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**A REVIEW OF RESEARCH PROGRAMS RELATED TO THE
BEHAVIOR OF PLUTONIUM IN THE ENVIRONMENT**

Prepared for

**U.S. Department of Energy
Office of Special Nuclear Projects**

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1.0 INTRODUCTION

Plutonium-fueled radioisotopic heat sources find application in a spectrum of space, terrestrial, and underseas applications to generate electrical power by thermoelectric or dynamic-cycle conversion. The principal radioisotope used for this purpose is plutonium (Pu)-238 in the form of plutonium dioxide. Such systems under postulated accident conditions could release radioactivity into the environment resulting in risks to the general population. Radioactivity could be released at high and low altitudes, on the land surface, or underwater. The released radioactivity could then be dispersed into various environmental media (air, soil, water) and interact with people through various exposure pathways leading to inhalation, ingestion, and external radiological doses and associated health effects.

NUS Corporation has developed short-term exposure (RISK II) and long-term exposure (RISK III) models for use in safety risk assessments of space missions utilizing plutonium-fueled electric power systems.^(1,2,3) These models have been documented in the Overall Safety Manual.⁽²⁾ In order to effectively use these models in risk assessments, representative input values must be selected for a spectrum of environmental transfer parameters that characterize the behavior of plutonium in the environment. The selection of appropriate transfer parameters to be used in a given analysis will depend on the accident scenarios to be modeled and the terrestrial and aquatic environments to be encountered.

In support of the U.S. Department of Energy, Office of Special Nuclear Projects, NUS Corporation was authorized to review the availability of plutonium environmental data for use in risk assessments and the status of research programs being conducted by various organizations related to the behavior of plutonium in the environment. This work has been performed under Contract No. DE-AC01-80ET-32079, Task Assignment 82-8.

This report summarizes the research programs presently being conducted and makes recommendations on areas where further research is needed in order to fill gaps in the data necessary for risk assessments. Current research programs were reviewed primarily by visiting organizations and researchers who

are actually conducting the work. A description of the current research programs being conducted is summarized in Section 2. Conclusions of the study and recommendations for future research are presented in Section 3.

Plutonium environmental data for use in risk assessments have been summarized in a separate report by NUS Corporation as part of this task.⁽⁴⁾ The data summary will be included in a future update to the Overall Safety Manual.

2.0 REVIEW OF PLUTONIUM ENVIRONMENTAL RESEARCH PROGRAMS

Much research has been conducted over the past ten years on the behavior of plutonium in the environment. This research has been prompted by environmental and safety studies designed 1) to evaluate the effects of fallout following nuclear weapons testing in the atmosphere, 2) in support of the Liquid Metal Fast Breeder Reactor (LMFBR) program, 3) in support of plutonium production and handling facilities, and 4) in support of plutonium-fueled nuclear systems used in space, terrestrial, and underseas applications.

Principal organizations involved in research on plutonium in the environment including the following:

- o Argonne National Laboratory (ANL)
- o Pacific Northwest Laboratory (PNL)
- o Lawrence Livermore National Laboratory (LLNL)
- o Los Alamos National Laboratory (LANL)
- o Oak Ridge National Laboratory (ORNL)
- o Savannah River Ecology Laboratory (SREL)
- o U.S. DOE, Environmental Measurement Laboratory (EML)
- o U.S. DOE, Nevada Applied Ecology Group (NAEG)
- o U.S. DOE, Office of Special Nuclear Projects (OSNP)
- o U.S. DOE, Office of Health and Environmental Research (OHER)
- o U.S. Environmental Protection Agency (EPA)
- o Universities.

A primary focus point of plutonium environmental research is the Ecological Sciences Division of the DOE Office of Health and Environmental Research and its coordinated research efforts in support of the LMFBR program. These research areas, summarized in Figure 1, cover not only plutonium but all the transuranics. Principal OHER sponsored research is conducted by PNL, ANL, SREL, ORNL, and LANL. OHER has sponsored annual workshops over the past several years that deal with transuranics in the environment with special emphasis on plutonium.⁽⁵⁻⁹⁾ Due to the tremendous amount of plutonium environmental data collected to date, OHER sponsored the compilation and summary of this work, resulting in the publication of Transuranic Elements

Figure 1.- Plutonium Environmental Programs Sponsored by DOE's Office of Health and Environmental Research

in the Environment.⁽¹⁰⁾ The document is an important source of data for use in estimating environmental transfer parameters.

Research related to plutonium in the environment resulting from nuclear weapons testing is being conducted by EML, NAEG, and ANL.

Environmental research related to plutonium-fueled space nuclear systems, sponsored by the DOE Office of Special Nuclear Projects, is being conducted by LANL and the Naval Ocean Systems Center (NOSC). This research work is designed to evaluate the interaction of plutonium dioxide fuel with terrestrial and aquatic environments.

As part of the present task, visits were made to various institutions to meet with individuals who are conducting research programs related to the behavior of plutonium in the environment. A summary of these visits is presented below, with detailed trip reports presented in Appendix A.

2.1 Los Alamos National Laboratory

Los Alamos National Laboratory is conducting a program for the DOE Office of Special Nuclear Projects to evaluate the response of plutonium dioxide fuel forms to various terrestrial and aquatic environments. Experiments are conducted in controlled environmental chambers simulating humid and desert terrestrial environments with temperature and rain cycling. Aquatic experiments are conducted in aquariums to simulate both fresh water and sea water environments.

NUS visited LANL on August 24, 1982 to review the ongoing experiments being conducted by G. Matlack and his colleagues. The meeting focused on summarizing key results from all the data collected from the experiments in the past. These results include fuel dissolution rates in fresh water and sea water, airborne release rates under dry and rain conditions, and vertical transport rates in soils under percolating water conditions. These results have been summarized in the plutonium environmental data summary report prepared as part of Task Assignment 82-8.⁽⁴⁾

NUS again visited LANL on September 21-22, 1982 to attend a meeting reviewing DOE/OSNP sponsored environmental programs. These programs include not only the one at LANL, but the orange tree experiment being conducted by J. Pinder of SREL, and the ocean experiments being conducted by H. Weiss of NOSC at San Clemente Island, California. The orange tree experiment is designed to evaluate the uptake of airborne deposited plutonium dioxide particulates on orange trees in the Cape Canaveral region following a launch pad abort. The experiments at NOSC are designed to evaluate the response of plutonium dioxide fuel forms to actual ocean conditions. During the meeting, NUS made a presentation on risk assessment methods for plutonium-fueled space nuclear systems and described the Overall Safety Manual.

Due to a reorganization at LANL and new direction from DOE/OSNP, the LANL environmental program was re-oriented from one designed to yield information on relative response of various fuel forms to terrestrial and aquatic environments to one designed to yield quantitative information on environmental transfer parameters for use in modeling. In support of this re-orientation, NUS again visited LANL on November 17, 1982 to assist LANL in developing an experimental plan for the new program. Meeting efforts centered on establishing qualitative and quantitative objectives of each experiment to ensure that the results would be useful in risk assessments.

2.2 Lawrence Livermore National Laboratory

NUS visited LLNL on August 26, 1982 to discuss the research being conducted by V. Noshkin and his colleagues at the Bikini and Enewetak Atolls in the South Pacific related to transuranic contamination associated with the nuclear weapons testing program. This work which spans several years is designed to monitor transuranic radioactivity in the marine environment at Bikini and Enewetak, including sediments, water, and marine life. Distribution patterns and transport mechanisms have been established and concentration factors determined for various kinds of marine life.

V. Noshkin was also part of a study designed to evaluate the contamination of the marine environment around San Clemente Island due to the program at NOSC

that placed plutonium dioxide fuel forms on the ocean floor to determine their response.

V. Noshkin, reviewed data on some rocket aborts over the Johnson Islands that resulted in the dispersal of plutonium during the weapons testing program. These data would be very useful to review because of the similarity of the abort scenarios with potential launch abort accidents at Cape Canaveral.

2.3 Savannah River Ecology Laboratory

NUS visited SREL on August 31, 1982 to discuss research being conducted by J. Pinder and other persons from SREL and the Savannah River Laboratory (SRL) related to the behavior of transuranics in terrestrial and aquatic ecosystems at the Savannah River Plant (SRP). Liquid and airborne transuranic source terms are generated at SRP as part of the operation of weapons-grade material production reactors and separation facilities. An extensive environmental program has been established to evaluate environmental concentrations of transuranics, transport mechanisms, and uptake in the varied terrestrial and aquatic ecosystems at SRP.

J. Pinder described the orange tree experiment being conducted for DOE/OSNP as discussed previously.

2.4 Oak Ridge National Laboratory

NUS visited ORNL on March 8, 1983 to review the work being performed by C. Garten and E. Bondietti. C. Garten has been conducting research programs on the behavior of transuranics in terrestrial ecosystems at ORNL resulting from contamination due to operations on the Oak Ridge Reservation. The terrestrial ecosystems studied include a floodplain and a deciduous forest. C. Garten's study on the behavior of plutonium in a deciduous forest is unique in that it integrates field measurements into a simulation model, thus providing a comprehensive description of the behavior of plutonium in a complex ecosystem.

E. Bondietti's research has focused primarily on the behavior of transuranics in aquatic systems. This has included trying to understand the distribution

of plutonium in aquatic systems in terms of the various oxidation states of plutonium.

2.5 Argonne National Laboratory

NUS visited Argonne National Laboratory on March 9, 1983 and met with D. Nelson and R. Larsen who have been conducting research programs on the behavior of transuranics in natural aquatic systems. This work in the past has included extensive evaluations of transuranics in Lake Michigan and other fresh water North American lakes in both the United States and Canada. More recently their work has focused on trying to understand the chemistry of transuranics in aquatic systems in terms of the various oxidation states of a given radionuclide. Much of this work has been directly related to plutonium. D. Nelson has also been supporting research programs at other institutions related to transuranics in the marine environment by providing analytical radiochemistry services. It should be noted that in all of these programs the transuranics, including plutonium, were derived from nuclear weapons testing fallout.

In addition to the above studies, R. Larsen has been doing research on the internal dosimetry of ingested transuranics. Specifically, he has examined oxidation state changes that occur in compounds of transuranic radionuclides as a result of water treatment and root/leaf uptake. He has found that these changes in oxidation state result in a much higher transfer of plutonium from the small intestine to the blood than had been previously assumed.

2.6 Pacific Northwest Laboratory

NUS visited Pacific Northwest Laboratory on March 10-11, 1983 to meet with O. Moss, R. Wildung, G. Sehmel, and others.

O. Moss has been conducting research characterizing the behavior of radioactive particulates and aerosols. These studies have been primarily in support of inhalation toxicology studies in which characterization and generation of specific particle size distributions are necessary. Three primary tools/techniques O. Moss utilizes with these studies include the electron

microscope, a wind tunnel, and the BET method. The BET method is used to determine surface areas of particles by depositing a thin film of liquid nitrogen on the particle surface. Using these techniques the particle shape and size distributions can be accurately characterized. A new wind tunnel will be used in resuspension and deposition studies using radioactive particles.

R. Wildung and his colleagues have been conducting research programs related to the behavior of transuranics, including plutonium, in terrestrial ecosystems. These studies have focused primarily on plant uptake using both greenhouse plants and field plots.

G. Sehmel has been conducting extensive research on resuspension. This has involved field studies at Hanford, the Nevada Test Site, and other locations where transuranic contamination of the ground has occurred in the past.

Other studies being conducted at PNL that are applicable to risk assessment of space nuclear systems include: 1) animal inhalation studies, 2) development of risk assessment models, and 3) development of statistical methods for the evaluation of environmental data and design of experiments.

2.7 Overall Plutonium Research Status

The overall status of plutonium environmental research programs with respect to specific data requirements for 1) risk assessment modeling of plutonium-fueled space nuclear systems and 2) recovery and decontamination efforts following postulated accidents are summarized in Table 1.

2

Status Summary of

Data Category	Parameter
<u>Terrestrial Ecosystems</u>	Airborne and leaching/ Runoff Source Terms

Inventory Ratios^a

2-8

^a. The proportion of the total plutonium in the ecosystem

Table 1 (Page 1 of 5)

Plutonium Environmental Research and Data Needs

Research/ Data Source	Status	Recommended Research Priority
NL	Airborne and leaching/runoff source terms due to PuO_2 chunks have been characterized under humid and arid conditions in controlled environmental chambers. More refined mass balance experiments are needed in order to characterize these source terms as a function of particle size. Since postulated source terms of most interest with respect to accident risk are dispersed particulates resulting from initial impact, explosion, or fire, the continuing need for experiments involving chunks should be re-considered.	Low
NL (Hakonson), ORNL	Inventory ratios have been developed for a contaminated floodplain forest at ORNL and Mortandad Canyon at LANL. Potentially, such information could be developed by SREL for ecosystems at SRP based on available environmental data collected to date that would be more applicable to ecosystems at Cape Canaveral. Finally, inventory ratios could be developed for actual Cape Canaveral ecosystems, but the lack of contaminated ecosystems except fallout precludes this. However, the evaluation of inventory ratios for fallout plutonium should be considered. The other option is to perform experiments in LANL controlled environmental chambers simulating Cape Canaveral terrestrial ecosystems. However, fine particulate PuO_2 should be used that is uniformly deposited to duplicate that expected from postulated accidents. These kinds of experiments are currently being considered by LANL.	High

to that found in each major compartment

Data Category	Parameter	
<u>Terrestrial Ecosystems</u> (Continued)	Concentration Ratio ^a (Field Grown Crops)	O R
	Concentration Ratio ^b (Animals)	O L S
	Watershed Erosion Rate	S

2-9

- a. The proportion of the total plutonium in the ecosystem
b. Ratio of concentration in plants (animals) to concentration in soil

Table 1 (Page 2 of 5)

Plutonium Environmental Research and Data Needs

Research/ Data Source	Status	Recommended Research Priority
NL, SRP, PNL, NTS, Rocky Flats, Enewetak	Concentration ratios are available for field grown crops, vegetables, and native vegetation at the indicated locations. J. Pinder at SREL is doing a comprehensive experiment to determine critical parameters for contaminated orange trees. Additional experiments should be undertaken for other economically important crops (principally grass for cattle grazing) in the Cape Canaveral region in a similar manner to that of the SREL orange tree experiments. Determination of inventory ratios for Cape Canaveral ecosystems described previously would also yield concentration ratio data.	High
NL, Rocky Flats, NL (Hakonson), REL	Concentration ratios are available for small mammals in a floodplain forest, a canyon ecosystem, and a grassland ecosystem. Additional data has been collected by SREL at SRP that needs to be pulled together. The latter data are somewhat applicable to Cape Canaveral ecosystems since they relate to game animals. However, impact on the cattle industry would be of more concern at Cape Canaveral.	Medium
L, Simpson, Mound	Watershed erosion rates for fallout plutonium have been estimated for the Savannah, Miami, Newport, and Hudson River watersheds. Due to the flat terrain in the Cape Canaveral region, the watershed erosion rate is expected to be small. Furthermore, most water would seep downward in the porous soil or evaporate. Data from the USDA Soil Conservation Service could be used to estimate soil erosional losses for given watersheds, allowing order of magnitude estimates to be made.	Low

to that found in each major compartment
ation in soil

2

Status Summary of

Data Category	Parameter	
<u>Terrestrial Ecosystems</u> (Continued)	Soil Leaching Rate	L
	Resuspension Factor	N F
<u>Aquatic Ecosystems</u>	Dissolution Rates	L

2-10

Table 1 (Page 3 of 5)

Plutonium Environmental Research and Data Needs

Research/ Data Source	Status	Recommended Research Priority
LANL, HASL	The downward movement rate of plutonium in soils is being evaluated at LANL in controlled environmental chambers under humid and arid conditions for different soil types, and in soil column experiments. Improved experiments characterizing downward transport will continue. Generally, the results indicate the downward movement rate is very slow. However, more attention should be given to short term transport in order for support cleanup activities. Short term in this case means what happens after each subsequent rain following an accident involving dispersed particulates.	Medium
NTS, Hanford, Rocky Flats, SRP, LLNL	Much resuspension factor data are available for the NTS, Hanford, Rocky Flats and SRP sites. Some limited data are available for the shoreline environment at Bikini. The SRP and Bikini data are somewhat applicable to the Cape Canaveral region. Resuspension studies in the Cape Canaveral region would be desirable, although these studies would have to be done with non-radioactive tracers. Sehmel of PNL questions a set of NTS resuspension data used in most models that indicate a decrease in the resuspension factor from 10^{-4} to 10^{-9} in two years. Sehmel believes that subsequent data do not support this kind of decrease. Further evaluation of this problem is needed.	Medium
LANL, NOSC	Dissolution rates of bare PuO_2 fuel forms have been measured by LANL in warm and cold fresh water and sea water in controlled aquariums. Future experiments are designed to account for removable mechanisms in the aquatic system that	High

2

Data Category	Parameter
<u>Aquatic Ecosystems</u> (Continued)	

2-11

K_d^c

c. Ratio of concentration in sediment to concentration i

Table 1 (Page 4 of 5)

Plutonium Environmental Research and Data Needs

Research/ Data Source	Status	Recommended Research Priority
	<p>will lead to improved estimates of dissolution rates. In addition, dissolution mechanisms and the effects of chemical and physical parameters of the water will be evaluated. These experiments are being planned by LANL.</p> <p>No data are available on iridium corrosion rates which are needed to determine the time frame required for encapsulated fuel to be more exposed to dissolution mechanisms. LANL should consider performing accelerated iridium corrosion rate experiments in sea water and fresh water to provide these data. In conjunction with this research, the fuel forms exposed to actual ocean environments at NOSC should be analyzed to determine the nature of encrustations and the degree to which iridium encapsulation is degraded or enhanced.</p>	
<p>LANL, ORNL, LLNL, SREL</p>	<p>The distribution of plutonium in aquatic systems between the water and sediment has been evaluated in numerous fresh water and marine environments. The speciation and oxidation states of plutonium, along with controlling chemical and physical parameters have also been characterized. However, data specific to Cape Canaveral aquatic ecosystems are lacking. G.R. Choppin of Florida State University has been conducting studies of the behavior of fallout plutonium in some Florida lakes. Perhaps this research effort could be extended to include the Cape Canaveral region. The primary orientation of such experiments should be directed at impacts on the Florida fishing industry.</p>	<p>Medium</p>

in water

7

Data Category	Parameter
<u>Aquatic Ecosystems</u> (Continued)	Concentration Ratios
	Particulate Retention Factor ^d and Residence Half-Life ^e

2-12

d. Fraction of PuO₂ particulates deposited on a water surface
e. Time required for one-half of the surface retained particulates to be removed

Table 1 (Page 5 of 5)

Plutonium Environmental Research and Data Needs

Research/ Data Source	Status	Recommended Research Priority
LANL, SREL	Concentration ratios for fresh water and marine life have been characterized for a number of locations in the Atlantic and Pacific Oceans, and fresh water lakes. The applicability of these data to the Cape Canaveral aquatic ecosystems needs further evaluation. Site-specific data may be needed in order to evaluate impacts of accidents on the Florida fishing industry.	Medium
US, LANL	NUS analyses indicated that PuO ₂ particles deposited on a water surface could be retained on the water surface due to surface tension effects. This effect could significantly alter risks and cleanup strategies. This phenomenon would be a function of particle size and the surface tension of water with respect to PuO ₂ . Experiments are needed to confirm or deny the existence of this phenomena. LANL is considering the feasibility of making such surface tension measurements. Experiments in which PuO ₂ particles of known size are deposited on a water surface and viewed optically in a sealed chamber would also be useful.	Very High
face that is retained on the surface due to surface tension effects iculates to penetrate the surface		

3.0 CONCLUSIONS AND RECOMMENDATIONS

Conclusions and recommendations of this study, based on a review of the literature and existing research programs related to the behavior of plutonium in the environment, are presented below. A summary of plutonium environmental data for use in risk assessments of plutonium-fueled space nuclear systems was prepared as part of this task and is presented in a separate report.

3.1 Availability of Plutonium Environmental Data

In evaluating the quantity and quality of plutonium environmental data available, a distinction must be made between mechanisms affecting the transport and distribution of plutonium in the environment. The general environment can be described as environmental compartments, such as air, water (aquatic ecosystems), land (terrestrial ecosystems), and people. Modeling in support of risk assessments attempts to describe the distribution of released plutonium within a given compartment and its transport in time from compartment to compartment.

Air and water transport mechanisms, weathering, resuspension, and runoff move plutonium from one compartment to another in a manner that is somewhat independent of the nature of plutonium. The plutonium environmental data and research addressed in this report relates to how plutonium is initially released within a given compartment and how the released plutonium redistributes itself within that compartment.

In this context the results of this study indicate there is a tremendous amount of data available in the literature on the behavior of plutonium in the general environment and various types of terrestrial and aquatic ecosystems. There is so much available that much effort should be devoted to reviewing, screening, and digesting the data and information already available. Accordingly, NUS recommends that currently available data be thoroughly evaluated by researchers prior to conducting experiments to ensure themselves that no duplication of efforts occurs and useful contributions will be made.

The research program at LANL fulfills the need to characterize the initial release mechanisms in various types of ecosystems. Research programs at SREL, ORNL, ANL, and PNL are designed to characterize how released plutonium redistributes itself in a given ecosystem. Future research needs in support of risk assessments of plutonium-fueled space nuclear systems should focus on the specific ecosystems to be encountered in the Cape Canaveral region. In this regard, it should be noted that all research to date focuses on the redistribution of plutonium within a small narrowly defined ecosystem. However, emphasis in modeling in support of risk assessments is placed on mass transport mechanisms and the relative distribution of plutonium among various compartments (ecosystems). In order to do this modeling detailed information is needed on the distribution of the compartments (ecosystems) themselves within the region, requiring a careful land use and demographic survey.

3.2 Recommendations for Research

Recommendations for further research activities are presented below.

1. Research priorities designed to satisfy data needs for risk assessments and post-accident recovery and decontamination operations established in Table 1 of Section 2.0 should be considered in the design of new experiments.
2. The program at LANL is well designed to characterize initial release mechanisms in various ecosystems. However, modifications should be considered in light of the research priorities identified in Table 1 of Section 2.0. For those experiments that are conducted, fuel configurations actually expected for given accident scenarios and environments should be used (e.g., dispersed particulates for terrestrial experiments, and aeroshell modules for sea water experiments). It is recommended that LANL establish further contact with D. Nelson and R. Larsen at ANL to draw upon their expertise on the radiochemistry of plutonium in aquatic systems and O. Moss of PNL to draw upon his expertise with regard to characterization and generation of plutonium dioxide particle size distributions. LANL is developing detailed experimental plans in accordance

with DOE/OSNP guidance letters of May 1983. NUS should continue to work closely with LANL in support of the experimental program.

3. The orange tree experiment being conducted by J. Pinder of SREL should be extended to include other economically important crops in the Cape Canaveral Region, primarily grass used for cattle grazing.
4. Efforts should continue by NUS in characterizing the Cape Canaveral regional environment in terms of demographic, land use, water, and crop distributions. This information is as important for modeling purposes as the plutonium environmental data itself.
5. NUS strongly recommends that further research be performed to assess the possibility that the surface tension of water might be such that plutonium dioxide particles could be retained on the surface. The implications of such a phenomenon include:

- o Water bodies could be decontaminated by skimming the surface
- o Increased hazards occur along shorelines where the particulates tend to accumulate by wave action
- o Surface feeding fish will have higher concentrations than what would normally be expected
- o Sea spray would be substantially enriched in plutonium in near shore environments.

In support of this effort, LANL has plans to evaluate the surface tension of water with respect to plutonium dioxide.

6. No data are available on iridium corrosion rates which are needed to determine the time frame required for encapsulated fuel to be more exposed to dissolution mechanisms in aquatic environments. It is recommended that LANL consider performing accelerated iridium corrosion rate experiments in sea water to provide these data. In conjunction

with this work, the fuel forms exposed to actual ocean environments at NOSC should be analyzed to determine the nature of carbonate encrustations, under what conditions they form, and the degree to which encapsulation is degraded or enhanced.

3.3 Experimental Design

Experiments designed to characterize the behavior of Pu-238 fuel forms in aquatic and terrestrial environments should have clearly defined qualitative and quantitative objectives. Such experiments should reflect realistic terrestrial and aquatic environmental conditions to the extent possible while maintaining control over carefully defined physical and chemical parameters. This can be facilitated by taking a systems approach to each experiment in which an underlying quantitative objective should be to obtain a total mass balance of Pu-238 as a function of time in each element of the environmental system. The results of each experiment should be directed towards understanding the physical and chemical mechanisms affecting the transfer of Pu-238 from one element of the system to another. Exponential box models are often useful in understanding the behavior of such systems, in which mass transfer rates from one element of the system to another can be represented by exponential transfer parameters and supporting system equations. The objectives of the experiment then reduce to quantifying the transfer parameters and understanding the underlying transfer mechanisms. Furthermore, the results of every experiment should be compared with results published in the literature on plutonium in environmental systems in general. This would also include statements as to how the data would be used in 1) risk assessment models, and 2) post-accident recovery and decontamination operations. It is recognized that the above represents an idealized approach, and not always achievable in such experiments as those under consideration. A comprehensive experimental plan should be prepared that addresses all these items. An example of the type of approach recommended, applied to the design of aquatic experiments, is presented in Appendix B.

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APPENDIX A

Trip Reports on Meetings Related to the Behavior of Plutonium in the Environment

810 CLOPPER ROAD
GATHERSBURG, MARYLAND 20878
(301) 258-8000

MEMORANDUM

TO: Distribution GEC-82-325-RAD
October 5, 1982

FROM: M. Wilkinson *myw*

SUBJECT: Trip Report on Meeting at Lawrence Livermore National Laboratories
for Environmental Data Support Task

This trip report summarizes a meeting on August 28, 1982 at the Lawrence Livermore National Laboratories (LLNL) that was conducted as part of the DOE Environmental Data Support Task. Present at the meeting were:

- c V. Noshkin (LLNL)
- c L. Wong (LLNL)
- c E. Bertrac (NUS)
- c M. Wilkinsor (NUS)

The specific purpose of the trip was to obtain information requested in GEC-82-277-RAD dated August 17, 1982 from NUS to V. Noshkin. LLNL has conducted radiological monitoring programs at Bikini and Enewetak Atolls, related to transuranic contamination associated with prior nuclear weapons testing programs, and at San Clemente Island, related to contamination resulting from testing the response of plutonium dioxide fuel forms to the marine environment. Information obtained during the meeting is presented below.

Plutonium from weapons tests in the Pacific Ocean is generally more associated with sediments than is dissolved in water. It is primarily the reduced state of plutonium that is associated with sediment; the oxidized state is primarily present in water. Plutonium will mobilize out of sediments into the water if it becomes oxidized. It has also been determined that americium is only present in sediments; it does not oxidize and mobilize into the water.

The distribution coefficient, K_d , is the ratio of material present in sediment to material present in water. When measured in the Marshall Islands, K_d agrees within 50% of the K_d measured at San Clemente. In fact, this parameter has been found to be approximately 10^5 for plutonium throughout the world. If all plutonium was in the reduced state, K_d would be about 10^6 ; K_d would be about 10^3 if all plutonium was in the oxidized state. It has also been determined that K_d decreases as the concentrations of dissolved organic carbon in water goes up.

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No differences in animal uptake have been observed between oxidized and reduced plutonium. Most animal uptake is through ingestion of sediments. Plutonium remains primarily in the gut. Muscle contents are lower by about 10^{-4} . Since muscle is the edible portion of fish, some care should be taken when scrutinizing concentrations of transuranics in fish. Results are often quoted that include gut contents. In the Marshall Islands, it has also been determined that there is decreased uptake of americium in fish, as compared to plutonium. The opposite situation, i.e., higher americium uptake, has been found at Windscale.

In the Enewetak Atoll, the soluble and insoluble states of plutonium, oxidized and reduced, respectively, appear to have come to a steady state. Predictions are that the sediment inventory of the lagoon will be remobilized into solution and transported out towards the Northern Pacific with a half-life of 400 years.

Seaspray on the windward side of the atoll was found to contain 20 times the concentration of plutonium of sea water. Levels of algae and other organic materials were also found to be enriched. It is thought that the plutonium enrichment may be due to complexing with organics.

In regards to the aborted rocket launches at the Johnston Islands, it was agreed that the scenario of the shuttle exploding on the launch pad would be quite similar to actual incidents at Johnston. Apparently drainage water in the pad area contains about 200 pCi/gm of plutonium. NUS will do further checking on these incidents to see whether source terms can be determined.

The meeting was quite successful; a great deal of information and knowledge was exchanged on both sides. NUS obtained from Noshkin some other leads on additional places to search for information on the behavior of plutonium in the environment. Noshkin was reluctant to give values for transfer rates between various environmental compartments because of uncertainties inherent in any such numbers.

/ds

Distribution: R.W. Englehart
B.W. Bartram

810 CLOPPER ROAD
GAITHERSBURG, MARYLAND 20878
(301) 298-8000

MEMORANDUM

TO: Distribution

GEC-82-326-RAD
October 5, 1982

FROM: M. Wilkinson *my*

SUBJECT: Trip Report on Meeting at Los Alamos National Laboratory for
Environmental Data Support Task

This trip report summarizes a meeting held at the Los Alamos National Laboratory (LANL) as part of the DOE Environmental Data Support Task. Attendees included:

o	G. Matlack	(LANL)
o	S. Bronisy	(LANL)
o	J. Patterson	(LANL)
o	N. Shalmaker	(LANL)
o	M. Wilkinson	(NUS)
o	B. Bartram	(NUS)

The purpose of the meeting was to obtain information requested in GEC-82-277-R41, dated August 17, 1982, from NUS to LANL. Discussions centered on reducing experimental data already collected into a form that could be used to characterize plutonium source terms in terrestrial and aquatic environments. Information obtained in the meeting is summarized below.

1. Aquatic Experiments

No significant differences in aquatic dissolution rates were found between bare PPG and GROS fuel. The highest release rates continued to be in cold (10°C) fresh water. The release rates are presented in Table 1. Release rates are determined every quarter, and appear to remain the same, 100%. No investigations of the reasons for differences in release rates have been conducted or are planned.

Vent release rates for encapsulated fuel in both deep and shallow ocean, as determined in laboratory experiments, were essentially zero. Some attack was present along the weld, and some plutonium was released as a result of chlorine attacking the clad.

2. Terrestrial Experiments

Airborne release rates in humid climates were determined in a series of five experiments in which plutonium concentrations were measured before, during, and after rain. Four experiments used fuel chunks and one used fines. The duration of rain was five minutes. In the experiments with fuel chunks, the airborne concentration was a few pCi/m³ prior to the rain. Within five minutes after the beginning of the rain, the airborne concentration had increased by a factor of 10⁴. Within one to two hours after the rain, however, the airborne concentrations had returned to pre-rain levels.

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In the experiment using fuel fines, there was no increase in airborne concentration as a result of the rain. By the time the rain had stopped, the airborne concentration was a factor of 10 lower than at the onset of the rain.

For this experiment, an assumption was made that, for chunks, the spallation rate is greatest at the onset of rain and decreases linearly to zero in 10 seconds. There was no spallation with fines. Source term data based on these experiments are summarized in Table II.

The only information available for dry climates was the airborne concentration of plutonium in the environmental chamber containing fuel chunks.

Soil leaching was evaluated under two different experimental conditions. One experiment placed paraticulate and ionic plutonium sources on the top of soil columns. The columns are saturated with water and the eluent analyzed for plutonium. In these set-ups, 99% of the plutonium remained at the top of the column. Other experiments evaluated the distribution of plutonium in core samples taken from environmental chambers. In these chambers, water percolates through the soil following "rain". Here, there is an irregular distribution of plutonium down to a depth of about 30 cm. The plutonium in the percolated water was very fine, with a diameter of 0.2-0.3 μ .

Water runoff source terms have not been evaluated by Matlack's group. We were referred to Hakonson's (LANL) work.

The effects of Cape Canaveral soil on fuel release rates has not been evaluated yet. Matlack is waiting for more fuel from SRP.

There has been no chemical characterization of soil, water, and rain used in these experiments. The only parameter quantified has been the conductivity of the water used for rain. Distilled water from taps in the laboratory is used for rain.

/ds

Distribution: R.W. Englehart
B.W. Bartram

TABLE 1
Plutonium Release Rates In Aquatic Environments

<u>Sample Type</u>	<u>Power (W)</u>	<u>Water</u>	<u>°C</u>	<u>Release (nCi/m²-s)</u>
PPO	15	Sea	10	1.2
GROG	15	Sea	10	120
GROG	15	Fresh	10	170
GROG	15	Fresh	37	88
GROG	15	Sea	37	0.2

TABLE II

Airborne Source Terms For Bare Fuel in Humid Conditions
Under Rain and No-Rain Conditions

<u>Fuel Type</u>	<u>Concentration Before Rain nCi/g-s</u>	<u>Spallation Rate nCi/g in 10 Seconds</u>	<u>Concentration After Rain nCi/g-s</u>
Chunks, > 6mm	2×10^{-8}	0.9	5×10^{-9}
Fines	4×10^{-8}	0	3×10^{-9}

MEMORANDUM

TO: Distribution

GEC-82-330-RAD
October 5, 1982

FROM: M. Wilkinson *mjw*

SUBJECT: Trip Report on Meeting at Savannah River Ecology Laboratory for
Environmental Data Support Task

This memorandum summarizes a meeting at Savannah River Ecology Laboratory (SREL) on September 1, 1982 to discuss research on the behavior of transuranics in the environment. Present at the meeting were:

- o J. Pinder (SREL)
- o D. Adriano (SREL)
- o D. Gay (SRL)
- o D. Hayes (SRL)
- o J. Alberts (SREL)
- o C. King (SRL)
- o M. Wilkinson (NUS)
- o B. Bartram (NUS)

The purpose of the meeting was to obtain information requested in GEC-82-281-RAD, dated August 17, 1982. Data obtained during the meeting are presented below; each researcher discussed his own project(s).

J. Pinder

J. Pinder conducts research on the behavior of crops grown in fields adjacent to the Savannah River reprocessing facilities. There are two different source terms seen in the crops: Pu-238 from the Savannah River Plant (SRP) and Pu-239/240 from global fallout. Pinder has examined deposition, uptake, and resuspension in a variety of crops. The main source of crop contamination was direct deposition, with some resuspension, and almost negligible root uptake.

D. Adriano

D. Adriano is involved in experiments with J. Pinder. In addition, in greenhouse studies, he analyzed root uptake in crops using soils from H area. No differences were found in the behavior of Pu-238 as compared to Pu-239/240.

D. Hayes

D. Hayes studied the behavior of transuranics in two different rivers: the Savannah River and the Newport River, in North Carolina. Inventory of plutonium has been determined to be 55 Ci from fallout (Pu-238) and 5 Ci from the operation of the SRP (Pu-239/240). In the Savannah River, about 10% of the plutonium inventory results from SRP. Above and below the plant there were no significant differences in the amount of Pu-238 as compared to Pu-239/240: about 10% of

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the plutonium inventory is Pu-238, with the rest from fallout. Concentration of plutonium in the Savannah River, including sediments, is 0.2 to 0.3 fCi/l. The removal rate of plutonium from the river is about 0.01% per year.

In the Newport River, which contains coastal waters, plutonium from fallout appears to be more mobile than in the Savannah River. Presumably this is due to the increased amount of organics in coastal plain waters. The floodplains of both rivers are quite different. Differences in hydrology are important in determining the behavior of plutonium in the aquatic environment.

D. Gay

D. Gay has studied the relationships of particle size to deposition and resuspension. He has determined that the majority of Pu-238 released at SRP is less than 0.5 μ in diameter. A trapping system has been devised to catch particles as small as 0.3 μ . Gay has also determined that most radioactivity is associated with the less than 0.5 μ particles.

Resuspension rates were determined in two fields. Field 1 was mostly clay and had a resuspension rate of 1.7×10^{-10} /m. Field 2 was very sandy and farther from the stack; the resuspension rate there was 4.7×10^{-10} /m.

C. King

C. King has been evaluating the effect of another plutonium source term. This source term results from shallow burial of low level waste in trenches 20 feet deep, 25 feet above the ground water. The total inventory buried between 1955 and 1968 was 500 Ci of Pu-239 and 2500 Ci of Pu-238. Burial ceased in 1968 as a result of Federal mandates. The ground water and soil in the burial area have been monitored for transuranics, tritium, and fission products.

Beginning about 1968, a compartmentalized model was initiated to predict movement of the radioactivity. The model uses partial differential equations and a number of experimentally or theoretically determined transfer coefficients.

An upper dose limit of 4-5 person mrem per year has been calculated using the model. The dose calculated assumes that crops grown and harvested on the contaminated soil would be ingested and that the site would be used for residences.

The model has four subroutines. One calculates hydrological transport of the radioactivity. A second subroutine computes doses that result from a five-vector food chain using methodology from Regulatory Guide 1.109. The final two subroutines calculate doses that would result from subsistence or commercial farming on the site.

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J. Alberts

J. Alberts has examined the factors that control partitioning between sediments and organisms in Florida rivers. Dissolved organics and sediment characteristics control partitioning. Alberts noted that at Cape Canaveral the watershed area is sandy with no clay, and that water there is low in organics. In searching the literature for information on the possible fate of plutonium in the Cape Canaveral area, NUS was cautioned to examine data from watersheds of similar characteristics.

Generally, plutonium and other metals are more soluble in acid pH. As pH increases to neutral, metals precipitate out. Biota uptake is greatest when plutonium is in a soluble state.

The area around the Cape has citrus groves which are heavily fertilized. The runoff from these groves will affect the chemical character of rivers in the area. Additionally, the sediment character affects the distribution of plutonium. With clay sediments, 99% of the plutonium in the river is associated with sediments. In sandy sediments or sediments containing carbonates, however, plutonium is generally not associated with sediments.

/ds

Distribution: R.W. Englehart
B.W. Bartram

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MEMORANDUM

GEC-82-337-RAD
October 11, 1982

TO: Distribution
FROM: B. W. Bartram **BWB**
SUBJECT: DOE/OSNP Environmental Programs
Review Meeting

CC: R. Englehart
M. Wilkinson
File: 2818-06

NUS attended a review meeting at Los Alamos National Laboratory (LANL) September 21-22, 1982 on the plutonium environmental programs sponsored by DOE's Office of Space Nuclear Projects (OSNP). The meeting agenda and a list of the attendees are attached. A summary of the meeting is presented below.

1. S. Bronisz, LANL Program Manager for plutonium related DOE/OSNP work, made introductory comments to the meeting.
2. P. Cunningham, head of the newly formed LANL Analytical Chemistry Division (CMB-1), described a recent reorganization within LANL affecting the work related to the DOE/OSNP environmental program. The newly formed Analytical Chemistry Division consists of an Environmental Chemistry Section under P. Wagner, and a Radiochemistry Section under G. Matlack. The LANL plutonium environmental program will now be conducted under P. Wagner.
3. G. Bennett provided an overview of DOE/OSNP programs related to the development, fabrication, testing, and deployment of plutonium-fueled and fission reactor space nuclear electric power systems.
4. G. Matlack provided an overview of the DOE/OSNP sponsored program at LANL related to the behavior of plutonium dioxide fuel forms in terrestrial and aquatic environments, and described the aquatic experiments in detail.

The dissolution rates of plutonium dioxide fuel (PPO) determined by aquarium experiments are as follows:

<u>Fuel Wattage</u>	<u>Water Type</u>	<u>Temperature, °C</u>	<u>Dissolution Rate, nCi/m²-s</u>
2.5	Fresh Water	10	180
"	Sea Water	10	6.3
"	Seawater	37	2.7
25	Fresh Water	10	330
"	Seawater	10	11
"	Seawater	29	1.6

The dissolution rate in cold fresh water is observed to be consistently higher than in other aquatic environments. The reason for this is not known, although G. Matlack speculated that it might be due to the formation of a less soluble plutonium hydroxide form on the fuel surface in conditions other than cold fresh water. Other observations included 1) a removal half-life of 40 hr for a soluble form of plutonium in seawater due to plate-out, 2) 30-50% of plutonium in the dissolution experiments plated out on the aquarium sides, and 3) the released plutonium is in the form of small particles rather than ions.

LANL has not made any correlating measurements of chemical parameters in the aquatic experiments. I questioned whether the increased dissolution rates in cold fresh water was due to a true increase in dissolution rate or perhaps a decrease in removal rate by surface plating. In order to obtain a true picture both the dissolution and removal rates should be characterized.

In the limited experiments conducted to date using GROG fuel (used in GPFS) a severe crumbling of the fuel was noted in the tidal experiments, probably resulting from thermal shock. The dissolution rate of GROG fuel is higher than PPO in seawater.

G. Matlack noted that internal dosimetry models for plutonium ingestion assume a transfer fraction from the small intestines to the blood, f_1 , of 3×10^{-5} . The plutonium is assumed to be in a reduced state (+3, +4 valence). However, R. Larson of Argonne National Laboratory has observed that f_1 , could be as high as 10^{-3} for oxidized plutonium (+5, +6 valence) such as would occur in chlorinated water supplies.

5. N. Stalnaker described chemical, physical and physical/chemical mechanisms that may be controlling the dissolution of plutonium in aquatic systems.
6. J. Patterson described the terrestrial experiments conducted by placing plutonium fuel forms on various types of soil in controlled environmental chambers.
6. H. Weiss described the experiments being conducted at the Naval Ocean Systems Center consisting of placing plutonium fuel forms on the floor of the Pacific Ocean off San Clemente Island and observing the effects of seawater. The experiments conducted with bare plutonium showed the following:
 - o All fuel tested in seawater released plutonium during exposure.
 - o Some dissolved plutonium remained in test chambers; some, either in solution or associated with suspended material, was lost to the environment.
 - o Released plutonium was transported to a considerable distance.
 - o In the area monitoring, 0.18 nCi of Pu-238 above 0.10 nCi from fallout can be accounted for by assuming a 1 ng/g per day dissolution rate.

Tests are now being conducted with only encapsulated fuel. Considerable encrustation and sinking into the sediment have been observed.

7. B. Bartram described the methodology utilized by NUS in the risk analysis of space nuclear electric power systems as contained in the Overall

Safety Manual. The requirements for environmental data in support of these analyses were described and recommendations were made for utilizing a uniform data cataloging system. The need for good experimental design and a literature search prior to conducting experiments was emphasized.

8. G. Bennett described the experiments being conducted by J. Pinder of the Savannah River Ecology Laboratory related to the contamination of dispersed Pu-238.

After the discussions were concluded LANL personnel conducted a tour of the laboratories where the aquatic and terrestrial experiments are being conducted.

List of Attendees
DOE/OSNP Environmental Programs Review Meeting
Los Alamos National Laboratory
September 21-22, 1982

<u>Name</u>	<u>Organization</u>	<u>Telephone</u>
Karl Summer	DOE/OSNP	FTS-233-3992
George Ogburn	DOE/OSNP	FTS-233-3234
Charles Osterberg	DOE/OHER	FTS-233-3035
Herbert Weiss	NOSC	(714)-225-6441
Garry Bennett	DOE/OSNP	FTS-233-3197
Bart Bartram	NUS	(301)-258-8666
Nelson Stalnaker	LANL/CHM-1	FTS-843-4068
George Matlack	LANL/CHM-1	FTS-843-4068
Bob Clark	DOE/ALO	FTS-846-5208
Tom Keenan	LANL/NP	FTS-843-8039
Gene Peterson	LANL/CHM-1	FTS-843-5182
Dick Heaton	LANL/CHM-1	FTS-843-1141
Roy Greiner	LANL/CHM-1	FTS-843-5966
Paul Wagner	LANL/CHM-1	FTS-843-4923
Bob Campana	General Atomic	(714)-455-4356
Bob Campbell	JPL	FTS-792-4148
Ernie Johnson	MRC	FTS-774-3311
Jim Patterson	LANL/CHM-1	FTS-843-5959
Stan Bronisz	LANL	FTS-843-4782
David Jacobs	AFWL/NTTV	(505)-844-9791

LOS ALAMOS-DOE/OSNP

ENVIRONMENTAL PROGRAM REVIEW

September 21-22, 1982

Conference Room 1, CMR Building

September 21 PROGRAM ACTIVITIES

9:00 AM	Introduction	S. Bronisz
9:10	Environmental Organization	P. Cunningham
9:20	DOE/OSNP Environmental Programs	G. Bennett
9:35	Los Alamos Programs	
	History	G. Matlack
	Aquatic	G. Matlack
	Aquatic Pu Levels	N. Stalnaker
	Terrestrial	J. Patterson
12:30 PM	Lunch	
1:30	NOSC Program	H. Weiss
2:30	NUS Program	B. Bartram
3:30	SREL Field Program	G. Bennett
4:00	Discussions	All

September 21

9:00	General Discussions	All
10:00	Tour

INTERNAL CORRESPONDENCE

TO: B. W. Bartram, R. W. Englehart DATE: 83-059-RAD
Project 2818
April 12, 1983

FROM: M. J. Wilkinson *mfw* COPIES: File: 2818-06

SUBJECT: Trip Report on Meeting at
Oak Ridge National Laboratory
for Environmental Data Support Task

This report summarizes the meeting that took place at Oak Ridge National Laboratory on March 8, 1983. This meeting was conducted as part of the DOE Environmental Data Support Task. Present at the meeting were: -

- o C. Garten (ORNL)
- o E. Bondietti (ORNL)
- o B. Bartram (NUS)
- o M. Wilkinson

The specific purpose of the trip was to obtain an update on research that has been done at ORNL on transuranics in the environment since the publication of Transuranics in the Environment in 1980.

Garten has been summarizing the relative environmental behavior of various radionuclides, including U, Th, Pu, and Am. For this work, Garten measured uptake, concentration, and concentration ratios of biota in the Oak Ridge flood plain. (The flood plain had been used for liquid wastes from the Manhattan Project.)

By determining these parameters for uranium, Garten believes that he will have determined an upper limit for plutonium. Because uranium is more mobile than plutonium, the mean value for uranium uptake, concentration, and concentration ratio is higher than the same parameter values for plutonium. As was the case with other researchers, Garten reiterated the fact that differences in plutonium mobility are due to plutonium's different oxidation states: in water, plutonium in the oxidized state is dominant; in sediments and other environmental compartments, the reduced form dominates.

Plutonium concentration ratios for biota in the flood plain were all within the 10^{-3} range. Biota studied included grass, rat carcasses, tree leaves, and root and leafy vegetables.

Garten also looked at the uptake of Pu(IV) and Pu(V) by placing plant roots in a solution of nutrients containing both species of plutonium. Over time, all plutonium in the solution became reduced. Additionally, the exudate in the plants' xylem was all in the reduced state. Therefore, Garten believes that the plutonium ingested by plant-eating animals is in the reduced state.

Memorandum

B. W. Bartram, R. W. Englehart
83-059-RAD
April 12, 1983
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Bondietti had been studying the annual cycling of plutonium in the water column in Pond 3513. The pond had been used for liquid wastes during the Manhattan Project, and received waste until about 1974. According to Bondietti, the pond represents the upper limit of availability of radio-nuclides in a hard-water system.

The annual cycling in the pond peaks in the summer, with a minimum in the winter; the concentration of plutonium is 10 times greater in the summer than in the winter. In the summer, plutonium concentrations are 5 pCi/l in water and about 2000 pCi/g in sediments. Seventy-five percent of the plutonium in the water at this time is in the oxidized state. Bondietti believes that the cycling may be due to changing pH within the pond. The pH of the pond in the summer is 9 to 10, while in the winter it is 8 to 9. As the pH increases, it is possible that more carbonate (from the water) is available to react with the plutonium.

Cycling of plutonium concentrations has also been observed in Lake Michigan, although there, the maximum occurs in winter. Bondietti would not hazard a guess at why these differences exist; he would only say that there are many factors that must influence cycling.

Neither Garten nor Bondietti are currently working on the behavior of transuranics in the environment. They are, however, examining the validity of standard radiological assessment models, including EPA's AIRDOS and NRC Regulatory Guide 1.109.

INTERNAL CORRESPONDENCE

TO: B. W. Bartram, R. W. Englehart
FROM: M. Wilkinson *myw*
SUBJECT: Trip Report on Meeting at Argonne
National Laboratory for Environmental
Data Support Task

83-060-RAD
Project 2818
DATE: April 12, 1983
COPIES: File: 2818-06

This memorandum summarizes the meeting that took place at Argonne National Laboratory (ANL) on March 9, 1983 as part of the DOE Environmental Data Support Task. Present at the meeting were:

- o R. Larsen (ANL)
- o D. Nelson (ANL)
- o B. Bartram (NUS)
- o M. Wilkinson (NUS)

The purpose of this meeting was to obtain an update on research that has been done at ANL on the behavior of plutonium in the environment since the publication of Hanson's summary (Transuranics in the Environment) in 1980.

Nelson and Larsen have been characterizing the behavior of plutonium in aquatic systems. They have found that the assimilation of mono-atomic plutonium by mammals is about 100 times higher than with any polymeric form of plutonium. With mono-atomic plutonium, the form that occurs in natural waters, true solutions of plutonium can form.

According to Nelson and Larsen, other researchers examining the behavior of plutonium in water have prepared their own "solutions", rather than using natural waters. Solutions made in the laboratory apparently tend to be composed of larger, polymeric particles of plutonium. In contrast to polymeric solutions of plutonium, solutions of mono-atomic plutonium tend to approach equilibrium fairly rapidly. In fact, Nelson has found that the half-removal time from water to sediment in Lake Michigan is about one year.

If assimilation of mono-atomic plutonium is indeed 100 times higher than with other forms of plutonium in solution, Larsen has suggested that the GI uptake factor used in dose models be increased by a factor of 100. With such an increase, the MPC for plutonium should be correspondingly lowered by a factor of 100.

Nelson and Larsen have also measured the plutonium concentration ratio, K_d , in a number of salt and fresh water sources. They have found that the amount of oxidized plutonium is related to the amount of dissolved

Memorandum

B. W. Bartram, R. W. Englehart
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Page 2

organic carbon (DOC). As the K_d of oxidized plutonium decreases, the concentration of DOC increases; the plot is an approximate straight line on log-log paper.

To quantify the effect of DOC on the K_d of Pu(IV), the following experimental technique was used.

- 1) Take samples (50 l) of natural waters
- 2) By ultra-filtration, remove the sediments
- 3) Add the sediments back in controlled concentration, as ppm of ligand
- 4) Determine the K_d of Pu(IV) at each ligand concentration.

These experiments showed that for all samples of natural water tested, the K_d with no ligands, i.e., low DOC, was about 10^6 . For each water sample, a similarly shaped curve was produced: on log-log paper, the curve has an initial shoulder that breaks to form a straight line with a slope of about one. Therefore there is probably only one DOC attached to each Pu(IV). Differences in the curves were in the width of the shoulder, with a broader shoulder corresponding to increased ligand concentration. The ligand concentration at which the shoulder breaks indicates the point at which Pu(IV) begins to be adsorbed onto the ligands.

Larsen should, by this time, be partially retired and working part-time. Presently, he and Nelson are examining the validity of groundwater migration hypotheses. They believe that plutonium should be mostly in the reduced form in groundwater because of the absence of DOC.

INTERNAL CORRESPONDENCE

TO: R. W. Englehart
DATE: 83-088-RAD
June 8, 1983

FROM: B. W. Bartram BWB
COPIES: M. Wilkinson

SUBJECT: Trip Report on Meetings at Pacific
Northwest Laboratories

This trip report summarizes meetings between personnel from NUS and Pacific Northwest Laboratories (PNL) during a visit to PNL March 10-11, 1983 in support of the DOE Space Systems Project, Task 82-8 (Plutonium Environmental Data). The purpose of the visit was to meet with researchers who are conducting studies related to the behavior of plutonium in the environment in order to gather information for use in risk assessments of plutonium-fueled space nuclear systems. Individuals involved in these meetings were as follows:

O. Moss	(PNL)	G. Shrekheise	(PNL)
R. Wildung	"	W. Napier	"
W. Templeton	"	W. Kennedy	"
R. Thompson	"	W. Robertson	"
E. Jenne	"	M. Wilkinson	(NUS)
J. Thomas	"	B. Bartram	(NUS)
R. Gilbert	"		

A summary of the work being done by each researcher is presented below:

O. Moss - O. Moss acted as overall coordinator of the meetings between NUS and PNL personnel. O. Moss has conducted studies in the past under contract to DOE/OSNP related to the effect of agglomeration on particulate plutonium dioxide source terms. His current research centers on aerosol/particle physics with one of his responsibilities being the characterization and generation of particle size distributions of various compounds, including plutonium dioxide, for use in animal inhalation studies.

Principal research tools utilized by O. Moss in this work include an electron microscope, a BET Apparatus, a wind tunnel, one of each type of aerosol generator, and one of each type of aerosol detector. A software package is used in conjunction with the electron microscope that essentially automates particle sizing, reducing the time required to roughly one tenth of that required by the manual method. A new wind tunnel has just been constructed that is much larger than the one previously used. It is designed to handle radioactive particulates for resuspension, deposition, and agglomeration studies. The BET Apparatus is used to characterize particle surface areas by noting weight changes by a microbalance due to the deposition of a layer of liquid nitrogen one molecule thick on the particle surface. The BET Apparatus can also be used to characterize the reaction of particles to gases. Other methods of particle sizing utilized include cascade impactors and a laser particle sizer that look at the time of flight between two laser beams.

Memo to: R. W. Englehart
83-088-RAD
June 8, 1983, Page 2

I described to O. Moss the current programs being conducted by LANL in support of DOE/OSNP in order to obtain any guidance he might have. One problem described by R. Englehart related to plutonium dioxide source terms due to impact testing of fuel forms is what is the best way to characterize released fuel mixed up in broken graphitics. This characterization would include quantifying the total quantity released and determining the particle size distribution. One technique LANL has considered is ashing the graphitics and characterizing the left over fuel. The concern with this approach is that the ashing process may alter the particle distribution.

O. Moss had several suggestions related to this problem. The ashing approach should not really cause any problems as long as the ashing is done at a low enough temperature that sintering does not occur. Handling of the sample after ashing should then be minimized to the extent possible. One way to minimize handling is to characterize the particle size distribution by the sedimentation method. The ashed fuel can be effectively broken up by using a good soap solution that breaks down interparticle forces. Then the particle size distribution is determined by measuring particle settling rates in viscous oil. However, I feel that surface tension effects of the oil surface may cause particles to be retained on the surface, causing problems with this method. Also, the particle size range of interest may not correspond to particle size range for which the sedimentation method is best suited.

O. Moss suggested a literature resource that may have information applicable to this problem as well as other problems related to particles. The publication is entitled Current Awareness of Particle Technology that is published once a month and summarizes all published information related to particle technology by title and cross references. It is published by:

Particle Science and Technology Information Service
University of Technology
Loughborough, Great Britain

O. Moss also suggested that S. Bronisz's group at LANL make contact with Sid Solderholm at LANL who does work in aerosol/particle technology.

O. Moss indicated that the problem of plutonium dioxide particles embedded in graphite is similar to particles embedded in lung tissue. In the latter case a standard technique is freeze the particles in place by pouring plastic over the mass and letting it harden. Then the material is sliced in a series of planes and examined microscopically to determine the particle size distribution. Then the mass can be ashed to determine the total radioactivity. Perhaps this same approach could be used for the graphitics case.

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O. Moss also had some suggestions related to the plutonium dissolution experiments being conducted at LANL. The topic of discussion was how can the dissolution process be best characterized. He suggested a dynamic system be used to answer the questions of 1) is the surface wearing away, 2) what does the surface look like in the process, and 3) what is the nature of the plutonium leaving the surface. The dynamic system would consist of water flowing past the fuel. Downstream released ionic plutonium and particulate plutonium dioxide would be separated by using selective adsorption media. He noted that in measuring rate limiting mechanisms leading to dissolution careful attention should be given to wall losses. Wall materials should be chosen so that they will not accept plutonium at the pH of the water.

R. Wildung - R. Wildung conducts research related to the behavior of transuranics in terrestrial ecosystems. He gave an overview of similar research being conducted by others at PNL. These studies are directed at understanding basic processes and mechanisms that control the behavior of transuranics in terrestrial ecosystems, rather than the mere collection of data such as concentration ratios. Most of this work seemed to emphasize plant uptake of radionuclides. In this regard, R. Wildung indicated that the solubility of a given radionuclide plays a very important role in root uptake. Research has indicated recently that another important factor in root uptake is that plants may selectively uptake radionuclides depending on how similar the radionuclides are to important plant nutrients. For example, a plant may selectively uptake a given radionuclide because the plant may view it as sulfur or some other nutrient that it requires. R. Wildung indicated that in about six months the research at PNL may be able to determine what nutrient, if any, plants view plutonium as.

W. Templeton - W. Templeton is involved in studies related to radioactivity in the marine environment. A lot of data is available on the behavior of radionuclides from Windscale, the Northeast Atlantic Dump Site and NEA Monoco. The NEA has a seabed working group on this subject. Sandia has a seabed disposal program that may have useful information.

He is a member of NCRP Committee 64 (Environmental) that has been examining dose assessment models. The results of this review will be published in a report in the near future. One conclusion is that the concentration ratios used by the NRC are not very good. Actual concentration ratios are highly variable. The committee has air, land and fresh water panels but has not looked at marine models yet. Owen Hoffman did parameters sensitivity studies in support of this work.

R. Thompson - R. Thompson is on ICRP Committee 2 that is in the process of revising ICRP Publication 19 related to the internal dosimetry of the actinides. The gut to blood transfer factor, f_1 , for ingested plutonium will be raised due to studies that have shown that ingested plutonium is more soluble than originally thought.

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In the original ICRP 19, f_1 was 3×10^{-5} . In ICRP 30 it was revised to 10^{-4} . In the revised ICRP 19, f_1 is expected to be 10^{-3} . This could make the ingestion pathway doses for released plutonium much more important in future risk assessments of plutonium fueled space nuclear systems. In addition to this change the residence half-time of plutonium in the lung may be reduced. This would tend to reduce lung doses and increase bone doses due to inhaled plutonium.

The NCRP does not have an overall risk assessment scheme that ICRP has in ICRP 26. NCRP feels the data does not support the organ weighting scheme in ICRP 26. The NCRP won't have a comparable scheme for several years. NCRP Committee 57 (Internal Emitters) has concluded that the internal dosimetry model and dose factors in ICRP 30 are the best available and preferred. With regard to the plutonium internal transfer parameters in ICRP 30, there is a question as to whether plutonium actually ends up on bone surfaces. The transfer factor may be reduced by a factor of 3 or 4.

E. Jenne - E. Jenne is involved in research related to process level simulation modeling of transuranic behavior in the environment. Much of his previous work was related to modeling plant processes in order to characterize plant root uptake of transuranics. His present work consists of the development of geochemical models for use in characterizing transuranic migration in groundwater systems. He is developing a geochemical model for plutonium that incorporates the thermodynamics of plutonium so that the distribution of plutonium among its various dissolved complexes can be calculated. There are two other plutonium geochemical models in existence developed by Lawrence Berkely Laboratory and a Canadian group, respectively. The PNL model incorporates 35 possible plutonium complexes. Iron, manganese, and particulate organic carbon are the primary plutonium absorption sites in the model. Silica may be important in clean sands. Primary and secondary minerals coated with iron, manganese, organic carbon, and aluminum silicates may also be important. For a given geochemical system, including aquatic systems, quantitative information on the density of these absorption sites represent the bulk of the input data.

With regard to Cape Canaveral, E. Jenne indicated the U.S. Geological Survey should have data on dissolved organic carbon in Florida lakes, as well as other pertinent data.

G. Sehmel - G. Sehmel's research area includes resuspension, dry deposition, and fugitive dust. He identified several papers and reports summarizing resuspension data and models. One problem he noted concerned a resuspension model incorporated in NRC dose assessment models as well as OSM models. The model assumes an initial resuspension factor of 10^{-5} that decreases to 10^{-9} exponentially after two years with the residual resuspension factor remaining 10^{-9} thereafter. He indicated that the original data on which this was based from the Nevada Test Site was highly scattered. Measurements taken years later indicated the resuspension factor did not really decrease. Another problem is that if residual resuspension factors are indeed as low as 10^{-9} , initial resuspended air concentrations must have

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been extremely high if the resuspension factor was initially 10^{-5} . This may have overlooked health implications.

J. Thomas - J. Thomas has been associated with the Pu-239 animal inhalation studies at PNL and research related to the statistical design of environmental experiments associated with released radioactivity. He noted that a lot of environmental studies designed to quantify parameters for use in dose assessments collect a lot of data but don't focus on the basic mechanisms that would lead to a better understanding of the behavior of radionuclides in the environment.

R. Gilbert - R. Gilbert conducts research on statistical methods in environmental studies. He prepares a periodic bulletin entitled "TRAN-STAT, Statistics for Environmental Studies" under contract to DOE/OHER that addresses the statistical design of experiments and the statistical analysis and interpretation of the resulting data. He described a new technique called "kriging" that is a moving average method that looks at the structure of the data. It is used to develop distribution patterns based on a set of point values. For example, it would be useful in constructing ground concentration isopleths from a set point measurements.

G. Shreckheise - G. Shreckheise conducts research related to plant uptake of radionuclides. He utilizes field plots and lysimeters in this work. The lysimeters are essentially PVC pipes that are put in the ground, filled with a desired mix of soil and radionuclides and then plants put in for study. He noted that biological mechanisms were a very important factor in redistributing radionuclides in soil. These biological mechanisms include plant roots, burrowing animals, earthworms, and microbes.

W. Napier and W. Kennedy - Both W. Napier and W. Kennedy perform radiological dose assessments and develop supporting models. They described the models utilized at PNL and discussed the advantages and disadvantages of models used by NRC and other groups.

APPENDIX B

A Sample Experimental Plan

**DESIGN CONSIDERATIONS FOR EXPERIMENTS
CHARACTERIZING PLUTONIUM IN AQUATIC ENVIRONMENTS**

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December 30, 1980

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DESIGN CONSIDERATIONS FOR EXPERIMENTS CHARACTERIZING PLUTONIUM IN AQUATIC ENVIRONMENTS

As an example of the type of approach recommended by NUS in the design of experiments characterizing plutonium behavior in the environment, design considerations applicable to plutonium aquatic environment experiments are summarized below. This example is not intended to be comprehensive, nor is the supporting model necessarily the most representative. The model was selected only for its simplicity and computational ease.

1.0 Literature Review

Aquatic transfer processes include the interaction of airborne dispersed activity and intact point sources with water bodies, such as rivers and streams, bays and large lakes, and oceans. These interactions include the dissolution of particulate and point sources, transfers to sediment, transfers to other water bodies, and transfers to food supplies such as fresh water food or seafood.

Plutonium behavior in aquatic systems is governed to some extent by the oxidation state in which plutonium is found. Plutonium can have four oxidation states, including Pu(III), Pu(IV), Pu(V), and Pu(VI). These are essentially valence states having differing chemical behavior, and are analogous, for example, to the two oxidation states of iron, Fe(II) and Fe(III). In general, Pu(III) and Pu(IV) are reduced states and Pu(V) and Pu(VI) are oxidized states of plutonium. The reduced states are generally found as precipitates associated with sediment and the oxidized states represent the waterborne, soluble fraction. Pu(VI) is usually rapidly converted to Pu(V). Plutonium tends to be in the oxidized state, and therefore soluble, in systems having high pH and high carbonate concentrations. Soluble, oxidized states also occur when plutonium becomes complexed with certain organics such as humic acid associated with decaying vegetation and chelating agents such as those found in fertilizers. Plutonium tends to be in the reduced state, and hence tied up in sediments, when pH, dissolved carbonates, and certain organics are low, and when reducing agents such as Fe and Mn are present.

Some data indicate that a transition from oxidized states to reduced states occurs when pH drops below 8. The conditions outlined above often result in flip-flop situations in the environment. For example, most of the fallout plutonium in Lake Michigan with low carbonate and pH levels (less than 8) is in the sediments. At the other extreme, the plutonium in Mono Lake with high pH and carbonate levels is in solution. The oxidation states of plutonium occurring in the environment are actually much more complex than summarized above, in that trace quantities of unidentified compounds may also affect the oxidation state in which plutonium occurs in a given situation. (1, 2, 3)

As in the case of terrestrial systems, the major sink (about 96%) for plutonium in the ocean or a body of fresh water is the sediment. (4,5) Over long periods of time the remaining few percent may recycle. This was found in studies of Bikini Atoll, where plutonium is being redistributed from the sediment to the overlying water column in a highly complex way. (5,6) Similar evidence of plutonium redistribution was found in Lake Michigan, where 3% of the plutonium is in the water and most of the plutonium (97%) is in the sediment. (7) In the top 6 cm it is evenly dispersed and has a removal half-time of 3 to 4 years. About 3% is recycling. This figure (3%) seems to be relatively constant over longer periods of time and has been observed in other Great Lakes.

2.0 Model Basis

The dissolution of fuel in the form of particulates or intact sources will be complex in nature, being a function of fuel encapsulation, chemical and physical form, and fuel geometry. The dissolution of fuel can be represented as shown in Figure 1. Encapsulated fuel in an aquatic environment is assumed to begin releasing dissolved fuel after time t_0 , which is the time required for the encapsulation material to be dissolved. The quantity of dissolved fuel in solution as a function of time, referring to Figure 1, can be shown to be

$$q(t) = \frac{Q(0)\lambda_d\delta(t-t_0)}{\lambda_{fs} + \lambda_d - \lambda_{ds}} \left\{ e^{-[(\lambda_{fs} - \lambda_{ds})t_0 + \lambda_{ds}t]} - e^{-[\lambda_{fs}t + \lambda_d(t-t_0)]} \right\}$$

where

$$\delta(t-t_0) = \begin{cases} 1 & \text{if } t \geq t_0 \\ 0 & \text{if } t < t_0 \end{cases}$$

t_0 = time required for dissolution of encapsulation material to occur, yrs.

This formulation can represent a spectrum of aquatic release scenarios, ranging from the immediate release of the entire inventory ($t_0 \rightarrow 0$ and $\lambda_d \rightarrow \infty$), delayed release associated with the dissolution of very soluble fuel (large λ_d), and the dissolution of very insoluble fuel (small λ_d).

Consider the application of this model to actual data. Studies of plutonium in aquatic environments indicate that dissolved fuel is transferred to the sediment sink with a 3.5 year half-life. (5)

Sediment sink in this case is taken to be that portion of sediment below the top 6 cm. Therefore,

$$\begin{aligned}\lambda_{ds} &= \frac{0.693}{3.5 \text{ years}} \\ &= 0.198 \text{ yr}^{-1}\end{aligned}$$

The λ_{fs} in Figure 1 and the equation represents the rate at which the intact^s fuel is removed from the aquatic environment by being covered with sediment or encrustation. The removal half-life associated with this removal mechanism has been somewhat arbitrarily taken to be 50 years, which is considered reasonable. Hence,

$$\begin{aligned}\lambda_{fs} &= \frac{0.693}{50 \text{ years}} \\ &= 1.386 \times 10^{-2} \text{ yr}^{-1}\end{aligned}$$

The quantity of dissolved fuel $q(t)/Q(0)$ has been evaluated parametrically as a function of time for a range of λ_d and t_0 values, and the results are presented in Figure 2.

The above model represents only one of many possible types of models that could be formulated to characterize such situations. A further discussion of alternative model formulations is presented in References 8 and 9.

3.0 Experimental Design Considerations

Based on the above information, experimental design considerations can be developed further. First, consider a possible set of idealized qualitative and quantitative objectives of experiments characterizing plutonium in aquatic environments.

- Qualitative Objectives - Characterize the behavior of plutonium in aquatic environments through experiments representative of typical initial source term conditions and a spectrum of actual aquatic environments. Develop an understanding of the chemical and physical mechanisms governing such behavior.
- Quantitative Objectives - Characterize the mass transfer rates of plutonium among all elements of a given aquatic environment as a function of specified chemical and physical parameters controlled by the experiment.

Experimental design should emphasize 1) realism reflecting actual aquatic environments, 2) obtaining a mass balance of plutonium in all elements of the system as a function of time, 3) control and measurement of selected chemical and physical parameters, and 4) interpretation of the results through model simulation.

The results of such an experiment can then be used as input to broader systems models describing the behavior of plutonium in a more global environment. (8) The above approach represents an idealization of course, but careful experimental design is considered the key.

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$Q(t)$ = QUANTITY OF FUEL INTACT AT TIME t , GRAMS

$q(t)$ = QUANTITY OF DISSOLVED FUEL IN SOLUTION AT TIME t , GRAMS

λ_d = FRACTION OF INTACT FUEL DISSOLVED PER UNIT TIME, yr^{-1}

λ_{fs} = FRACTION OF INTACT FUEL COVERED BY SEDIMENT PER UNIT TIME, yr^{-1}

λ_{ds} = FRACTION OF DISSOLVED FUEL IN SOLUTION TRANSFERRED TO SEDIMENT SINK PER UNIT TIME, yr^{-1}

FIGURE 1 DISSOLUTION OF ENCAPSULATED FUEL IN AQUATIC ENVIRONMENTS

Figure 2 Dissolution of encapsulated fuel in aquatic environments as a function of time and dissolution rate