ERDA-76/134

Workshop On Environmental Research For Transuranic Elements

Proceedings Of The Workshop November 12-14, 1975

Battelle Seattle Research Center
Seattle, Washington

Energy Research & Development Administration
Division Of Biomedical & Environmental Research

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

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Th	Pa	U	Np	Pu	Am	Cm

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Introduction

The Workshop on Environmental Research for Transuranium Elements was organized to provide a forum for communication and coordination among the research scientists who have ongoing projects and to reveal needs and ideas for additional research. This was the second workshop sponsored by the Division of Biomedical and Environmental Research on this subject. The first was held at Estes Park, Colorado, July 1974 to aid those scientists just beginning research in this expanding field by introducing them to the problems and questions encountered by those already conducting research with the transuranium elements. The first workshop focused on land and freshwater research while this second workshop included marine sciences as well.

The three day meeting consisted of a plenary session on the first day in which each scientist described very briefly the objectives and results of his research project to date. This was followed on the second and third days by group discussions in four separate topical panels. These panels and their chairmen were:

Plant Uptake----------Roger C. Dahlman
Transport and Diffusion in
Aquatic Systems----------Vaughan Bowen
Soil and Sediments----------John C. Corey
Ecosystems and Food Chains----Wayne C. Hanson

On the afternoon of the third day a closing plenary session was held where the panel chairman summarized the discussions.

This report is a description of the workshop and its results as observed by BER staff and as summarized in detail by the panel chairman.

The BER staff wishes to thank all of the participants for their enthusiastic response and extends special thanks to the panel chairman for the time and energy which they expended in planning, conducting and reporting the panel discussions.

Plenary Session

The workshop was begun with welcoming remarks from Burton E. Vaughan, Manager, Ecosystems Department, who represented the Battelle Pacific Northwest Laboratories, the host organization. He then introduced Robert L. Watters, BER, who described the workshop as a communication and coordination device among the growing number of scientists in the United States who were concerned with environmental research for the transuranium elements.

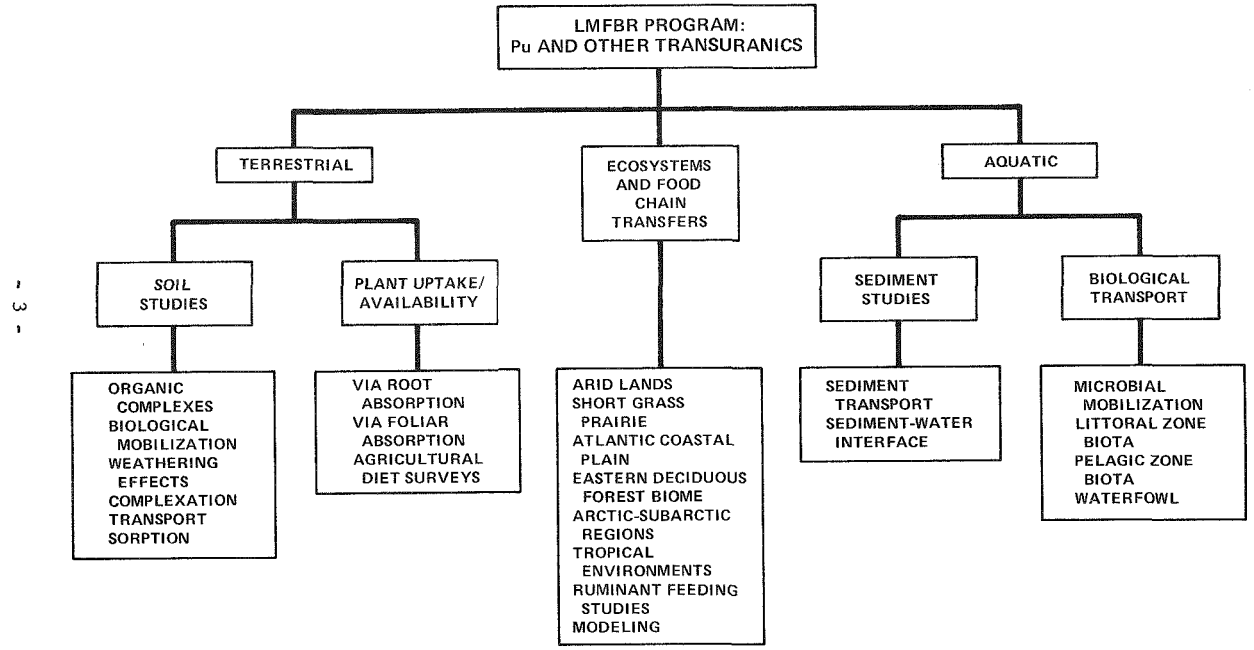
He briefly described the BER-ERDA program in this area as one which was developed to determine how and to what extent the transuranium elements released to the environment would enter the human population. As a corollary to that objective, he expressed the hope that a portion of the research would eventually describe to what extent the long-lived transuranium elements might persist in the environment as potential contaminants to the human population. The overall research program was developed from a logic tree which identified necessary tasks to obtain the necessary information. (A simplified version of this logic tree is seen in Figure 1.) Research was funded on the basis of this plan after identifying what studies were already supported by ERDA or EPA.

John W. Bartlett (Battelle Pacific Northwest Laboratory) (PNL) reviewed the needs for research relative to waste storage and disposal which he described as the Achilles Heel of the nuclear industry.

Generally, it is possible from an engineering standpoint to design a waste facility which could be generally effective for thousands of years. What is needed is information on the extent of release that could be allowed under various environmental conditions. This information is needed to arrive at engineering design which is economical and effective. Within this broad need several research tasks are apparent. For instance, site selection for waste storage is strongly dependent upon the geology, seismology and hydrology of the area. In the area of waste management, radiation and heating effects upon the environment need to be evaluated and the behavior of the transuranium elements in soils and ground water must be understood to make decisions about burial of low level wastes.

Burton E. Vaughan (PNL) pointed out the need for increased analytical capabilities for transuranium assays. He pointed out some of the pitfalls to reliable results which included interfering ions within

FIGURE 1


LMFBR PROGRAM:
Pu AND OTHER TRANSURANICS
TERRESTRIAL
ECOSYSTEMS AND FOOD CHAIN TRANSFERS
AQUATIC
SOIL STUDIES
PLANT UPTAKE/ AVAILABILITY
SEDIMENT STUDIES
BIOLOGICAL TRANSPORT
ORGANIC COMPLEXES
BIOLOGICAL MOBILIZATION
WEATHERING EFFECTS
COMPLEXATION
TRANSPORT
SORPTION
VIA ROOT ABSORPTION
VIA FOLIAR ABSORPTION
AGRICULTURAL DIET SURVEYS
ARID LANDS
SHORT GRASS PRAIRIE
ATLANTIC COASTAL PLAIN
EASTERN DECIDUOUS FOREST BIOME
ARCTIC-SUBARCTIC REGIONS
TROPICAL ENVIRONMENTS
RUMINANT FEEDING STUDIES
MODELING
SEDIMENT TRANSPORT
SEDIMENT-WATER INTERFACE
MICROBIAL MOBILIZATION
LITTORAL ZONE BIOTA
PELAGIC ZONE BIOTA
WATERFOWL

the sample matrix, and interfering ions and isotopes introduced with transuranium spike solutions. These interferences should be carefully considered before starting plant uptake studies or field studies where unusual chemical conditions may exist such as those encountered in waste disposal ponds. He urged more intercalibration on samples among the various laboratories.

The next series of presentations were grouped to correspond roughly to the subsequent panel participations.

Roger C. Dahlman (ORNL) described the research at Oak Ridge as dealing primarily with the behavior of the transuranium elements in the humid southeastern United States. These studies are concentrated on determining the mobility in soils and biota, and changes in the nature of the various TU elements during various cycling processes. This work includes field studies on a small flood plain of two hectares which was contaminated in 1944, aquatic studies in White Oak Lake and environmental chemistry studies designed to characterize plutonium and other TU elements in soils, sediments and plant extracts.

Michael H. Smith (SREL) discussed the work at Savannah River Plant where a cooperative program is carried out by the University of Georgia's Savannah River Ecology Laboratory, (SREL) and Du Pont's Savannah River Laboratory. They take advantage of the low level but readily detectable plutonium which has been released to the streams and deposited from atmosphere releases. These studies include an agricultural field plot where wheat and soybeans have been harvested and corn will be planted. Concentration ratios between soil and plants for plutonium isotopes are generated from this study. Studies are also underway to determine the movement of plutonium through terrestrial and aquatic food chains.

Frederick Au (EPA - Las Vegas) reviewed his work on the uptake of TU elements by soil microorganisms. These include the identification of TU elements in fungal spores as a possible transport mechanism and the comparison of the effect of various chemical forms and substrate conditions upon uptake. The influence of microbial populations on the movement of TU elements through soils is also under investigation.

John Koranda (LLL) described the agricultural program at Enewetak which is designed to measure fission products and plutonium. These studies are in support of the program to safely return the inhabitants to the atoll. Food crop plantings occurred this summer and the results are not yet available. Earlier surveys at Engebi Island in the Atoll indicated plant-soil concentration ratios for selected plants to be in the range of 10^{-4} to 10^{-3}.

Evan M. Romney (UCLA) is studying the uptake of Pu by desert plants at the Nevada Test Site. Field studies show plant-soil concentration ratios from 10^{-2} to 10^{-1}, but these are largely attributable to resuspension and foliar deposition rather than root uptake. Greenhouse experiments produce concentration ratios of 10^{-5} to 10^{-3}. The ratios of Pu to Am appear to be lower in plants than soils on which they grow which indicates an enhancement of Am in plants. This observation leads to the inference that Am is more mobile than Pu in this environment. Soils at NTS have shown little alteration over the past 20 years.

Arthur Wallace (UCLA) has been conducting greenhouse studies on plant uptake of TU elements in the presence of commercial chelating agents of agricultural importance such as DTPA and EDTA. Results show that in the pH range of 5 to 8, chelates of Am become increasingly available in soils with increasing pH. Plutonium, on the other hand, shows enhanced plant uptake with the same chelates at lower pH values. Americium appears to show some similarity to other heavy metals such as Pb and Ra in its behavior to chelating agents.

Raymond E. Wildung (PNL) described his research on the role of soil and plant factors in Pu availability. Specific studies have included the effect of soil microbes on uptake, the effect of the chemical form of Pu introduced to the soil and Pu complexes which are formed either by biological action or by soil amendments. Pu solubility appears to be the critical factor, i.e., any mechanism which renders Pu more soluble increases its uptake by plants. The conclusions so far developed are that (1) Pu in soil is consistently less than 0.1% water soluble, (2) Pu components which show apparent water solubility have molecular weights of about 10^4 daltons (amu), (3) organic complexes of Pu are long lived in soil, (4) Pu-resistant fungi modify Pu behavior in soil, and (5) seeds of plants show 10^{-7} concentration ratios.

Gene Schreckhise (PNL) is studying the effects of weathering and aging on plant uptake of the TU elements. The radionuclides under study are ^{238}Pu and ^{241}Am with Np and Cm planned for future experiments. They are mixed with soil which is then placed in PVC pipes which act as containment vessels and lysimeters. These are placed vertically in the soil in the field and seeded with various agricultural crops and wild grasses. Results so far show that seeds have low concentration ratios relative to leaves and that plant uptake may be increasing with time over the first year.

Robert Schulz (University of California) conducts laboratory experiments on Pu and Am uptake in agricultural plants. He uses Nevada Test Site soils as well as soils spiked in the laboratory. His results for barley in soils spiked with nitrate forms of Pu and Am show plant-soil concentration ratios in the following ranges:

	Plants	Seeds
Pu	10^{-5}	10^{-8} to 10^{-6}
Am	10^{-4}	10^{-7} to 10^{-5}

Dominic Cataldo (PNL) conducts experiments on foliar deposition and foliar absorption of Pu. His studies include measurements of the bio-availability as a function of particle size, influence of chemical form and effects of rainfall. Results so far indicate that Pu compounds become decreasingly available with time. The accumulation ratios, pCi/plant tissue/pCi/leaf tissue, are usually 10^{-6} to 10^{-5}.

John C. Corey (Savannah River Laboratory) described resuspension studies which were performed during the cultivation of a field contaminated with ^{238}Pu and $^{239-240}Pu$ at a level 200 times background. A calculation indicated a 70-year dose commitment of 49 mrem to a tractor driver if he continued this work for 100 days without respiratory protection. On the other hand, the wheat which was grown on the field had a concentration of 1 fCi/g and would have produced a dose commitment of 0.04 mrem to a person eating 150 g per day for one year.

He has also studied the concentrations of Pu on the inlet and outlet sides of water treatment plants on the Savannah River. Levels on the inlet side varied from 0.4 fCi/l upstream of the Savannah River Plant to 2.2 fCi/l downstream. Outlet water (potable water) is consistently 0.07 to 0.09 fCi/l at all locations.

Al Boni (SRL) discussed the analytical support to the Savannah River Plant studies which includes air sampling, particle characterization and radiochemical analysis. He stressed the importance of source term characterization and the development of techniques to determine what chemical alterations are occurring in the environment.

Their studies with air sampling downwind from separations plant stacks have shown changes in $^{238}Pu/^{239}Pu$ ratios which indicate changes in deposition velocities. This should be investigated in greater detail as to the nature of the chemical and physical forms of the airborne material.

Ernest A. Bondietti (ORNL) is studying the soil chemistry and physical chemistry of the TU elements. He noted that with leaching experiments on high fired Pu oxides that about 70 percent of the radioactivity in solution is Am.

T. Tamura (ORNL) characterizes the Pu present in soils where known contamination events have occurred. He identifies the soil size fractions, density fractions and mineralogical components associated with the radionuclide. He characterizes the chemical parameters of the plutonium such as requirements for total dissolution, degree of dissolution by selective solvents, selective complexing, ion exchange characteristics and oxidation-reduction characteristics. Leachability studies have shown considerable differences in contaminated soils at various locations which are probably due to different original forms of Pu and to different soil chemistry. For example, NTS soil treated for 30 minutes with HNO_3 solution leached 10 percent of the TU while the Oak Ridge soil under the same treatment lost 70 percent of the TU.

Donald A. Brown (University of Arkansas) is studying the diffusion of Pu in soil-water systems and desorption characteristics. Experiments have been performed both with aqueous diffusion systems and with soils. The range of diffusion coefficients in soils is 10^{-8} to 10^{-7} cm^2/sec, but 10^{-9} cm^2/sec has been measured in some instances for less soluble forms.

John C. Shephard (Washington State University) is studying the distribution and diffusion of Np, Cm and Am in soils. The objectives are to define diffusion coefficients and to evaluate the degree of complex formation with natural ligands such as those in humic and fulvic acids. Observations so far indicate the distribution ratios between soil and soil-water decrease with time. It is inferred that radiocolloid complexes are formed to influence behavior.

Frend J. Miner (Rockwell International - Rocky Flats) studies the movement of Pu in soil/water systems. Sorption measurements at equilibrium have been made on various U.S. soils. The effect of Pu particle size on soil sorption has recently been the subject of study and the elution of Pu from soil columns has started. Results so far are:

1. For plutonium nitrate all soils sorbed greater than 99 percent of added tracer.

2. Sorption is influenced by chemical and physical properties of Pu and by soil factors.

3. Soil column elution indicates movement, especially in the initial fractions indicating a small but highly mobile form present in the initial nitrate form.

4. Early experiments indicate that the equilibrium sorption of Am is higher than for Pu in the same chemical form.

W. A. Polzer (Idaho National Engineering Laboratory) works in cooperation with F. J. Miner. He discussed the effects of polymers of Pu upon results in cation exchange and sorption experiments. Where care is taken to remove polymers there are no concentration effects in the molarity range 10^{-8} to 10^{-6}. Increase of pH within the range from 4 to 8 causes decreased sorption in the absence of polymers.

H. Nishita (UCLA) is studying the soil chemistry of Pu, Am and Cm. His objectives are to delineate the various factors which affect the mobility of the radionuclides in soils. These will be followed by plant uptake experiments. He expects to develop a method of solvent extraction on soil which will define availability and which could be used for site evaluation for fuel processing plants.

E. H. Essington (Los Alamos Scientific Laboratory) described the function of the Nevada Applied Ecology Group, Soils Element Group. In general, NTS soil profiles show rapid decline in Pu concentration with depth but there are complexities at some locations. Plutonium has been accumulated at the base of desert shrubs in wind deposited blow sand but quantitative description is not yet available.

(Ed. note.) This project is funded through NAEG and does not receive funds from DBER.

J. H. Patterson (LASL) is concerned with environmental consequences of the release of ^{238}Pu from radioisotope thermal generators. This program has three aspects which are:

1. Distribution of ^{238}Pu in air, soil, and water are measured at times after ^{238}Pu microspheres are distributed on soil in simulated terrestrial environments. Simulated rainfall increased the air concentration probably due to spallation, when water hits the thermally hot microsphere. Percolation of rain through soil has produced some vertical movement of ^{238}Pu.

2. Simulated seawater experiments using fuel elements made from a Pu and Mo alloy are run to determine rates of corrosion and dissolution. This work is funded by Space Nuclear Systems-ERDA.

T. E. Hakonson (LASL) discussed the work in progress at the liquid effluent areas in Los Alamos and the studies at Trinity, the site of the first nuclear weapons test. The present studies have been concerned with the distribution and behavior of Pu and Am, mechanisms of movement and experimental studies. Plutonium at 16 km from Trinity has penetrated 5-10 cm below the soil surface in 30 years. Rain water penetration correlates with Pu distribution. At 1.6 km from Trinity the soil fraction less than 53 μm contains little Pu but at 44 km the less than 53 μm fraction contains much of the Pu. At LASL (73%) of the Pu is in the less than 53 μm fraction. One experiment which involved sampling stream sediments in a liquid effluent canyon before and after runoff shows significant lateral distribution and vertical distribution changes within the stream bed for plutonium. It is inferred that the nature of the movement is due to the strong association with the fine sediments.

S. M. Fried (Argonne National Laboratory) studies the behavior of Pu and Am in rock (LASL tuff and EBR basalt). He simulates ground water under various hydrostatic pressures with Pu on rock surfaces. Cores of the rock are then analyzed to determine penetration. Two components of Pu have been observed, one slow and one fast. The fast moving component is believed to be a polymer. Americium behaves much like Pu in the rock specimens.

W. C. Weimer (PNL) is using chemical analogs in an attempt to infer the long term behavior of the TU elements. The objectives of this work are first to demonstrate the feasibility and if successful to carry out experiments which rely upon the natural concentration of the analog in soil. The work will start with neodymium as an analog for Am. Later studies will explore the possibilities of Th^{+4} and Ce^{+4} as analogs for Pu^{+4}.

D. N. Edgington (ANL) discussed his work on Pu associations with sediments in Lake Michigan. He noted that in this environment ^{137}Cs follows Pu in sedimentation with distribution on the bottom generally on the east side of lake. Fully 97 percent of the calculated fallout to the watershed is in the lake sediments already. The Pu is associated with the chemical reducing fraction of the sediments.

Wayne C. Hanson (LASL) described the arctic studies which LASL conducts with BER support. The objectives of the research are to describe routes, rates and amount of TU elements in arctic ecosystems. The simple food chain lichen-caribou-man is a good one for developing discrimination

factors for the upper trophic levels including man. Comparisons of TU element cycling behavior in a variety of arctic ecosystems, plant communities and animal types is also possible because of a collection of specimens which span 16 years. The results of these studies can be compared to other studies at Trinity, NTS, and Thule to note similarities or differences which may be due to source term or environmental factors.

Paul Dunaway (NVO, Director of NAEG) quickly outlined the NAEG program noting that the experimental work was represented by several other NAEG participants. He mentioned that the basis for the studies had been the evaluation of hazards and need for clean up at NTS. Much work has been done on movement through food chains and the importance of wind resuspension as a method of transport. Determination of isopleths of Pu concentrations about known contaminated sites has been a major effort. NAEG also maintains a Transuranium Information Center at Oak Ridge and a study farm at NTS which is available for agricultural experiments with crops and cattle.

D. C. Adriano (SREL) has been concerned with the Agricultural experiments at the Savannah River Plant. Results of the wheat planting in the open field mentioned by Smith and Corey indicate that most of the small amount of activity detected in the wheat is due to resuspension of dust. This is inferred from the ^{238}Pu ratios.

Experiments with $^{239,240}Pu$ on different soils, with liming, and with application of chelating agents produced significant effects on Am uptake. In bacterial/algae studies with living and dead cells, Am concentration increased with increased cell density and greater uptake occurred with living cells.

J. E. Pinder (SREL) discussed the experimental design for the Savannah River Plant studies. The objectives were to develop designs which would give a detailed analysis of the conditions within 0.5 km of the release point where a fairly steep concentration gradient occurs and which would give reliable information at 5 km to 30 km. The ultimate objective is to arrive at a predictive capability regarding plant uptake, ecosystem cycling and estimated exposure to man of TU elements which are emitted by a separations plant. Greenhouse studies of concentration ratios and the agricultural plots should produce estimates of root uptake and foliar deposition. Natural plant communities show concentration correlations with distance over 1-10 km for soil samples and vegetation samples.

F. W. Whicker (Colorado State University) had been conducting a field study at Rocky Flats for the past three years. His study site is in a generally southeastern direction, downwind of the area contaminated by drum storage of cutting oil.

The site has been subject to deposition of Pu due to wind resuspension from the drum storage area before asphalting and small additional deposition due to resuspension from pockets of contaminated soil between the asphalt pad and the study site. There is also some small chronic release from the plant ventilation stack.

The objectives of the study have been to

1. Define the distribution of ^{238}Pu and $^{239,240}Pu$ throughout the various ecosystems components.

2. Determine the mechanisms and rates of transport through soil block transplants between contaminated and uncontaminated areas, soil erosion studies, and large mammal transport (deer and cattle).

3. Develop a predictive model which will probably have to be empirical and site specific.

The results to date show that Pu is widely distributed due to various source terms and that results in all compartments show large coefficients of variation because of the particulate nature of the contaminant (CV range from 100 to 600 percent). The data are not normally distributed and less than 25 percent are log normally distributed. Concentration ratios (plant/soil) average about 2×10^{-2} and half of this is due to root uptake as indicated by soil block transplants. Concentration factors in herbivores are about 10^{-3} and in carnivores about 10^{-5}.

R. Gilbert (PNL) discussed possible statistical approaches to the treatment of field data for the TU elements. These data are usually highly variable and require careful approaches to sampling design and application of statistical techniques. He is funded by BER to assist contractors who may need help in the setting of experimental and sampling protocals.

W. E. Martin (Battelle-Columbus) described his efforts to model the NTS contamination sites for human hazard from Pu. The conceptual model is fairly easily developed but the values for transfer coefficients

(i.e., loss rate constants) are still not available. He thinks that the assumption that all of the components reach a steady state is an over simplification and that closed system equilibria are better approximations in many compartments because of the persistence of the TU elements.

D. G. Sprugel (ANL) discussed the studies on the Miami River Watershed where measurable ^{238}Pu can be detected from Mound Laboratory. Liquid wastes are released twice weekly into the Miami River at less than the Radiation Protection guide level. Some low level chronic release occurs through the stack. In addition, a 1967 spill at the plant caused some contamination in the canal and ponds below the plant.

The routine liquid waste release has made an interesting pulse of ^{238}Pu to follow downstream. This can be followed to determine the kinetics of uptake in suspended particles and aquatic plants.

Fish have been transplanted into the contaminated ponds and then are removed to uncontaminated water to study loss rates.

Terrestrial studies in the environs are dealing with essentially fallout background levels but the $^{238}Pu/^{239}Pu$ ratios are higher so that an identification of local source can be made. A small watershed will be identified and studied for Pu movement. Comparison of disturbed and undisturbed fields will be part of this study.

G. Potter (EPA-Las Vegas) presented the program on ruminant studies at NTS. Range cattle are assayed for physical/biological transport across organs and placentas and fistulated steers are monitored to determine source and amount of TU elements in diet. Cows are fed Cm and Np in uptake and milk transfer studies funded by DBER-ERDA.

F. G. Lowman (Nuclear Regulatory Commission) discussed the work which the Puerto Rico Nuclear Center did at Bikini Atoll. Measurements showed that sea water to normal sediments ratios were 6×10^{-5} but for anoxic sediments the ratio was 3×10^{-6}. The distribution of Pu in the water has been shown to be current driven. The concentration ratios for plankton are dependent upon the size of the plankton population and ranged from 2×10^{4} up to 5×10^{5}. The change of Pu concentration in water is faster than one would expect by physical parameters alone. Plankton tend to favor ^{238}Pu over ^{239}Pu, but ^{238}Pu is discriminated against by filter feeders but favored by those organisms that have membrane transport systems.

B. G. Bennett (Health and Safety Laboratory) outlined the research and monitoring program at HASL. A variety of radionuclides including ^{238}Pu and $^{239,240}Pu$ are measured in air samples. These concentrations have generally declined since 1963 but Chinese tests in 1973 increased 1974 values threefold. These historical data have been used to calculate an average plutonium body burden and this estimate agrees with the limited information on tissue samples analyzed at LASL.

Diet analyses from New York grocery stores indicate that shell fish are the highest source for ingestion (0.011 pCi/kg). A comparison of calculated results used for inhalation and ingestion show inhalation is 1000 times more important in contribution to body burden than is ingestion of the food in the survey.

An experiment which will assay the plutonium concentration in vegetables and the soil in which they are grown is in progress. The source of plutonium is world wide fallout which is in very low concentration in the soil.

Soil samples from a soil profile on Cape Cod are routinely taken and analyzed to observe the downward movement of Pu.

Vaughan Bowen (Woods Hole) compared the observations obtained in Lake Ontario and the Atlantic Ocean. Americium is leached differentially from Pu in Lake Ontario but not in the marine environment. The ^{137}Cs observed in the lake is not moving similarly in Lake Ontario.

Plutonium correlation with depth holds for both the North Pacific and North Atlantic environments. He also noted that Fe is sinking much faster than Pu in the marine environment.

A study of the spatial distribution and isotopic ratios indicates that the major input of Pu to Lake Ontario is from the Niagara River.

Wally Weimer (PNL) discussed the results of work performed by Dave Robertson. Study areas for this research are the Marshall Islands and Thule, Greenland. The recent results have been from Thule where the accident site has been studied in a follow-up survey. These indicate that detrital materials are the major Pu reservoir; the source term appears to be 25-30 Ci; limited biological activity redistributes the Pu; the highest ocean water concentrations were at 5m above the bottom near the accident site and mostly particles; and surface water concentrations indicate world-wide fallout levels of Pu.

Dave Hayes (SRL) discussed a comparative study of estuarine environments which he is doing in cooperation with F. A. Cross. Their results show that Pu at the mouth of the Savannah River totals about 55 Ci which can be attributed mainly to fallout (6 percent of the alpha activity is from ^{238}Pu) within the river's watershed while the Savannah River Plant effluent contributed about 0.3 Ci of Pu (80 percent of the alpha activity is from ^{238}Pu). At the mouth the freshwater sediments contained about 5 fCi/g while there were about 40 fCi/g in bay water sediments. In general, Pu concentrations in the various environmental components of the Savannah River estuary were not higher than reported values for other coastal marine areas.

H. J. Simpson (Lamont-Doherty Geological Observatory) has used ^{137}Cs to indicate samples expected to have Pu in a Hudson River survey. In a series of sediment cores he observes that the Pu concentrations can be related to the fallout input and not to Indian Point reactor. However, about 50 percent of the ^{137}Cs is ascribed to the reactor.

W. R. Schell (University of Washington) described his work at Bikini Atoll where his results indicate that the circulation pattern is wind driven. Most of the Pu is in the northwest quadrant of the sediments with a maximum concentration of 120 pCi/g. The maximum deep water concentration is 240 pCi/m^3.

V. E. Noshkin (LLL) has measured the Pu and Am concentrations in the soils and aquifers at Bikini and Enewetak Atolls. In addition, he has done extensive sampling of the lagoons. He states that the terrestrial food chains are more important than marine food chains for purposes of human health hazards evaluations.

Plutonium is moving through the ground water at Bikini with detectable quantities 80 meters below the surface. There seems to be a linear correlation between Pu in surface soils and ground water.

A study of coral growth increments for Pu isotopes revealed that ^{239}Pu now seems to be a constant value in yearly fallout. Ratios of ^{241}Pu and ^{240}Pu to ^{239}Pu have maintained a constant ratio in the past years. However, $^{238}Pu/^{239}Pu$ ratios are highly variable.

Maurice Wahlgren (ANL) has studied the distribution of Pu in Lake Michigan food chains and finds the following average concentrations:

10^{-4} pCi/kg_{wet}	in surface waters
10^{2} pCi/kg_{wet}	in sediments
10^{-1} pCi/kg_{wet}	in zooplankton
10^{-3} pCi/kg_{wet}	in carnivorous whole fish

The indications from sediment and water sampling are that the plutonium in surficial sediments is re-equilibrating with lake waters and is exported to Lake Ontario. Inventory of fallout Pu in Lake Michigan is about one third of that expected of a closed system. It is believed exportation and the low concentration in the incoming waters result in this reduction of inventory.

R. M. Emery (PNL) has been studying the redistribution and movement of Pu in a 30-year old waste pond. There are about 15 biological components in the pond and these are sampled and analyzed for ^{238}Pu and $^{239,240}Pu$. The sediments average about 170 pCi/g for ^{238}Pu and about the same for $^{239,240}Pu$. Field and laboratory experiments are conducted to determine the kinetics of uptake within biological components.

L. D. Eyman (ORNL) is conducting research at White Oak Lake which is similar to Emery's studies. In this case, however, Am and Cm, as well as Pu, are being studied. Tracer studies with ^{237}Pu are incorporated in this work in controlled experiments. The results are applied to a 12 compartment model for preliminary dose assessments.

E. D. Goldberg (Scripps) mentioned his work in establishing the history of pollutants in coastal marine environments. Rates of sedimentation and the age of sediments have been determined by stratagraphic and radioactive dating techniques. Plutonium increased to a maximum of 1000 dpm/kg in 1964-1965 along the California coastline. On the east coast detected levels increased to 100 dpm/kg in the same time period. He has observed that reducing environments in the ocean sediments may have four to five mCi/km^2 which is a higher concentration than on land at the same latitudes. In general, Pu is rather immobile in the marine environment and appears to be moved to lower depths in sediments by burrowing organisms.

T. R. Folsom (Scripps Institute) described his work on Pu concentrations in giant brown algae. Apparently, this accumulation is a surface phenomenon and this is demonstrated by exposing paper and other surfaces to similar environments and accumulating the same surface concentrations. His work on marine food webs shows that Pu does not concentrate as it moves up the food chain.

PARTICIPANT LIST

WORKSHOP ON ENVIRONMENTAL RESEARCH
FOR THE TRANSURANIUM ELEMENTS

November 12-14, 1975

Held at the Battelle Seattle Research Center

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REPORT OF PLANT PANEL

The plant panel addressed the following topics in their discussions of incorporation of transuranic (TU) elements by plants.

1. Uptake of Pu and Am by rooted plants from soils contaminated by transuranic elements.

2. Mechanisms responsible for uptake by rooted plants.

3. Translocation and relative concentrations of Pu in different organs, tissues, and cells of plants.

4. Mechanisms of contamination of foliar surfaces of terrestrial plants.

5. Measurement of fractional movement of transuranic elements from soil to plants.

6. Analytical chemistry of transuranic elements in biological matrices.

Topics 4 and 5 were evaluated in considerable detail with respect to (a) definition of terms for expressing uptake, (b) magnitudes of uptake by plants growing in contaminated substrates, (c) factors influencing uptake by the root pathway, (d) modes of contamination of foliar surfaces, and (e) format for reporting results.

Definition of terms

Numerous terms have been used to describe relationships and distributions of TU elements in plants and soil. One convenient expression is the ratio of concentration in plant material to that of substrate or reference material commonly defined in terms of

$$\frac{\text{Act/wt of plant part}}{\text{Act/wt of substrate or reference material}}$$

Because the expression represents a simple ratio of concentration, the plant panel recommends use of the simple term, concentration ratio (CR), to describe this relationship. The plant panel proposes that CR terminology be used to express concentration ratios in plants and soil in place of terms such as discrimination factor, uptake coefficient, accumulation ratio, distribution ratio, etc., - all of which are based on the above definition.

Another relationship requiring convention definition is the ratio of activity on an area basis. The plant panel proposed use of the term,

$$\text{Inventory Ratio (IR)} = \frac{\text{Activity/unit area in product}}{\text{Activity/unit area in source}}$$

This ratio represents distribution or budgets of activity in definable components of crops and ecosystems.

Transfer coefficient (k) was defined as fraction of TU element transferred per unit time. The expression, k, would represent

$$k = \frac{\text{fraction TU}}{t} \text{ or } k = \frac{\text{fraction TU}}{\Delta t}$$

depending on attributes of the data set. Where such information is available, the kinetics of transuranic mobility can be described by this convention.

Ratio of concentration of Pu and Am in plants and soil

Participants of the panel summarized data on CR's which they had gathered or with which they were familiar. The CR's are summarized in Tables 1, 2, and 3, respectively, for Pu values from pot culture, Pu values from field studies, and Am values from pot culture and contaminated field sites. Results were summarized on differences in CR's between leaf-stem vs seed-fruits (Table 4). The CR results exhibit considerable variability (a range of eight orders of magnitude). Factors of 10^1 to 10^3 differences between foliage and fruits were evident. Direct deposition and surface contamination accounted for at least 90 percent of foliar concentrations of Pu when CR's approached 10^0. For cases of limited comparison, CR's for Am appear to be at least 10^2 to 10^3 higher than for Pu. Much of the remaining variability was attributed to soil factors and source characteristics, but the cause and effect relationships for all TU elements are not clearly understood. The following source characteristics and soil processes likely play a role in the uptake of Pu by plants:

1. Weathering or depolymerization of Pu-oxide or polymeric forms.

2. Formation of complexes involving carbonates or organic matter.

3. Transformations by soil organisms (microflora).

4. Biotic influences related to root metabolism, degradation, and recycling of transformed products.

5. Oxidation state of Pu.

6. Soil fertility and pH.

Discussions with the soil-sediment panel on the role of these processes on the uptake of Pu by plants reinforce the fact that insufficient information exists to explain the wide range of CR's. Further, it was pointed out that little is known about factors such as complexing agents (naturally occurring organic acids, chelators used in trace element amendments to soil) and their effects on the availability of TU elements to plants. For example, microorganisms produce citric and oxalic acids, known Pu chelators, and these complexers may affect the long-term availability of TU elements. It was emphasized in these discussions that an understanding be obtained on the soil mechanisms affecting availability and uptake of TU elements in order to extrapolate limited experimental data to a variety of potential contamination events, to different source term characteristics, to diverse ecosystem cycling processes, and to assessments of consequent hazard to man.

Contamination of foliar surfaces

The state of information was reviewed on interactions involving TU elements at the air-plant interface. It appears that initial contamination by deposition on foliar surfaces is governed by particle aerodynamics and characteristics of the plant surface. Although difficult to generate controlled particle size distribution, deposition of carrier particles is predictable within a factor of 5 if aerodynamic variables are known. Particulate distributions are best characterized in terms of AMAD. Typical mass loadings of Pu aerosols or carrier particles in different environments would aid predictions of deposition.

For size classes of aerosols and particles likely to be associated with releases from fuel reprocessing facilities, the panel concluded that limited information is available on retention and translocation of surface deposited transuranic substances. Although much of the existing literature is ambiguous with respect to submicron size particles, retention is likely governed by behavior of particulate carriers, although surface adsorptive phenomena are not understood. Likewise, limited information is available on translocation (absorption at site of deposition and transport to other plant tissues) of transuranic substances deposited on foliar surfaces. Results from wind tunnel experiments suggest that foliarly deposited Pu is available for translocation from leaf to root and seed tissues. CR values for these tissues ranged from 10^{-6} to 10^{-3} following foliar contamination with fresh ($0.4\mu m$) and aged ($0.1\mu m$) $^{238}PuO_2$. Other evidence indicates high translocation rates if Pu enters leaf tissues in ligand form.

Resuspension of contaminated soil by wind and concomitant deposition on foliar surfaces primarily involves particles greater than 1μm in diameter. The panel concluded that magnitudes of resuspension for different environments are not well understood. Likewise, the degree of direct foliar contamination by raindrop splash is not known.

Format for reporting results

While formats for reporting results are frequently a function of experimental objectives, thus they remain the prerogatives of respective investigators, the plant panel recommends inclusion of minimal supplemental information for subsequent use for comparative analyses, hazard assessment and modeling. If possible, results should be reported on a dry weight basis, and a dry wt/wet wt ratio should be given. Exposure geometry (pot size, quantity of soil, conditions of amendment to soil, leaf-area ratios, etc.) should be described. Yield (harvested weight per pot or plot), standing crop of biomass (weight per unit area), and bulk density of soil are additional parameters to be reported in conjunction with concentration ratio data.

Research Needs

1. Determine why CR's for pot culture and field studies differ by factors of at least 10 to 100 for Pu.

2. Achieve a better understanding of the soil and plant factors influencing the uptake of transuranic elements by plants. Questions on the role of soil properties involve potentially increased or decreased availability as a function of time as a result of polymerization or depolymerization, formation or breakdown of complexes, and fertility influences. With respect to plants, age, species differences, physiological stress, root-mycorrhizae may influence the availability of TU elements.

3. Determine the factors that influence retention of small particles on foliar surfaces. Mechanisms and routes of entry to internal tissues of foliar contaminants need delineation. Chemical transformation, translocation, and accumulation of TU elements absorbed by the foliar pathway need to be determined for foliar modes of contamination.

4. Develop better methods for distinguishing between TU elements assimilated by foliage via the root pathway vs TU elements deposited on foliar surfaces.

5. Understand the physiological mechanisms (passive, coupled transport, pinocytosis) governing root uptake of Pu in order to extrapolate among the diverse plants and to predict uptake by vegetative components of ecosystems on a generic basis.

6. Develop numerical data on plant-soil relationships for Np, Am, and Cm and consider relationships specified in items 1-4 above.

7. A plant sample, possessing uniform or certified quantities of isotopes of Np, Pu, Am, and Cm is needed for interlaboratory comparison of radiochemical analysis. Reference material could be prepared and distributed possibly by the Bureau of Standards or the IAEA.

8. Establish field sites in different climatic regimes for long-term studies of TU uptake by native and crop plants. Given the wide range of observed CR's, the combined effects of time, soil chemistry and ecological processes on availability of TU elements to plants need to be resolved.

9. Another question needing investigation is the influence of plant biochemical transformations that may cause different nutritive characteristics of TU elements with respect to animal metabolism. Once reacted biochemically in plant cells, is the animal digestability of TU elements increased or decreased?

Table 1. Plutonium concentration ratios for plants grown in potted soil

Plant	Range of CR's	Conditions	Rapporteur
Ladino clover	10^{-5} to 10^{-4}	In 120 kg of NTS soil; 70 n Ci g^{-1} soil; CR's increased by seven times in 5 years.	Romney
Alfalfa	10^{-5} to 10^{-4}	In 3 kg of NTS soil; 0.6 nCi g^{-1} soil; highest CR's involve chelate treatment.	
Barley			
Straw	10^{-5} to 10^{-3}		
Grain	10^{-6} to 10^{-3}		
Soybean		In 3 kg of NTS soil; 5 nCi g^{-1} soil; highest CR's involve chelate treatment.	
Forage	10^{-4} to 10^{-3}		
Bean	10^{-6} to 10^{-4}		
Barley			Schulz
Grain	10^{-7}	NTS soil; high-fired Pu oxide; 10 to 50 nCi g^{-1} soil.	
Leaf	10^{-5}		
Wheat			
Grain	10^{-8} to 10^{-6}	Pu-chloriade and Pu-nitrate (^{239}Pu at 0.5 Ci g^{-1}).	
Leaf	10^{-6} to 10^{-3}		
Barley			
Leaf and stem	10^{-5} to 10^{-3}	Pu-nitrate (^{238}Pu and ^{239}Pu) 10 Ci g^{-1}.	Wildung
Grain	10^{-7}		

Table 2. Plutonium concentration ratios for plants in field contaminated sites

Plant	Range of CR's	Conditions	Rapporteur
Wheat		Within 300 m of reprocessing plants; 1 pCi g^{-1} soil; represents greater than 90% surface contamination from aerial deposition.	Adriano
Straw	10^{-2}		
Grain	10^{-3}		
Native vegetation	10^{-1} to 10^{0}		
Native vegetation		Flood plain contaminated for 30 years; 25 to 100 pCi g^{-1}; evidence for Pu-organic complex; suspect monomeric forms of Pu; nominal surface contamination.	Dahlman
Tree foliage	10^{-3}		
Herbaceous	10^{-3}		
Soybean leaf	10^{-4} to 10^{-3}		
Millet	10^{-4} to 10^{-3}		
Bush bean			
Leaf	10^{-3}		
Bean	10^{-4} to 10^{-3}		
Native vegetation			
Foliage	10^{-4} to 10^{-1}	Enewetak; soil pH = 8	Koranda
Coconut	10^{-3}		
Native perennials		NTS safety shot; resuspension and surface contamination suspected on hirsute plants.	Romney
Shrubs	10^{-2} to 10^{0}		
Mixed native grasses	10^{-2}	Metallic Pu in oil at RF; 1 nCi g^{-1}; root and foliar surface contamination contribute equally.	Whicker
Transplanted soil blocks			

Table 3. Americium concentration ratios for plants grown in potted soils and contaminated field sites

Plant	Range of CR's: Soil	+ Lime	+DTPA	+Lime and DTPA	Conditions	Rapporteur
Pot culture						
Corn						
Soil pH = 6	10^{-2}		10^{-1}		10 nCi g^{-1}; chelate applied to soil.	Wallace
Soil pH = 8.5	10^{-1}		10^{0}			
Barley						
Soil pH = 7.5	10^{-1}		10^{0}			
Soil pH = 6	10^{-1}		10^{0}			
Bush bean						
Soil pH = 7	10^{-1}		10^{1}			
Bush bean						
Sand texture	10^{-1}	10^{-1}	10^{0}	10^{+1}	2 nCi g^{-1}; chelate added to Am stock.	Adriano
Clay texture	10^{-1}	10^{-1}	10^{-1}	10^{+1}		
Corn						
Sand texture	10^{-1}	10^{-1}	10^{-1}	10^{0}		
Clay texture	10^{-1}	10^{-2}	10^{-1}	10^{0}		
Barley						
Leaf	10^{-4}				Contaminated soil from NTS areas 11, 13; 3 kg pot^{-1}.	Schulz
Grain	10^{-6}					
Wheat grain	10^{-7} to 10^{-5}					
Alfalfa	10^{-4} to 10^{-3}				Am-sol'n added to soils; 3 kg pot^{-1}.	
Barley					In 3 kg NTS soil; 0.6 nCi g^{-1} soil; highest CR's involve chelate treatment.	Romney
Straw	10^{-5} to 10^{-3}					
Grain	10^{-5} to 10^{-3}					
Soybean						
Forage	10^{-3} to 10^{-1}				In 3 kg NTS soil; 5 nCi g^{-1} soil; highest CR's involve chelate treatment.	
Bean	10^{-4} to 10^{-2}					
Field studies						
Native perennial shrubs	10^{-2} to 10^{0}				NTS safety tests surface contamination suspected	Romney
Native plants	10^{-2} to 10^{0}				Enewetak	Koranda

Table 4. Difference factors of Pu concentration ratio for leaf-stem vs seed-fruit

Plant	Seed-fruit CR's relative to leaf-stem	Condition	Rapporteur
Soybean	0.01 to 0.001 lower	Pot culture	Romney
Barley	0.01 lower	Pot culture	Schulz
Barley	0.02 to 0.01 lower		
Barley			
Soybean	0.01 to 0.001 lower	Pot culture	Wildung
Peas			
Barley	0.01 lower	Field lysimeter	Cline
Cheatgrass			
Bush bean	0.1 lower		
Millet	0.5 lower	Garden plot	Dahlman
Wheat	0.1 lower	Field crop	Adriano

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REPORT OF SOILS AND SEDIMENTS PANEL

Summary

Soils and sediments do or will soon provide the principal locations in which the transuranium elements will reside over geologic time. Of principal concern is the understanding of the processes that affect the mobilization or immobilization of transuranium elements once they enter the soil or sediment. To evaluate our current understanding of the processes influencing the interaction of plutonium with soil and sediment, the panel summarized current knowledge of the various factors that affect the interaction, including source term, medium, redistribution processes, reservoirs, and general areas requiring further study. The summarization suggested considerable general information is available; research, currently in progress, is aimed at developing needed information; and a limited amount of general relationships exist to estimate the rate materials are currently being redistributed on the earth's surface. The panel recommends that additional information be obtained on transuranium chemistry in dilute solutions, the role of complexing agents on the mobility and availability of transuranics be clarified, and suitability of tracer studies for characterizing the behavior of transuranics under environmental conditions be confirmed.

Discussion

The general objective of this panel was to determine the relationship between physical, chemical, and biological factors affecting the movement of transuranium radionuclides in the soil system in order to answer the question, "What fraction of the total inventory of transuranics will reside in man as a function of time?" Table I outlines the areas of interest and provides a general statement of current knowledge. Table II elaborates on the problem areas. Table III lists the panel participants. Table IV lists the heavy element activities in nuclear fuels as a guide to future transuranics. Table V lists heavy element activities for a 3.3 percent enriched ^{235}U and a 3.5 percent enriched ^{239}Pu charge. Table VI lists ^{238}Pu fate from space program.

General Areas Requiring Further Study

1. Suitability of tracer studies in characterizing real world situations.

2. Plutonium behavior in extremely dilute solutions.

3. Modeling of environmental systems using existing values for estimating ecosystem responses.

4. Role of chemistry and physics in making plutonium available or unavailable for transport.

5. The lack of information on Pu concentration variability in the soils and sediments component of the environment was identified as an important problem area related to the design and interpretation of data from experimental and field studies. Specific emphasis should be placed on characterizing the components of this variation in a variety of contaminated environments in order to better define the level of sampling effort required to detect statistically significant differences in Pu distributions and transport.

Table I

Source	
1. Fallout (SNAP and Weapons)	Amount well known; chemical and physical form poorly characterized in available literature - likely to be heterogeneous - makes interpretation of experiments difficult.
2. Reactor and Reprocessing (Liquid and Atmospheric)	Amount known; behavior and character poorly defined at present - under study (atmospheric).
3. Burial Ground (Solid Wastes)	Amount poorly known; diverse chemical and physical forms, makes interpretation of experiments difficult.
Medium	
1. pH, CEC, Particle Size, Mineralogy	Techniques available, relationships to movement and uptake not clear. Information on K_d vs pH, Pu vs particle size, etc., known on some soils. These characteristics are variable among sites.
2. Ion Content (Salts, Organic Complexing agents)	Complex and roles not clearly defined, under study.
3. Pu, Am, Cm Behavior in Dilute and Soil Solutions	Are transuranics behaving as polymers, radiocolloid, or colloidal aggregate - unknown and important.
Redistribution Processes	
1. Erosion	Magnitude on gross scale well known; site specific studies underway; principal mechanism moving transuranics.
2. Biotic (Recycling)	Effect of cycling through biological material on mobility and availability to plants and man is unknown; inventory can be estimated (small).

Table I (Continued)

3. Resuspension	Large amount of information available; important role in transuranics to man.
4. Diffusion	Small effect over short term, but likely important in availability to plants; under study.
5. Freezing and Thawing	Small effect.
6. Wetting and Drying	Important processes in soil that shrink and swell.
7. Man's Activities	Important factor - highly diverse.
Reservoirs - Short and Long Term	
a. Soil	Relative magnitude of each can be defined and rate material is moving from one compartment to a second compartment estimated.
b. Sediments (Marine and Freshwater)	
c. Water (hydrosphere)	
d. Atmosphere	
e. Plants, Animals (Biosphere)	

Table II

SPECIFIC RESEARCH QUESTIONS

1. How does weathering of small particles that reduces initial resuspension by three orders of magnitude occur? Humid area values are needed since magnitude of reduction and time dependence of reduction are known only for arid environments.

2. Evaluate the use of defined microsphere sizes to determine availability of plutonium to plants as function of particle sizes.

3. What characteristics do different source types (fallout, stack effluent, liquid effluents) have in common initially or after introduction to the soil and sediment that would permit generalizations to be drawn concerning their behavior in the environment?

   a. What are the nature and behavior of the agglomerate species where the source is (1) plutonium dioxide (refractory and/or low temperature), (2) hydrous plutonium oxide ($PuO_2 \cdot XH_2O/Pu(OH)_4$), (3) formation of polynuclear plutonium hydrolytic colloids ("polymer") formed by increasing pH, (4) formation of sorption colloids on host colloids naturally existing.

   b. Characterization of particle size and state of aggregation of transuranium elements in soil.

4. Resuspension component on agricultural plants is poorly defined as to magnitude, particle size, segregation in atmosphere, dust load and translocation in the plant.

   a. The determination of the range of resuspension in non-desert areas and the change in these factors with time and downward movement.

5. Are the differences in the environmental behavior of plutonium-238 and -239 source term related or due to fundamental differences in the radioisotopes?

6. Summarize the values from transport studies of plutonium in soil and sediment systems and perform sensitivity analyses to evaluate the relative importance of the various mechanisms.

   a. What are the mobilities of ^{238}Pu and ^{239}Pu in soils and sediments?

Table II (Continued)

b. What are the physical mechanisms in plutonium transport?

c. Determination of diffusion coefficients for transuranics as indicators of mobility of these elements in soil aquatic environments.

7. Improved analytical techniques are needed to determine the chemical form of plutonium in water-soil solutions and with complexing agents as well as for determining particle sizes over range of interest from small colloids and up.

   a. Techniques for measuring the dissolution rates of plutonium from plutonium-oxide microspheres - in soils, chelates and in soil solutions.

   b. Characterization of chemical species involving plutonium.

   c. Plutonium valence state in the environment.

   d. Chemical species of plutonium that are stable in a soil/water environment.

8. Evaluation of the importance of recycling of organic matter on subsequent availability to plants and interaction with soil.

9. Increased emphasis on Am and Cm behavior under environmental and laboratory conditions.

   a. Comparative studies on the behavior of americium and curium in the environment from presently contaminated areas.

10. Physico-chemical associations of Pu in soils and how chemical or biological mechanisms affect these associations.

    a. Extraction procedure for plutonium adsorbed on soil surfaces.

    b. Interaction of clays and organic matter on plutonium adsorption and diffusion.

    c. Type of sorption (physical and/or chemical) as a function of type of soil and the chemical form of plutonium.

Table II (Continued)

d. Mechanism for plutonium sorption.

e. Continued emphasis on behavior of plutonium in different kinds of soils - plant uptake, discrimination factor, solubility and movement.

f. Analytical and experimental methodology for extracting "non total" plutonium with various solutions from the soil.

g. Reliable leaching agent to correlate with plant available forms of transuranics.

h. Role of iron, aluminum, manganese oxides in the mechanisms of transuranic adsorption.

11. Measurement of precise, meaningful distribution ratios of Pu, Np, Am, and Cm so that they will have high predictive value.

    a. Determination of factors that influence distribution ratios for transuranics, such as:

        (1) Polymerization under environmental conditions

        (2) Radiocolloid formation

        (3) Aging

        (4) Adsorption and complex formation with soil organic material.

12. Role of organisms in sediments for redistributing plutonium.

Table III

Panel Members

N. F. Barr, ERDA

E. A. Bondietti, ORNL

D. A. Brown, U. of Arkansas

E. A. Bryant, LASL

J. C. Corey, SRL

D. Elle, ERDA

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T. Garland, PNL

T. E. Hakonson, LASL

B. Lotz, ERDA

F. J. Miner, Rocky Flats

H. Nishita, UCLA

J. Patterson, LASL

R. Perkins, PNL

J. F. Relyea, U. of Arkansas

D. R. Rogers, Mound Laboratory, MRC

J. C. Sheppard, Washington State University

D. Sprugel, ANL

T. Tamura, ORNL

B. Wachholz, ERDA

TABLE IV

ACTIVITY OF HEAVY ELEMENTS IN ACCUMULATED WASTE FOR ENTIRE NUCLEAR INDUSTRY

Normalized to 1 for ^{239}Pu

Isotope	1972	1974	1976	1978	1980	1982	1986	1990	2000
^{237}Np	0.22	0.21	0.22	0.21	0.20	0.18	0.15	0.14	0.076
^{239}Np	11.5	11.2	11	10.8	14.5	26.5	43.1	31.7	7.84
^{236}Pu	0.00092	0.00076	0.00075	0.00067	0.00056	0.00061	0.0036	0.010	0.012
^{238}Pu	36.1	44.1	49	50	55.6	80.6	167	283	361
^{239}Pu	1	1	1	1	1	1	1	1	1
^{240}Pu	1.54	1.65	1.7	0.58	1.89	2.55	4.45	5.33	3.14
^{241}Pu	315	306	300	287	295	337	403.3	367	255
^{242}Pu	0.0043	0.0043	0.0044	0.0042	0.0047	0.0065	0.0099	0.011	0.011
^{243}Pu	--	--	--	--	--	--	--	--	--
^{241}Am	96.1	95.3	96.5	93.6	103	137	178	141	77.2
^{242m}Am	5.69	5.64	5.5	5.4	7.23	12.2	19.5	14.2	5.68
^{242}Am	5.69	5.64	5.5	5.4	7.23	12.2	19.5	14.2	5.68
^{243}Am	11.5	11.2	11	10.8	14.5	26.5	43.1	31.7	7.84
^{242}Cm	5637	3887	3020	2255	2624	3876	3183	953	278
^{243}Cm	2.31	2.23	2.25	2.16	2.34	3.16	4.03	3.00	2.16
^{244}Cm	1561	1511	1510	1427	2441	5671	9813	6464	919
^{245}Cm	0.23	0.22	0.23	0.22	0.50	1.43	2.64	1.92	0.33
^{246}Cm	0.046	0.045	0.046	0.046	0.072	0.26	0.49	0.35	0.063
^{247}Cm	--	--	--	--	--	--	--	--	--
TOTAL CURIES	7.2×10^5	3.9×10^6	1.4×10^7	3.0×10^7	7.9×10^7	2.7×10^8	9.3×10^8	1.2×10^9	1.8×10^9

TABLE V

HEAVY ELEMENT ACTIVITIES IN NUCLEAR FUELS*

(Normalized to 1 for ^{239}Pu)

	3.3% Enriched ^{235}U		3.5% Enriched ^{239}Pu	
Isotope	Charge	Discharge	Charge	Discharge
^{234}U	23.2	.0023	.00022	.00038
^{235}U	1	.000052	.00001	.00001
^{237}Np		.001	--	.00014
^{238}U	4.55	.00086	.00022	.00047
^{238}Pu		8.62	8.84	21.0
^{239}Pu	--	1	1	1
^{240}Pu		1.49	1.46	2.45
^{241}Pu		324	291	662
^{242}Pu		.0043	.0042	.016
^{241}Am		.264	.762	1.26
^{242}Am		.0127	--	.072
^{243}Am		.055	--	.276
^{242}Cm		114	--	668
^{243}Cm		.017	--	.129
^{244}Cm		8.05	--	73.1
^{245}Cm		.00087	--	.015
^{246}Cm		.00019	--	.003

*PWR, Power = 30 MWT Burnup = 33,000 MWDT.

**Normalized to 1 for ^{235}U.

TABLE VI

SPACE PROGRAM

System	Launch Date	Planned Earth Orbit Ci ^{238}Pu	Abort Ci ^{238}Pu	Earth Escapes Ci ^{238}Pu	Fuel Form	Comments
1. SNAP 3	6-61	1800			Metal	1000 yr orbit
2. SNAP 3	11-61	1800			Metal	1000 yr orbit
3. SNAP 9	9-63	17,000			Metal	1000 yr orbit
4. SNAP 9	12-63	17,000			Metal	1000 yr orbit
5. SNAP 9	4-64		17,000		Metal	Burnup in atmosphere
6. SNAP 19	5-68		34,000		Microspheres	Recovered from channel
7. SNAP 19	4-69	37,600			Microspheres	3000 yr
8. SNAP 27	11-69			44,500	Microspheres	On lunar surface
9. SNAP 27	4-70		44,500		Microspheres	Impacted in Pacific
10. SNAP 27	1-71			44,500	Microspheres	On lunar surface
11. SNAP 27	7-71			44,500	Microspheres	On lunar surface
12. SNAP 27	1-72			44,500	Microspheres	On lunar surface
13. Pioneer F	3-72			80,000	PMC	Escaped solar system
14. Transit	9-72	24,000			PMC	1000 yr orbit

TABLE VI

SPACE PROGRAM

System	Launch Date	Planned Earth Orbit Ci ^{238}Pu	Abort Ci ^{238}Pu	Earth Escapes Ci ^{238}Pu	Fuel Form	Comments
15. SNAP 27	12-72			44,500	Microspheres	On lunar surface
16. Pioneer G	4-73			80,000	PMC	Escaped solar system
17. Viking 1	8-75			41,200	PMC	Planned Mars landing
18. Viking 2	9-75			41,200	PMC	Planned Mars landing
19. Les 8/9	2-76	288,000			PPO	Planned 100,000 orbit
TOTALS		387,200 Ci	95,900 Ci	464,900 Ci		948,000 Ci

REPORT OF ECOSYSTEMS AND FOOD CHAINS PANEL

Publication of Results and Dissemination of Present Data

The Panel was impressed with the expansion of research effort in transuranium elements that has occurred since the Plutonium Workshop held at Estes Park, Colorado, on July 11-12, 1974. However, there appeared to be some duplication of efforts within the overall program which indicated that continuous encouragement should be given to investigators to publish results in the open literature, in order to communicate their progress. One means of doing this would be to initiate the drafting of a publication that would summarize the available information on transuranic nuclides in the environment. The Panel discussed this at length and proposed that a book (or other suitable document) entitled "Transuranic Elements in the Environment," similar to the NAS-NRC publication (1971), "Radioactivity in the Marine Environment," be prepared under ERDA-DBER auspices. An initial outline was suggested as follows:

(Book Outline)

A. Title: Transuranic Elements in the Environment

B. Preface

1. Objectives of book
2. Caveats

C. Introduction

1. Restate and extend objectives of BER research program

D. Part A

1. Source Terms
2. Inventory and Distribution

a. Chemical analytical procedures
b. Physical analytical procedures

E. Part B

1. Physical Transport Mechanisms

a. Atmospheric
b. Hydrologic

2. Biological Transport Mechanisms

    a. Terrestrial
    b. Semi-aquatic (sublittoral zones)
    c. Aquatic

3. Consequences

    a. Ecological
    b. Human

F. Statistics and Experimental Modeling

1. Limitations
2. Overkill
3. Case histories

G. Monitoring Design

1. Radiological
2. Ecological

H. Future Needs

1. ERDA input

The Panel further recommends that a select committee be formed to determine principal authors of various sections and chapters, or to see that they are reviewed by certain people or subcommittees. Divergent opinions should be incorporated wherever plausible, as was done in "Radioactivity in the Marine Environment," so long as the differences are mainly philosophical and deal with common bases of fact. Emphasis should be on new material and what needs to be done.

The Panel felt that investigators would be motivated to publish their results through either (1) summarizing and synthesizing material for inclusion in the book or (2) wanting to avoid being "scooped" by holding results for future publication.

Availability and/or Need of Environmental Information Regarding Transuranic Elements

The Panel felt that environmental investigations were generally lagging behind the need to address current and pending problems. Two immediate

approaches that were recommended to correct this situation were (1) spiking experiments and (2) the establishment of long-term study areas.

Spiking experiments should be conducted with the caveat that they may require that double spiking be done to circumvent mass flow through certain systems which may yield results that are highly influenced by the amount of the spike material. The Panel concurs in the proposal to use chemical analogs for certain studies, but recommends that such studies be carefully done to select the proper material in order to correctly interpret the results and avoid confounding the total picture of environmental behavior of transuranics.

The ERDA-BER concept of National Environmental Research Parks, such as those currently established at Savannah River and the Idaho Nuclear Engineering Laboratory and those proposed at Hanford, Rocky Flats, and Los Alamos, would provide the kinds of areas envisioned as necessary to study long-term behavior of transuranic elements in a variety of ecosystems. The Panel recommends that this concept move forward at all possible speed so that suitable study areas will not be lost and that integrated ecological research directed at programmatic needs of ERDA-BER can be conducted in the time frames needed to provide the necessary information.

Clarification of Rate Constants/Transfer Coefficients

The current confusion over which values to use in calculations of food chain transfer of transuranic elements appeared to the Panel to result from incomplete data banks that need input from current investigations. Much information has not been summarized and synthesized and this argues for the proposed publication discussed under Topic I.

These parameters are subject to change with time and environmental influences, which may require a stochastic approach to their treatment. This, in turn, requires a large data base and, perhaps, spiking experiments such as those discussed under Topic II. Dietary information is critical for pathway analysis and will require more realistic transfer rates than are now available. The Panel felt that a starting point could be established by growing plants in transuranics and to feed the biologically-processed radionuclides to animals. Similarly the need for more information on microbial changes of transuranics, thus enhancing their biological availability, would be a logical companion investigation.

Ingestion of many radionuclides, particularly the transuranics is not restricted to food items; intake by way of ingested soil may overwhelm TU intake in forage and may be highly variable by season, location, animal species, and physical/chemical form of the element. This points up the need to optimize parameters in design on the basis of as current a knowledge as possible and to more rigorously define source terms. Values for nCi/g, nCi/m^2, plus soil densities should be provided in order to make proper compartmental estimates. Standard soils should be established and used for uniformity of analytical results and for interlaboratory calibrations.

Several sources of information were identified that may well assist investigators. Gilbert (PNL) and Tamura (ORNL) can make valuable contributions in this general area; Bondietti (ORNL), Boni (SRL), and Wildung (PNL) have available information on chemical forms of transuranics; and Soinski (LFE) has been consulted by Whicker (CSU) about physical forms of transuranics. The Panel recommends that these sources be consulted more often by investigators and that they be incorporated into the proposed publication.

Concluding Remarks

The Panel commends the ERDA-BER for sponsoring this worthwhile Workshop, and felt that a staggering amount of information had been presented representing a great deal of progress in a short period. The first indications of duplication of efforts, sometimes within larger laboratories, prompted the Panel to recommend the publication of a major synthesis of available information and greater communication among the investigators. Coordinated, unified efforts to properly design field studies require much talent; however, the transuranic research program has now progressed to the point that such effort should be made to obtain the needed information as efficiently as possible.

The Panel suggested that a third Workshop on transuranics would probably be appropriate in the Spring or Summer of 1977 and felt that it should be organized more or less along the same lines as this second workshop; there was much more work accomplished in the smaller working groups than at the first such meeting. The meeting length was considered about right, the working groups were logical, and the influence of BER staff was low-profile; whether this last point was cogent to the results of the Workshop is left to your evaluation.

ECOSYSTEMS AND FOOD CHAINS PANEL

Discuss the research needs for ecosystem and food chain transport of the transuranium elements. Problems of experimental design and sampling strategies should be major topics.

Chairman: Dr. Wayne C. Hanson, LASL

B. G. Bennett, HASL

A. L. Boni, SRL

F. A. Cross, NMFS

P. B. Dunaway, NV

R. Gilbert, PNL

F. G. Lowman, NRC

O. D. Markham, INEL

W. E. Martin, Battelle Columbus

J. E. Pinder, SREL

G. Potter, EPA

M. H. Smith, SREL

D. Sprugel, ANL

R. L. Watters, ERDA

F. W. Whicker, CSU

REPORT OF TRANSPORT AND DISPERSION PANEL

Introduction

The assignment of this panel was to identify and assess the research needed to support a quantitative description of the movements of transuranic elements in aquatic ecosystems, both freshwater and marine. During the discussions it became evident that the basic charge should be clarified by specification that the descriptions needed must be extrapolatable for 1000 to 1,000,000 year time spans, and these cycling processes will ultimately have to be evaluated in terms of their relationship to man.

As illustrated in Figure 1, the first steps taken were the identification of a variety of sources from which transuranic elements have been, or may be expected to be, released into aquatic environments, and the classification of aquatic environments into a small number of intercomparable compartment classes. Although it was realized that some of the sources identified are common to all of the panels involved, and that some compartments (such as ground water or the soil interstitial solution) might properly be considered by more than one panel, time was not available either for cross checking among panels, or for full consideration of all the problems clearly included in our charge even when narrowly defined. For these reasons we restricted our discussions to the aquatic environments.

Source Terms

As shown in Figure 1, the known or anticipatable sources of transuranics to aquatic environments could easily be identified and an agreed on classification be achieved. The panel, however, realized quickly that any attempts at quantitative descriptions were seriously constrained by its not knowing either the characteristics or the amounts of transuranic materials being released or potentially to be released from civilian or military nuclear activities.

This panel concurs with the recommendation made at the closing session by the Food Chain panel that a document "Transuranic Elements in the Environment" be prepared under BER auspices. This document should have a chapter dedicated to an inventory, to chemical and physical descriptions, and to the global distributions of each of the potential radionuclide source terms illustrated in Figure 1.

In realizing that each of these source classes must be more thoroughly characterized, and better evaluated quantitatively it was possible to identify a number of specific research needs:

1. Fallout has certainly been the best and most thoroughly studied source term, although much less so for transuranics than for the fission products. Even here, however, questions have been raised concerning the amounts of various nuclides that have been distributed world-wide by this mechanism, and the details of the geographical distributions. Recent unpublished data suggest that, in respect to uptake by marine algae, the ^{238}Pu from the SNAP 9A burnup was less available, in the southern hemisphere, than was $^{239,240}Pu$ from test fallout. The fact that no such difference has appeared in careful studies of northern hemisphere ecosystems raises the possibility that there may have been a north to south difference in particle-size or reactivity of either SNAP 9A ^{238}Pu or test fallout $^{239,240}Pu$ delivered to the earth's surface.

2. Terrestrial Remobilization needs to be quantitatively evaluated as a means of transporting transuranics (delivered as fallout or from other sources) from the land surface to aquatic environments. This question has been raised especially in relation to coastal marine areas, but the process should be placed in proper quantitative perspective relative to other mechanisms of input to coastal zones.

3. Accidents have already introduced a variety of forms of transuranic elements to aquatic environments, and may be expected to introduce many others. By their nature such occurrences are predictable only in a statistical sense, and the basic data are still inadequate for establishing any statistics. Both study of the known incidents, and evaluation of those that can be predicted show that such introductions will involve materials whose rates of solubilization cover a very wide range. Data are not available to support long-term evaluation of these rates in the cases of materials of considerable potential significance. Among these, for instance, may be listed such disparate instances as:

   (a) The long-term integrity of the reactor pressure vessels lost with sunken nuclear powered ships.

   (b) The effect of ThO_2 buildup in aerospace heat-source ceramics on their corrosibility under water.

(c) The nature and rates of solubilization to be expected in case of the particulate materials released from core melt-down of coastal reactors.

4. Disposal of solidified wastes has introduced, and is now introducing, measurable amounts of transuranic nuclides into marine environments; much evidence suggests the rate of these introductions will increase. These activities have been very little studied, but data exist showing that the mobility and biological availability of the nuclides so introduced depend strongly on the overall composition of the materials involved, as well as on that of the environments receiving them. Study of these questions is made very difficult both by the long-term nature of the release processes, and by the near impossibility of obtaining specific adequate data either on the wastes' composition or on their nuclide content.

5. Power Reactors and Allied Plants have been shown to be sources of transuranic nuclides. Because the transuranics so released have not been found in amounts large enough to represent direct health hazards they have been left virtually unexamined. We have consequently no information on the absolute amounts of transuranics leaked, or on the relative abundances of the elements represented. These questions deserve serious attention from at least two points of view:

   (a) A variety of data indicates that much of the transuranic radioactivity from these sources will be locally immobilized in sediments but in forms that may become ultimately available to the local biota. Obviously this has high relevance to the projection of long-term effects of coastal reactors, especially when sited in clusters.

   (b) Leakage of this sort appears to be the least complicated source we have of environmental transuranic nuclides. Thus, each coastal reactor represents a "transuranic experiment" that should be studied.

6. Fuel Cycle Wastes are operationally divided into high-level and low-level. Although at present all known high-level waste is being stored on land under controlled conditions, study is actively being pursued to assess the feasibility and economic advantage of several procedures for the emplacement of high-level waste in stable marine basins. In support of these efforts, a large number of basic studies are needed.

Low-level fuel cycle wastes have been released as aged, fluid effluent, by pipeline to the Irish Sea from British reprocessing plants at Windscale, since the early 1950's. Recently, several other countries have copied this procedure, the the British are reported to be giving serious consideration to a significant increase in their reprocessing operations. Information is lacking about the transuranic nuclide composition of these wastes, about the quantitative patterns of release (rates per month are the best data available), and about the horizontal trajectories of the transuranics, whether in the water column or attached to sediment, after leaving the pipeline. Obviously this last set of data must bear a relationship to the overall chemical composition of the waste stream, and so needs to be studied in several different situations. Equally obvious is the relevance of all these questions to the prediction of long-term build-up, and consequent long-term effects of transuranium nuclides from this sort of disposal.

Low-level fuel cycle wastes are also being released to coastal water by India, Germany and Italy; and there is reason to believe that France is similarly releasing, sporadically, wastes from its military nuclear plants. Less data are available from these operations than from the Windscale operations.

7. Military Fuel Operations give rise to wastes similar to those deriving from the civilian fuel cycle; the isotope composition and probably the transuranic element ratios however, are quite different. Although in the U.S.A. these two waste streams are kept separate, we have less assurance this is so for other foreign nuclear countries. Certainly obtaining adequate data concerning these military wastes will be orders of magnitude more difficult than from the civilian fuel cycle, but we do wish to emphasize the importance of this information to the validity of any long-term assessment of the environmental impact of transuranic nuclides from nuclear technology.

Source Terms Specific Research

Glacier Records

Present data and past fluxes of transuranic elements from the atmosphere to the earth's surface may be more accurately recorded in permanent glaciers than in soil profiles. The use of glaciers has these advantages:

(a) They exist at all latitudes and under all wind-systems.

(b) Their recent strata can be more accurately dated by using ancillary data such as varving, oxygen isotopes and ^{210}Pb techniques.

(c) They also contain records of other atmsopheric pollutants such as trace elements and chlorinated hydrocarbons.

(d) Seasonal and annual accumulations can often be differentiated as soils are ablated over long time periods whereas some glaciers are not. The glaciers provide an untapped advantage of assessing the fallout record. This panel recommends, therefore, that such records be sought in a systematic way that will provide insight into any of the questions listed above.

Close-in Fallout

It can be argued that close-in fallout from tests where the fireball incorporated sizable amounts of soil or of structural materials (tower shots) bears much similarity to the products that would arise from core melt-down. Study of close-in fallout from both the Nevada and Pacific test sites, planned for this purpose, could give us a good deal of insight to what may be expected from the most feared of accidents to a nuclear power reactor. Especially relevant would be data showing the chemical and physical inhomogeneity of close-in fallout particles, and the rates at which they release transuranic nuclides under various environmental conditions.

Solidified Nuclear Materials

Both as result of accidents, and as planned disposal of wastes, solidified nuclear materials have entered the oceans in many forms over the past few decades. Careful study should be undertaken, coordinating with the studies of EPA on the USA packaged nuclear waste dump sites, to provide data concerning the rates, and patterns, of release of transuranics so introduced, and their geochemical fates, and biological availability once released.

Nuclear Technology Leakage

Some leakage of material is unavoidable in any large-scale industrial process. The Panel was made aware where data were available that the

leakage rate approximated 10^{-5} per major technology step. The establishment of the value of such a parameter, or of the range of values, is an essential base for any long term evaluation of source terms from nuclear technology. We urge that this question be examined with care, by suitably qualified technical personnel.

Compartments

The lower section of Figure 1 shows the series of compartments into which the aquatic world could be classified. A vast amount of data relevant to distributions of transuranic nuclides in most of these compartments is available; however, only a fraction of this data base is available in the open literature. All investigators are urged to publish their results in timely fashion and to communicate both their progress and an understanding of those things that still need to be done.

Each compartment should be thought of as containing at least two parts: the water column and the sediment. In most situations the transuranic nuclides have been shown, no matter what the source term, to associate principally with the sediments. In the open ocean because of the great depth of the water column, there is a time lag on the order of decades between transuranic addition, and final association with the sediment, and there appear to be some systematic local perturbations of the sinking processes that have been described. The panel wishes to emphasize the importance of understanding the transient nature of the association of transuranic nuclides with sediments on the order of long time scales. It was probably unavoidable that all existing examinations of transuranic-sediment interactions have been directed to the steady-state prediction; clearly understanding of the steady-state situation is necessary. The Panel realized, however, that serious redistributional terms will be from episodic, catastrophic events, and that these studies have not yet begun. Prediction of the frequency, nature, and magnitude of catastrophic remobilizations, and of their effects, locally, on the biological availability of transuranic nuclides, would be a high-priority research goal. Each compartment should be studied to ascertain whether expected catastrophes may increase or decrease the rate of return of transuranic nuclides to man. The example which brought this question to the attention of the Panel was that of hurricanes: On an atoll, a hurricane may be expected to transfer large amounts of lagoon sediment (with its contained transuranics) to the surrounding deep ocean where the rate of return to man

will be much diminished; in an estuary, on the contrary, a hurricane may deposit large amounts of sediment on beaches, salt marshes, or filled land, where transuranic availability will be much enhanced. Another example shows rivers may be subdivided into those with flood plains, where a major flood may deposit inches to feet of sediment onto agricultural land, with much enhanced transuranic availability, or those without flood plains, and emptying into estuaries or large lakes, where a major flood may sweep much accumulated sediment into deeper water, with diminished transuranic availability.

Compartment Classes

It is by no means clear to what extent data concerning transuranic behavior can be extrapolated from one compartment class to another, or even among specific instances within a class. Some data already show that the relative biological availability of Pu and Am is different in fresh water from that in salt. Although it is believed that many such effects will prove to be the results of local differences in concentration of specific soluble complexes, other possibilities remain for examination.

Parameters

Table 1 illustrates one attempt to classify the important parameters and processes in aquatic systems that influence transuranic nuclide movements. Evidence suggests that removal from solution of many heavy elements (including the transuranics) in environmental systems depends greatly on the electrical charges and surface polarization of suspended solids. There is evidence that suggests a dependence upon the surface-area to volume ratio of the solid particles and this leads to the conclusion that colloidal particles represent a most important initial removal mechanism. The techniques of classical colloid chemistry have much to offer in following up these suggestions. Future investigation of transuranics in the environment should include serious consideration of pH (relative to the isoelectric point of transuranic-bearing colloids), the concentration and oxidation states of electrolytes, water velocities relative to particle motions, the concentrations and natures of organic complexes, whether solid or soluble, and the age of any possible transuranic-bearing colloids.

It is important to note that the parameters listed in Table 1 are not of equal importance in their applications to the different source classes or different compartment classes in Figure 1. For instance, sinking rate becomes of real importance only in the open ocean or in

very deep lakes; redox equilibria probably are significant only in sediments, stratified lakes, or the few marine basins that are anoxic; and so on. Some disagreement was felt over the value for distribution coefficients for biotic uptake: Some panel members suspect that these vary little from 10^{-5}, whereas others are convinced that no such generalization can be made.

Transport Models

Of especial relevance to the assignment of this panel is the construction of Transport Models. These describe the movement of transuranic nuclides within, through, and between aqueous systems, whether from a point source or diffuse chronic releases. Even though our present state of knowledge permits construction of models only of great crudeness, they can be useful to make first estimates of the availability and distributions of transuranic concentrations in water, sediment or biota. Building of models requires, first, insight into what the important transport processes are in the compartment under study. To implement these models, we require information on these transport parameters (source/sink terms) plus other parameters such as velocity, dispersion, sediment deposition velocities, etc. For instance, for pulse releases, velocity and dispersion coefficients are important, but are not as important in describing chronic releases or long-term residuals from accidental releases still in the system. In general, crude models can be used with rough estimates for the parameters, but refined estimates require specific validation experiments that must extend over long time spans.

The panel is aware that the cyclic nature of most ecological parameters, when measured over long time periods, reduces the precision of our predictive capability. Nevertheless, these models and predictions should be made so refinements can be made. For this purpose, obviously, each model should be subjected to a careful sensitivity analysis, the results of which should be used equally to refine both the model and the estimate of its prediction uncertainty, and to feed back into the program of measurement and experimentation. In this context the panel urges that attention be paid to each of the relatively long-term experiments that have resulted from transuranic releases to the environment.

Intercalibration

During its discussions the panel became increasingly conscious of the strain that is placed on analytical accuracy and precision in transuranic

measurement by the requirement of 10^3 to 10^6 year extrapolatability of predictions. Every effort should be made to improve our analytical quality control for transuranic measurement.

A strong recommendation by the Aquatic Transport panel, therefore, is cooperation with ongoing bilateral intercalibration efforts of other ERDA groups, as well as with the present IAEA activity along these lines.

Certainly the reliability of predicting transport processes from specific injections to the aquatic environment and comparisions with other investigators in other systems would be necessarily increased by this exchange of analytical information. Some necessary ingredients of this intercalibration program would include:

(a) Development of standard reference materials (perhaps by DOC-NBS) for a variety of matrices: soil, sediment, water, biota (plant/animal) all at various concentrations (environmental, intermediate, contaminated ~Enewetak, NTS).

(b) An exchange of common samples with a variety of matrices.

(c) Blind samples from sponsoring groups (~IAEA).

Summary of Research Needs

Most of the research needs summarized briefly here are noted at greater length above as shown by the page and paragraph references in parentheses:

1. Characterization, in terms of transuranic element and isotope ratios, chemical and physical nature, inventory, and global distribution, of each of the potential source terms listed in Figure 1.

2. Refine knowledge of transuranic distributions from world-wide fallout.

3. Verify reported southern hemisphere difference in biological availability of SNAP-9A versus fallout Pu, and assess its physico-chemical basis.

4. Begin program of using glacier ice for evaluation of fallout deliveries in relation to latitude and time.

5. Verify general importance, and dependence upon local conditions, of Terrestrial Remobilization as a means of redistributing transuranics.

6. Refined evaluation of rates of solubilization, and of the geochemical behavior of soluble leachates, of transuranics in solid materials deposited in aquatic systems, whether by accident or by disposal.

7. Begin study of transuranic leakage by power reactors, especially with reference to the magnitude of the source term, and to their use as local experiments in curium environmental behavior.

8. Maximize the information on transuranic distributions about the existing long-term environmental releases: e.g., Hanford, Enewetak, Bikini, Thule, Oak Ridge, Windscale, Karlsruhe, etc.

9. Begin study of close-in fallout from weapons tests as an analog of the debris to be expected from a core-melt-down disaster.

10. Begin study to evaluate the rate of loss of transuranics at each step in their industrial technology.

11. Refine evaluations of the interactions of transuranics with sediments, and with the interstitial or overlying solutions, with special reference to slow leakage.

12. Emphasize evaluation of the kinetic aspects of transuranic associations with solids or organisms; examine systems for evidence of the importance of non-equilibrium processes in controlling transuranic distributions and movements.

13. Begin study to support prediction of natural catastrophic events, and of their potential to remobilize transuranic nuclides in the environment, and speed their return to man.

14. Establish ways of evaluating the physical-chemical state of elements present in aquatic systems at such low concentrations as are the transuranics.

15. Improve communication between model makers and those studying transuranics in aquatic environments to ensure that models are constructed with relevance to real worlds.

16. Emphasize analytical quality control with special reference to the preparation of Standard Reference Materials, and to the support of, and the participation in, intercalibration exercises.

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FIGURE 1

Sources and Compartments

Source Classes

Fallout	Land Resuspension	Accidents	Disposal	Reactors and Allied Plants	Fuel Cycle Wastes	Military Fuel Operation
World-wide		Military			Low-level	
					High-level	
Close-in		Aerospace				
		Civilian				

COMPARTMENT CLASSES

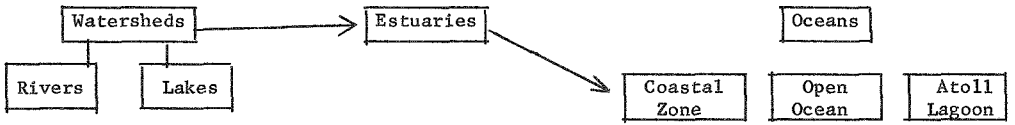

TABLE I

PARAMETER CLASSES

Chemical Speciation	Physical Speciation	Distributional Processes
	Particle associations	
Anionic	Size	K_D Abiotic - Kinetic, Equil.
Cationic	Sinking Rate	K_D Biotic - Kinetic, Equil.
Redox Conditions	Nature	Transport - Mass Balance
Colloidal State	Device Debris	Physical
Complexation	Mineral Detritus	Biotic
Biological Availability	Organic Detritus	
Surface Activity		