism and several transport models exist which may be suitable to describe this movement.

If material is moved to a "sink," it will redistribute within that system based on the physiocochemical regime of the environment. For present "sinks," this redistribution may be effectively predicted by existing dispersion models coupled with our knowledge of the phenomenalogical models which have been constructed for Lake Michigan and the world oceans. If the physiochemical regime of a "sink" changes to the extent that the actinide elements will be mobilized out of the sediments, the source term will no longer be an acute input, but rather a chronic release. In addition, a physiocochemical change which would change the form of the actinides would also drastically affect other major biogeochemical cycles such as iron, manganese and carbon. The knowledge of the kinetics of these changes is limited.

Finally, the processes which control food web dynamics of actinide elements in aquatic systems are poorly understood. Data from ORNL (Trabalka) demonstrate that the concentrations of plutonium, americium and curium in livers of freshwater fish cannot be predicted by the concentrations of those elements in the water column. Similar data are available for the flesh of fish from oceanic environments (Noshkin). Hence, more investigations into food web processes in aquatic systems is required before values can be placed in any model of impact on man of actinide elements through a freshwater food web.

The question was raised as to whether a model is required for actinide transport in the aquatic environment. The panel felt that several reasons existed for the construction of such a model: (1) to predict the ramifications of an acute source with a high probability of dose to man, (2) to gather the existing data base from a wide variety of aquatic environments, (3) to be used as an aid in defining future research, and (4) to ensure the general public that all habitat types are being considered in risk analysis. The panel further felt that if it is decided that a model should be required, the process for developing the initial model be the same as that for the development of the current modelling effort. A small group of people, familiar with the aquatic actinides programs and knowledgeable in state-of-the-art aquatic modelling, be assembled to "rough out" the initial effort. These people should come primarily from the existing group, but outside expertise should be sought if required. After an initial model has been conceptualized, it should then be given to the group at large for critique.

Summary of Panel Recommendations

1. Emphasis must be placed on understanding the environmental chemistry of actinide elements other than plutonium.

2. Our knowledge of the environmental chemistry of plutonium must be expanded to include the range of diverse chemistries extant in the environment, including groundwater and soil solution.

3. Use of existing thermodynamic data to predict the environmental chemistry of actinide elements must be done with caution. Consideration of the whole solution chemistry, concentrations of possible reactants, and rates of reaction are necessary to quantitatively describe the system.

4. Further work is required to understand the sedimentary processes which govern concentration ratios of the actinide elements.

5. Studies are required into the dynamics of actinide elements in aquatic food webs.

6. Current modelling efforts in the actinide program have little bearing on aquatic systems. If the models of the transport of actinide elements in aquatic systems are required, an effort must be made to synthesize already existing aquatic models with our knowledge of actinide element biogeochemistry.

MARINE - PANEL SUMMARY

WORKSHOP ON ENVIRONMENTAL RESEARCH FOR ACTINIDE ELEMENTS

March 17-19, 1981
Lawrence Livermore Laboratory

Panel Participants:

Chairman - D. Nelson

T. M. Beasley
G. R. Choppin
N. Cohen
D. N. Edgington
R. Fukai
E. D. Goldberg
L. S. Gomez
J. E. Halverson
M. Koide

R. P. Larsen
J. W. Morse
A. Nevessi
R. Ray
D. E. Robertson
W. R. Schell
M. R. Scott
H. J. Simpson
H. V. Weiss

The marine panel covered a broad range of topics during the meeting with major emphasis on the problems and potential of transport modelling. The discussions centered on plutonium since the data set is much more extensive for plutonium than for the other man-made actinides. Even for plutonium, however, there was considerable disagreement as to the mechanisms responsible for transport and there was considerable support for the view that extensive mathematical modelling efforts may still be premature.

One area in which the marine panel showed almost total agreement was in its response to the type of soil-plant models presented during the meeting. We felt that, while these models may be useful for certain problems, this approach would be exceedingly difficult to adapt to the marine transport problem because the models were formulated to describe a physical environment which is very different from that found in the oceans. In this system the principal mechanisms responsible for moving plutonium and the other actinides are particle settling and large-scale circulation. Neither of these processes has a counterpart in the terrestrial environment and hence they were not considered in these models. Any model which attempts to describe actinide transport in the oceans must be built around these two processes. The panel felt that useful models have been developed to describe other transport phenomena in the oceans and that these other models could be used to advantage in the formative stages of the plutonium model.

The dynamic nature of the marine environment imposes some severe limits on the oceanographers' ability to make unambiguous time-series measurements, a problem the terrestrial scientist normally does not encounter. For instance, the plutonium in a marine sample may have entered the system hundreds or even thousands of miles from the sampling location and, because of differences in current structure with depth, the plutonium at one depth could have a very different history from that at another. The data needed to calibrate and validate a marine model must therefore be collected over an extensive area and the measurements must be repeated with some regularity to establish trends.

In practice, because of the great expense of doing research at sea, the amount and type of data available have been limited. This places a major constraint upon our ability to construct marine models. Our basic data set has been derived from observations made over a limited time span and at relatively few locations. For the open ocean we are almost entirely dependent upon observations of fallout derived nuclides.

Fortunately, fallout plutonium is an excellent tracer as it was distributed over wide areas and was apparently delivered in an easily solubilized form. Its behavior is assumed to be that toward which all more insoluble forms will tend with time. To a large extent, then, our task involves following the course of a large tracer experiment in which the surface waters of the world's oceans were contaminated with plutonium, primarily during the late 1950's and early 1960's, and in which this plutonium is now being redistributed. Unfortunately, few measurements were made during the period of maximum input and consequently the best time to make measurements of the rapid transport processes (which are critical to model development) was lost. This lack of early measurements complicates, but does not render hopeless, the task of model development.

Substantial progress has been made in the past few years in collecting the data needed to describe the movements of plutonium (and to a lesser extent americium) in the ocean. Regularities in this data are emerging particularly with regard to the physical distributions of the elements and to their chemical speciation. These regularities suggest that the principle interactions may be common over wide areas and offer encouragement that satisfactory transport models can eventually be developed.

The panel recognized that an adequate knowledge of the input function, in regards to both its magnitude and isotopic composition, is essential to the model building process. Isotopic composition proved to be a major topic of discussion throughout the meeting since in many cases a change in isotopic ratio can be used to observe events which would otherwise go undetected. Recent measurements of the isotopic composition of plutonium in glaciers show a distinctive signature for fallout from the early years of weapons testing. For these early years the magnitude as well as the isotopic composition of the input must be established from the glacial record since direct measurements are not available. The fallout from the early 1950's is characterized by ratios of ^{241}Pu to ^{239,240}Pu which are about twice as high as those observed later. These high ratios are also found in the deepest plutonium bearing layers of sediment collected from anoxic basins off the west coast of North America. The existence (or absence) of measureable changes in isotopic ratio with depth in cores or with location is a powerful tool which can be used to test the validity of proposed transport processes both to and within sediments.

An illustration of the usefulness of plutonium isotopic ratios comes from measurements made in cores collected from the Gulf of Mexico. Changes in ^{240}Pu/^{239}Pu as a function of location indicate that at least two distinct sources have contributed to the plutonium inventory in Gulf sediments. Nearshore, where the total plutonium inventory is high, the ratio is near the global fallout average of ∿0.17. In deep water at the center of the Gulf, where the inventory is low, the ratio is much lower, ∿0.06. While the source of this "low ratio" plutonium has not been identified, it was apparently delivered in a form having more efficient transfer to deep water sediments than is typical of global fallout. Any estimate of this transfer rate based on the assumption of a single input (global fallout) and measurements of total plutonium would, therefore, be erroneous in this instance.

Another area in which isotopic ratios have been used to advantage in decifering a multiple input problem is the Columbia River system. The Hanford reactors contributed significant amounts of ^{239}Pu to the Columbia due to neutron activation of ^{238}U in the river water that was used to cool the reactors. This plutonium is detectable (using its distinctive isotopic ratio) in cores collected at the McNary reservoir downstream. At this location it constitutes an appreciable fraction of the total deposition. The isotopic ratio of plutonium in sediments at the mouth of the Columbia, however, is indistinguishable from that in global fallout. The amount of plutonium transferred from Hanford to the Pacific Ocean is apparently insignificant relative to fallout derived plutonium.

The general question of the source of the plutonium which is accumulating in sediments near the mouths of major rivers has been addressed for both the Columbia and Mississippi Rivers. Although the plutonium flux to individual cores frequently exceeds fallout delivery, in neither instance does the river appear to be the primary source. Not only do the total plutonium inventories off the mouths of these rivers exceed the estimated transport down the river, but the concentrations found in surficial sediments are several fold higher than the concentrations in suspended solids in the rivers. Furthermore, no substantial difference in plutonium inventory is observed between sediments collected outside the influence of the river discharge and those within.

A consistent pattern of plutonium accumulation, often in excess of fallout delivery, is found in coastal sediments from many parts of the world. The majority of the panel felt that this "excess" plutonium was introduced, as global fallout, elsewhere in the ocean and that it has since been moved by advective processes into these areas where transfer to the sediments is more rapid. Typically, the plutonium concentration in these sediments is fairly constant down to a depth of several centimeters and it then decreases logarithmically with increasing depth. The plutonium concentration in the upper layers is in the range of 30-70 fCi/g at a variety of locations. Most of these shallow water cores have been collected in oxic environments having low sedimentation rates where post-depositional mixing by organisms obscures the historical record of the inputs. A few cores, however, were collected from anoxic basins where bioturbation is absent and these do record faithfully the temporal history of the inputs. The depositional patterns in these cores, which also have a several fold excess of plutonium, indicate an essentially constant input from the mid-1970's back to the early 1960's with an approximately logarithmic decrease at greater depth. The shape of these deposition curves resembles the integral of the fallout delivery function and suggest that the plutonium being added to these sediments is coming from a reservoir whose concentration is proportional to the sum of the fallout input. Some of the panel argued eloquently that this reservoir was desert soil and that the plutonium was moved by the wind from land into the coastal oceans. Most of the panel remained unconvinced, however, citing data from the long history of fallout monitoring which has not shown resuspended dust to be a major contributor to airborne radionuclide concentrations.

The mechanism responsible for generating the plutonium profiles found in oxic cores remained a topic of active debate. There was general agreement among those present that, for most sediments, the observed vertical distributions are produced by biological mixing after deposition. Most of those present did not feel that chemical migration within the sediments was an important mechanism for redistribution although we were aware that some investigators do feel it is an important factor. There is clear evidence that plutonium is returning from the sediments of the Enewetak and Bikini Atolls to the lagoon water, but this return seems explainable in terms of a simple exchange equilibrium between the surface sediments and the overlying water. No chemical migration of plutonium within the sediments is necessary to account for this return. The question of plutonium migration within sediments is critical to any prediction of the ultimate fate of plutonium in the oceans and our ability to produce trustworthy, long range models. This issue awaits resolution.

Chemical speciation of plutonium, in the water column, was another topic which received considerable attention. The oxidation state of plutonium has now been determined in samples collected around the British Isles, in the Mediterranean Sea, the north Pacific Ocean, and at the Pacific test sites. In each of these areas the oxidized form, probably Pu(V), is a major component. In samples of water from the open ocean about 50 percent of the dissolved plutonium is Pu(V) while in coastal water samples it is ∿90 percent. The oxidation state distribution at the depth of the plutonium maximum in the Pacific (∿500 m) is not substantially different from the distribution at other depths, indicating that a change in oxidation state is not the direct cause of the maximum. Although the measurements seem reproducible, there was no general agreemen as to the cause or long-term significance of the actual values.

Differences in speciation between plutonium and americium are evident from comparisons of the ^{241}Am to ^{239,240}Pu ratio in solution and on suspended particles, which is always greater than the ratio in solution, often by a factor of 10. This tendency is consistent with the observation that much of the dissolved plutonium is present as the weakly sorbed Pu(V) species. The ^{241}Am to ^{239,240}Pu ratio in sediments is often higher than that calculated from the inputs (or measured in terrestrial environments where losses have not occurred). This again is consistent with the generalization that plutonium is slightly more "soluble" than americium and that consequently the delivery of americium to the sediments is slightly more efficient than is the delivery of plutonium.

The only truly puzzling (i.e., not rationalizable) research discussed at the meeting concerned the accumulation of plutonium in fish from the Pacific test sites. It is generally assumed that the concentration of plutonium in an organism is directly related to the concentration in some part of its environment with the proportionality constant being the concentration factor. Recent measurements at Enewetak, Bikini, and some lesser contaminated atolls suggest that for some fish species this assumption is not adequate. No clear dependence of plutonium concentration in either water or sediment could be demonstrated for samples collected over a substantial range of environmental concentrations. The panel had no helpful suggestions as to the cause of this phenomenon.

It was apparent during the course of the meeting that most current marine research emphasizes the physical and chemical aspects of actinide transport. In general it appears that our understanding of these features, while still not complete, exceeds our understanding of the factors which govern bioaccumulation. With our increased capability to characterize the chemical species of plutonium present in solution, more realistic bioaccumulation experiments can now be conducted.

In order to more effectively describe and predict the behavior of transuranic elements in the marine environment, the marine panel suggests the following actions be taken to complement the current programs:

1. Apply existing capabilities in marine modelling to problems of artificial radionuclides.

2. Increase research into biokinetic behavior of transuranics with marine and freshwater organisms. This information is essential for more detailed models. As a corollary, there is an urgent need for a continuing source of ^{237}Pu, the most useful plutonium isotope for conducting such studies.

3. Expand mass spectrometric capability to ensure adequate sample throughput in light of the increased use this technique is certain to receive.

4. Document the latitudinal fallout input record of transuranics, with special emphasis on isotopic composition, at some mid-latitude sites using glacial samples (or rapidly sedimenting water bodies).

5. Organize periodic laboratory exercises to ensure the accuracy of the data being obtained (especially important for new techniques).

6. Recognize the continuing need to obtain open-ocean samples at a time when ship facilities are diminishing. The panel points out the problems without proposing solutions.

The panel feels that these measures are essential to the formulation and validation of better models and that for the present they should take priority over extensive new modelling efforts.

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