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Environmental and Radiological Safety Studies

*Interaction of $^{238}\text{PuO}_2$ Heat Sources with
Terrestrial and Aquatic Environments
January 1—March 31, 1981*

Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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QUARTERLY REPORT
ENVIRONMENTAL AND RADIOLOGICAL SAFETY STUDIES

**Interaction of $^{238}\text{PuO}_2$ Heat Sources
with Terrestrial and Aquatic Environments
January 1 to March 31, 1981**

Compiled by

Glenn R. Waterbury

ABSTRACT

The containers for $^{238}\text{PuO}_2$ heat sources in radioisotope thermoelectric generators are designed with large safety factors to ensure that they will withstand reentry from orbit and impact with the earth and safely contain the nuclear fuel until it is recovered. Existing designs have proved more than adequately safe, but the Space and Terrestrial Division of the Department of Energy Office of Advanced Nuclear Systems and Projects continually seeks more information about the heat sources to improve their safety. The work discussed here includes studies of the effects on the heat source of terrestrial and aquatic environments to obtain data for design of even safer systems. The data obtained in several ongoing experiments are presented; these data tables will be updated quarterly. Discussions of experimental details are minimized and largely repetitive in succeeding reports. Compilations of usable data generated in each experiment are emphasized. These compilations include data from environmental chamber experiments that simulate terrestrial conditions, experiments to measure PuO_2 dissolution rates, soil column experiments to measure sorption of plutonium by soils, and several aquatic experiments.

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I. INTRODUCTION

Radioisotope sources provide heat for electric power generators that can supply constant voltage for several years. Therefore, they are used in many space missions to provide electric power for instruments and data transmission. In five of the Apollo manned landings on the moon, the scientific instrument packages deployed for postmission measurements were powered by such radioisotope ther-

moelectric generators (RTGs). Because the half-life of ^{238}Pu is relatively short (87.7 yr), the α -particle specific power is high enough (equivalent to 0.56 W/g of ^{238}Pu) to be used as a heat source in electrical power generation. In nearly all space applications to date, plutonium 80% enriched in ^{238}Pu has been used to supply the heat for the generators. The heat source container is designed with a great safety factor to withstand both reentry from orbit and impact

with the earth, so that the source will be safely contained until it is recovered. Although existing designs have proved more than adequately safe, the Space and Terrestrial Division of the Department of Energy Office of Advanced Nuclear Systems and Projects continually seeks more information about the heat sources to improve their safety. As part of this program, Los Alamos National Laboratory is studying the interaction of ^{238}Pu fuel material with seawater, fresh water, and marine organisms, and with air, soil, and rain to supply data for design of even safer heat sources.

Several chemical forms of ^{238}Pu have been used as heat sources. The forms now in use or being studied for future use are various pressed shapes of pure plutonium dioxide (PPO). We have run several aquatic experiments using plutonia-molybdenum cermet (PMC), but all our terrestrial experiments have involved PPO.

Discussions of details of the experiments are intentionally brief and will be repeated in subsequent progress reports. The main purpose of these reports is to compile in one convenient reference all usable data generated in the Los Alamos experiments. The compilation will be updated each quarter. When an experiment or set of experiments is completed or terminated, a final topical report will be issued and that phase of the work will not be included in subsequent quarterly reports.

II. BEHAVIOR OF ^{238}Pu FUEL IN TERRESTRIAL ENVIRONMENTS

(James H. Patterson, Frederick Steinkruger, and George M. Matlack)

A. Environmental Test Chambers

Meaningful data on the environmental effects of the radiation and heat from radioisotope heat sources can best be obtained under simpler, less varied conditions than exist normally in nature. Use of environmental test chambers permits selection of the parameters to be tested and ensures control over them. Determination of the effects of these parameters on the fuel material and simulation of different climates and soils are also possible.

Each environmental test chamber has an interior volume of 1.8 m^3 and can be programmed for diurnal temperatures of -20 to 95°C and relative humidities from 100 to 25% or lower, depending upon the temperature. A manually operated spray system

simulates rainfall at rates up to $30\text{ }\mu\text{m/s}$ ($\sim 4\text{ in./h}$).

Each chamber contains a 0.9- by 0.9-m tray, filled to a depth of 0.3 m with soil. The soil is supported by a grill covered with a thin perforated plate to drain off the rainwater that percolates through the soil. The bottom of the tray tapers to a pipe that runs to the outside of the chamber so this water can be collected. Three of the trays are divided into compartments, each of which has separate percolate collectors.

B. Soils

About 1.4 Mg each of loam, clay, sand, and silt loam were obtained for terrestrial experiments. Compositional analyses of these soils were provided by the U. S. Soil Conservation Service Laboratory at Riverside, California (Table I). In addition, about 1 Mg of black humus-type soil was collected from Canada Bonita in the Santa Fe National Forest (Jemez Division), Los Alamos County.

C. Test Conditions and Analyses Performed

The first experiments were started with the chambers programmed for humid summer conditions (20 to 40°C , 87 to 96% RH). Cams installed later provided humid winter conditions (0 to 17°C , 71 to 100% RH). The alternate summer and winter periods each last 6 months. Two chamber experiments are operated alternately under arid winter conditions (3 to 18°C , 21 to 87% RH) and arid summer weather (26 to 53°C , 7 to 33% RH). Three experiments were started under humid winter conditions, but one was terminated after a year (see Sec. II.D.2). Another experiment was carried on in a humid environment for 3 yr in the same chamber (see II.D.4). In the other two experiments the sources and the top layer of soil were removed after 5 yr and 8 months, and the experiments were continued as simulated cleanup operations. An experiment with four compartments, each with a different soil, was started under humid conditions. The other six current chamber experiments are now operating under winter conditions, two in arid climate, the other four under humid conditions.

The fuel material sample is placed in the center of the tray on the soil surface, except in the four-soils experiment, where it is placed in a small cup which drains into the dehumidifier waste bottle. Each sample under humid conditions receives a 3-cm rain once a week. The rains for arid conditions occur approximately once a month, except that there were no

TABLE I
COMPOSITION OF SOILS USED
IN TERRESTRIAL EXPERIMENTS^a

	<u>Loam</u>	<u>Clay</u>	<u>Silt</u> <u>Loam</u>	<u>Sand</u>
Sand (%)	49.5	1.5	25.3	88.6
Silt (%)	37.6	31.4	60.1	4.6
Clay (%)	12.9	67.1	14.6	6.8
C, organic (%)	0.6	0.12	0.03	0.01
Carbonate as CaCO ₃ (%)	trace	8	10	4
H ₂ O at 15 bars (%)	7.6	23.8	7.5	2.6
H ₂ O at saturation(%)	40	91	45	22
pH	8.0	7.8	8.4	9.1
Ca(meq/100 g)	16.3	43.9	26.4	17.8
Mg(meq/100 g)	1.9	10.2	3.7	1.7
Na(meq/100 g)	0.5	4.6	3.5	0.9
K(meq/100 g)	1.0	1.1	0.5	0.5
Cation exch cap	13.6	42.0	14.7	6.0
Elect. Conductivity (mmho/cm)	0.6	4.2	3.3	0.6
Clay mineral ratios:				
Mica	4	---	---	---
Montmorillonite	3	5	5	5
Kaolinite	3	3	2	3

^a Soil analyses provided by the U. S. Soil Conservation Service Laboratory, Riverside, California.

rains during the first arid summer. The rainwater that percolates through the soil is analyzed for plutonium. The condensate from the refrigeration-type dehumidifier in the chamber conditioning system also is analyzed to get a relative indication of the airborne plutonium in the chamber. Before 1978, core samples of the soil were collected at approximately 6-month intervals. Each core was divided into 5 to 14 sections that were analyzed separately to determine the plutonium variation with depth in the soil.

Small air samplers, fitted with approximately 40-mm-diam filter papers, were operated several times during rains in three chambers to measure the airborne plutonium. The filter papers were changed frequently during operation, and the plutonium was measured by α -particle counting the papers and in

some cases by dissolving them and α -particle counting the resultant solutions by liquid scintillation procedures.

Time-lapse movies were taken in four chambers during rains. Emissions from one fuel sample were studied by time-exposure photographs, without external lighting, in the near-ir, visible, and near-uv regions of the spectrum.

D. Test Samples and Results

Seven terrestrial environmental chamber experiments are under way. They are summarized in Table II.

1. *MHFT-12*. This was a PPO sphere that shattered during a simulated reentry and impact test for

the Multihundred Watt (MHW) program. The sphere fragments were removed from the container and sorted by size. The pieces with diameters greater than 6 mm, weighing 224 g, were placed on loam in one environmental test chamber on June 21, 1973, and 28 g of 0.01- to 6-mm-diam material were placed on loam in another on September 27, 1973. These experiments are now in their eighth winter in a humid environment.

Photographs of the emissions from the large pieces in the near-ir, visible, and near-uv spectral regions showed that the external temperature of the pieces was 250 to 300°C, and that an air glow around them was emitted mainly in the visible region, although some of it may have been uv. This air glow was probably caused by ionization of air by the alpha or gamma activity of the pieces.

Plutonium airborne concentrations increase during periods of rain on the large pieces. This effect results in a greater plutonium pickup from the air by the dehumidifier, as measured by plutonium analyses in the dehumidifier condensates. Analyses for a period of 2 months are shown in Fig. 1. The rate of collection of plutonium in the dehumidifier condensates from the chamber containing the large pieces was 13 times that for the fine material. The explanation is that material spalls from the hot pieces of PPO because of thermal shock when the cold rain first hits them. Spallation has been observed in nature when some kinds of rock, heated by the sun, are hit by a sudden shower.¹ This mechanism for the airborne plutonium observed over the PPO pieces seems to be confirmed by the striking results (Table III) of an experiment in which air particulate

TABLE II
TERRESTRIAL ENVIRONMENTAL CHAMBER EXPERIMENTS

Experiment No.	Description	Date Started
1	MHFT-12 chunks on loam; humid climate	6-21-73
3	MHFT-12 fines on loam; humid climate	9-27-73
6*	MHFT-50 fines on loam; circular soil partitioner; humid climate	4-9-75
7	MHFT-27 fines on sand; arid climate	2-26-75
8*	MHFT-50 chunks on loam; circular soil partitioner; humid climate	4-9-75
10	MHFT-27 chunks on sand; arid climate	2-26-75
12	ET1-1 cylinder on 4 soils, humid climate	2-27-81

* Source removed 12-19-80

TABLE III
AIRBORNE PLUTONIUM CONCENTRATIONS
DURING A RAIN IN AN ENVIRONMENTAL
CHAMBER CONTAINING LARGE PIECES OF
PLUTONIUM DIOXIDE

Filter No.	Collection Time (min)	Pu Concentration (pCi/m ³)	Remarks
1	24	3	Before rain
2	5	30 000	First 5 min of rain
3	5	6 000	
4	10	4 000	
5	10	3 000	
6	15	900	
7	15	800	Last 15 min of rain
8	50	200	
9	180	3	
10	860	3	

samples were taken continuously during a rain sequence. The average airborne plutonium concentration during the first 5 min of rain was 10 000 times the prerin concentration. After this, it dropped until it reached the prerin level about 1 h after the end of the rain. The principal cause for the rapid decrease was gravitational settling of the particles. The rain washed plutonium out of the air, but it could not have been the only removal mechanism because the decrease continued after the rain had stopped. A repetition of the experiment, but with the dehumidifier turned off, produced similar rates of decrease, indicating that pickup by the dehumidifier was not the principal removal mechanism. The results in Table III were obtained by counting the fiber glass filters with a 2- π gas proportional counter. Some of the filters were subsequently dissolved, and a more accurate analysis by liquid scintillation counting showed that most of the initial activity came from relatively large particles caught on the filter surface, and therefore they were counted efficiently by the proportional counter. Later, the large particles settled out, and the remaining smaller particles penetrated deeper into the filter and were counted less efficiently. This experiment was repeated twice, with Millipore membrane filters of two different pore sizes, and at two air collection

points within the chamber, in order to obtain samples for particle size and agglomeration studies. The four experiments in which particles were collected from the air in this chamber during a rain sequence showed a remarkable similarity in the variation of the plutonium concentration in air with time, even though filters of two types and of two pore sizes were used in two different locations in the chamber, and the dehumidifier was off in some experiments and on in others. In every case the plutonium concentration during the first 5 min of rain was 4 orders of magnitude greater than the prerin concentration to which it returned in about 2 h. The same behavior was observed in the liquid nitrogen experiment described below.

The first rainwater that percolated through the soil supporting the large fuel pieces contained 1.8 μ Ci of plutonium. This very rapid plutonium breakthrough indicated that material in the form of extremely small particles or a colloid was released from the surface of the fuel by the rain water. Plutonium ions would not be stable in solution at the slightly basic pH (about 8) and, if present, would be held in the soil by ion exchange.

The amount of plutonium found in the rainwater that percolated through the soil supporting the large pieces was strongly influenced by the climate, as is

shown in Table IV. Each weekly rain was equivalent to a 32-mm rainfall and deposited 27 ℓ of water. Under summer conditions, about 6 liters of rainwater percolated through the soil. This water contained an average of 0.8 μCi of plutonium. Winter rain produced more percolated water because of lower evaporation losses and a higher plutonium content, the early winter rains yielding as much as 18 μCi of plutonium. It seems that the higher summer temperatures dried the soil more between rains so it took longer to saturate during a rain. Because of this lower water flow in summer, fewer plutonium dioxide particles were washed through the soil. At the beginning of the winter, some of the particles left in the soil during the summer were washed through, giving higher results for the first few rains. These seasonal effects have occurred each summer and winter cycle, until the last four winters, when the plutonium content was approximately the same as in summers.

In the case of the fine material, the same behavior was seen in the average volumes of the percolated rainwater with changes in climate, but the average plutonium content was not only smaller but also showed little change with climate until the last eight seasons, in which the plutonium contents were a factor of 10 higher than before (Table V).

Autoradiography² of residue from evaporation of a percolated rainwater sample that was taken in winter from the chamber containing the large pieces of MHFT-12 revealed 0.05- to 0.8- μm -diam plutonium oxide particles with a count median diameter (CMD) of 0.095 μm . These accounted for only 1 ppm of the total plutonium in the sample, indicating that nearly all the plutonium particles carried through the soil by water were less than 0.05 μm in diameter.

Usually there was good agreement in measured radioactivities among aliquots from the same percolated rainwater sample. Occasionally there was disagreement with one aliquot counting as much as several hundred times the others. This indicated the presence of a significantly larger than average particle of PuO_2 . Assuming a single large particle, we estimated that the largest particle found in the high-activity aliquots was equivalent to a 4- μm -diam sphere, five times the diameter of the largest particle seen in the autoradiographic analyses of a rainwater sample in which the count rates of the aliquots agreed.

The fine particles of MHFT-12 interacted differently with the simulated environment. These particles were too small to have temperatures above ambient, so rain-induced spallation was negligible. Plutonium concentrations in airborne particulates

during a rain in the chamber also were measured. The results (Table VI) show that the airborne plutonium concentration before the rain was about the same as the prerin concentration in the chamber containing the larger pieces of MHFT-12. This concentration did not increase significantly during the first 5 min of rain, in contrast to the experiment with the large pieces. Instead, airborne plutonium decreased, and after the rain, the concentration was an order of magnitude lower than before. These lower concentrations probably indicated that soil wetting inhibited resuspension of plutonium-bearing particles. The counting rates were so low that we had to combine the six filters used during the rain to obtain meaningful statistics. The same treatment was necessary for the first five filters used after the rain.

We analyzed 19 soil cores in chamber 1 and 18 cores in chamber 3 for plutonium. The results and core locations for the first eight cores in each chamber are shown in Figs. 2 and 3. Each core was taken 25 cm from the central pile of fuel material. Cores 1 and 2 were collected at the end of the first summer, cores 3 and 4 during the first winter, cores 5 and 6 during the second summer, and cores 7 and 8 during the second winter. The top two or three sections of cores 3 and 4 in both of these chambers were divided into two nearly equal parts by a vertical cut. Six of these half sections, as indicated in Figs. 2 and 3, were examined for particle size distribution and agglomeration by LFE Environmental Analysis Laboratories. The rest of the half-sections were analyzed at Los Alamos for plutonium. Zeros indicate that less than 0.03 ng of plutonium was found.

The data in Figs. 2 and 3 show that, in addition to downward movement of plutonium through the soil, there was surface movement to the right and back of each chamber, the effect being greater in the chamber containing the fine particles of fuel. This movement was confirmed by a radiation contour map (Fig. 4) of the gamma- and x-ray intensities in the chamber containing the fine particles. An array of 17 thermoluminescent detectors (TLDs) was placed 10 cm above the center of the tray containing the fine material, one at each horizontal and vertical intersection of the grid in Fig. 4, except for the 10-cm tick marks. The intensities ranged from 0.3 R/h at the front (lower) left corner to 8.8 R/h at the center. If all the plutonium was still concentrated at the center of the tray, the contours would be circular. Their deviation indicates that some of the plutonium on the surface has been moved toward the back and to the right. This movement seems to have been caused by water released from a gate valve directly above core position 1 whenever summer humid conditions

TABLE IV
CLIMATIC EFFECTS ON AMOUNT OF
PLUTONIUM CARRIED BY WATER PER-
COLATING THROUGH SOIL
(Large Pieces of PuO_2)

<u>Climate</u>	<u>Conditions</u>	<u>Av Percolated Water Per Rain (liters)</u>	<u>Av Pu (μCi)</u>
Winter	0 - 17°C	10	2.7
	60 - 100% RH		
Summer	20 - 40°C	6	0.8
	85 - 97% RH		

TABLE V
CLIMATIC EFFECTS ON AMOUNT OF PLUTONIUM CARRIED
BY WATER PERCOLATING THROUGH SOIL
(Fine Material from MHFT-12)

<u>Climate</u>	<u>Av Volume per Rain (liters)</u>	<u>Av Pu (μCi)</u>
Winter	10	1.0
Summer	5	0.5

TABLE VI

CONCENTRATIONS OF AIRBORNE PLUTONIUM
DURING RAIN IN AN ENVIRONMENTAL CHAMBER
CONTAINING FINELY DIVIDED 238-PLUTONIUM DIOXIDE

<u>Filter No.</u>	<u>Collection Time (min)</u>	<u>Pu Concentration (pCi/m³)</u>	<u>Remarks</u>
1	103	1.9	Before rain
2	5	2.1	First 5 min of rain
2-7	60	1.9	Average during rain
8-12	164	0.12	After rain
13	926	0.14	

prevailed and the valve was opened to the building exhaust to provide air flow when the chamber door was opened. (This artifact was eliminated in June 1975, by deflecting the water away from the tray of soil.) The radiation map measurements were confirmed by an autoradiograph of the activity of the soil surface.

A seasonal effect was seen in three cores from the chamber containing the fine particles (Fig. 2). Cores 2, 4, and 6 all had higher plutonium concentrations than cores 1, 3, and 5 because of the gate valve artifact mentioned above. Cores 2 and 6, however, which were collected in the summer, had higher concentrations than core 4, collected in the winter, and there was more plutonium in sections below the surface, especially in the bottom sections. These high concentrations in the subsurface soil during the summer agree with the reason postulated to explain the low plutonium concentrations in summer rainwater that percolated through the soil; that is, the smaller flow of water in the summer did not wash as much plutonium out of the soil as did the greater winter flow. The high concentration in the bottom sections of these summer cores, also seen in Fig. 3, was less easily explained. Possibly some of the plutonium was held up by the perforated plate under the soil when the flow was low; the holes are only 22% of the surface area. Also, plutonium oxide particles may have collected at the soil and air interface

under low-flow conditions.

The seasonal effect was less marked in cores 7 and 8, which were taken during the second winter, in the chamber containing the fine particles. This may have been caused by gradual fixing of the plutonium dioxide in the soil with the passage of time. Core 7 looked like a winter core except for the bottom section, which was very high in plutonium. Core 8 was more like a summer core, with small amounts of plutonium in all sections.

Analyses of the cores from the large-piece chamber (Fig. 3) showed similar, but less pronounced, spatial and seasonal correlations. Part of this blurring of the effects may have been caused by water from the gate valve splashing onto the pieces and causing spallation. Evidence of this complication was seen in the experiment, described earlier, in which the air concentrations during a rain were measured with the dehumidifier off. In that experiment the prerain concentration was high, probably because the gate valve was opened just before the rain. Unfortunately, the sample of the bottom section of core 6 (Fig. 3) was lost, and we do not know whether the concentration was high at the bottom of this core, as it was in the other cores taken in similar places in the summer.

The six half-sections from cores in the two chambers were analyzed at LFE Environmental Analysis Laboratories for particle size distribution and agglomeration with the soil.³ There were no particles

Fig. 1.

Effect of rain on plutonium content of dehumidifier condensates.

large enough to form stars in a photographic emulsion in either section of core 3 from the chamber containing the fine particles. There were stars in the top section of core 4 in both chambers. The particle size range was slightly smaller than that in the rainwater that percolated through the soil; 90% of the particles had equivalent diameters of 0.04 to 0.2 μm . The lower limit may not be significant, as it is close to the detection limit of the procedure. The particles in the cores from the soil supporting the fine particles were slightly larger than those in the soil supporting the large pieces. They had a CMD of 0.072 μm compared with 0.056 μm , but they were still very much smaller than the 10- μm minimum parent particles. This fact agrees with observations that $^{238}\text{PuO}_2$ is much less stable than $^{239}\text{PuO}_2$.⁴⁻⁷ This instability seems to be associated with the intense radiation field of the isotope's alpha activity. Radiation damage to the oxide crystal lattice, reaction with radiolysis products in soil and water, and aggregate recoil may contribute to this degradation. Fleischer and Raabe have observed minute plutonia particles containing 60 to 10 000 plutonium atoms in aqueous media around $^{239}\text{PuO}_2$ powder.⁸ The diameters of these par-

ticles are less than 0.01 μm . They apparently originate from the disruption of material by the recoiling uranium atom following alpha decay. These particles may be washed off the fine material during a rain.

Twenty-five of the particles that formed stars, chosen at random on the photographic plates of widely dispersed material from the core of soil supporting the large fuel pieces, were examined microscopically for agglomeration. Each particle was found to be associated with one microscopically visible soil particle. This indicated that trapping of plutonium dioxide particles by soil was not a matter of filtration of the fine oxide particle, but that there was agglomeration of individual plutonium oxide and soil particles. This factor may determine which particles will be retained by the soil and which will pass through it. The soil particles in the agglomerates had the same size distribution as the bulk of the soil.

Cores 9 and 10 were taken from the chamber containing the large pieces of MHFT-12 during the second winter cycle, at locations 20 cm to the left and right of the PPO pieces, respectively. Core 10

a. Sample to LFE Environmental

Core positions:

8 4 6
1 Pu 2
5 3 7

Fig. 2.

Plutonium-238 in soil cores from chamber containing fine material from MHFT-12 (ng).

a. Sample to LFE Environmental

b. Sample lost

Core positions:

4
 8 6
 1 Pu 2
 5 7
 3

Fig. 3.

Plutonium-238 in soil cores from chamber containing large pieces from MHFT-12 (ng).

Fig. 4.

Radiation contours from a TLD grid above soil in chamber containing fine material from MHFT-12.

was sent to LFE Environmental Analysis Laboratories for particle size and agglomeration studies. There the copper tube containing the core was cut into eight sections with a tubing cutter. Core sections 1, 2, 4, and 8 (numbered from the top) were spread on slides and autoradiographed to determine the number and size of the plutonium oxide particles. No particles were found on these slides with diameters greater than $0.25\ \mu\text{m}$, and very few were greater than $0.1\ \mu\text{m}$. The numerical distributions of the particle sizes for sections 1 and 2 were almost identical. The mass distributions were also close, except for three particles in section 2 that were greater than $0.15\ \mu\text{m}$ in diameter, which heavily biased the mass distribution in this section. The particles in sections 4 and 8 had diameters that averaged less than half of those in sections 1 and 2. The number

distributions of sections 4 and 8 were about the same as were the mass distributions. It would be expected that the particle size distribution of samples like these would show large particles on the surface, with smaller, approximately equal-sized particles in the subsurface sections. Unless the results for the top two samples have been inadvertently reversed, it would seem that there had been mixing of the soil down to the level of section 2, approximately 2 cm.

Core 9 was divided into 10 vertical sections for plutonium analysis. The top section contained 24 ng, but the other nine sections each contained less than 0.03 ng of plutonium. This agrees well with the results of core 1, collected during the first summer, 25 cm to the left of the heat source pieces.

Cores 11 and 12 in the chamber containing the large pieces of MHFT-12 and cores 9 and 10 in the

chamber containing the fine material were taken during the third summer from positions approximately 13 cm in front of and behind the plutonium dioxide, respectively. The analyses of these cores for plutonium (Tables VII and VIII) show that core 1-12 is typical of cores taken during the summer weather cycle, with high total plutonium and at least 1 ng in each section. Core 1-11 has much less plutonium, with nearly all of it in the top and bottom sections. Cores 3-9 and 3-10 have similar plutonium contents. Core 3-9 has practically all of the plutonium in the top section. In Core 3-10, the plutonium generally decreases down the column, with a slight increase in the bottom sections.

Cores 1-13 and 1-14 were collected in the chamber containing the large pieces of MHFT-12 during the third winter from positions approximately 13 cm to the left front and the right rear, respectively, of the source pieces. Cores 3-11 and 3-12 were collected at this same time at positions 13 cm to the left and right, respectively, of the source material in the chamber containing the fine material from MHFT-12. In all four of these cores nearly all of the plutonium was found in the top section (Tables IX and X). In cores 1-13 and 1-14 measurable amounts were seen in the bottom sections and one other sec-

tion, with none in the other six sections. In cores 3-11 and 3-12 small amounts of plutonium were seen in most of the lower sections, a characteristic of previous summer cores. The plutonium seems to peak a few centimeters from the bottom of these columns.

The plutonium profiles from these cores are similar to those found by Essington in Nevada Test Site soils.⁹ He divided the profiles into three classes: normal, low, and abnormal. The normal ones had most of the activity at the surface, with decreasing amounts down the profile. This corresponds to our cores 1-12, 1-13, 3-3, and 3-10. (Activity in the bottom section is ignored as a possible artifact.) The low profiles had plutonium near or below the detection limit for most of the profile. Our cores 1-6, 1-7, 1-9, 1-11, 1-14, 3-1, 3-5, 3-7, and 3-9 have this characteristic. All the other profiles were classed as abnormal, many of which had peaks of activity below the surface, a feature of many of our core profiles.

Cores 1-15 and 1-16 in the chamber containing the large pieces of MHFT-12 and cores 3-13 and 3-14 in the chamber containing the fine material were taken during the fourth summer from positions approximately 13 cm to the front right and back left, respectively, of the plutonium dioxide sources.

TABLE VII

PLUTONIUM IN SOIL CORES
(Finer Material from MHFT-12)

Core 3-9			Core 3-10		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-3	33.8	1	0-3	30.1
2	3-7	0.33	2	3-5	7.6
3	7-9	0.086	3	5-8	1.8
4	9-12	0.034	4	8-12	0.66
5-11	12-25	<0.03	5	12-14	0.20
			6	14-16	0.049
			7	16-19	0.037
			8	19-20	<0.03
			9	20-22	0.035
			10	22	0.037
			11	22-24	0.044
			12	24-25	0.26
Total Pu		34.3			40.8

TABLE VIII

PLUTONIUM IN SOIL CORES
(Larger Pieces from MHFT-12)

Core 1-11			Core 1-12		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-5	4.26	1	0-3	95.6
2	5-8	<0.03	2	3-8	35.1
3	8-11	<0.03	3	8-11	1.44
4	11-15	0.086	4	11-13	1.28
5	15-18	1.45	5	13-17	1.62
Total Pu		5.80	6	17-18	33.9
					168.9

TABLE IX

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING LARGE PIECES FROM MHFT-12

Core 1-13			Core 1-14		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-4	56.2	1	0-5	593
2	4-7	0.87	2-4	5-15	<0.03
3-5	7-16	<0.03	5	15-18	0.15
6	16-18	0.13			
Total Pu		57.2			593

TABLE X

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING FINE MATERIAL FROM MHFT-12

Core 3-11			Core 3-12		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-3	2.91×10^3	1	0-2	2.15×10^5
2	3-5	4.21	2	2-4	246
3	5-8	0.92	3	4-7	79
4	8-10	0.09	4	7-8	1.20
5-6	10-14	<0.03	5	8-11	1.31
7	14-16	0.05	6	11-13	0.37
8	16-18	<0.03	7	13-15	0.95
9	18-20	0.35	8	15-17	0.04
10	20-22	10.7	9	17-19	5.43
11	22-23	3.39	10	19-21	2.96
12	23-24	66.8	11	21-23	215
13	24-25	10.6	12	23-24	2.71
Total Pu (ng)		3.01×10^3	13	24-25	31.2
					2.16×10^5

Analysis of soil cores 3-13 and 3-14 (Table XI) indicates that both have abnormal profiles because they have two subsurface peaks. The plutonium profiles of soil cores 1-15 and 1-16 (Table XII) are also considered abnormal because of subsurface peaks. During the fourth winter, soil core samples 1-17 and 1-18 were obtained from positions 38 cm to the left and right, respectively, of the large pieces from MHFT-12, and cores 3-15 and 3-16 were obtained from positions 13 cm to the front left and back right, respectively, of the fine material from MHFT-12. Plutonium analyses of these four soil cores (Tables XIII and XIV) show low plutonium in cores 1-17 and 1-18 and profiles that are abnormal because of subsurface peaks in cores 3-15 and 3-16. The plutonium distributions of cores 1-17 and 1-18 are similar to those previously observed in winter, but those of cores 3-15 and 3-16 are more like previously observed summer cores.

During the fifth summer, soil cores 1-19 and 1-20 were obtained from positions 38 cm in front of and behind, respectively, the large pieces from MHFT-12, and cores 3-17 and 3-18 were taken from positions 38 cm to the left and right, respectively, of the fine material from this source. The analyses of cores 1-19 and 1-20 show abnormal profiles of plutonium in that there are higher plutonium contents in sections below the surface in both cores than in the top and

bottom sections (Table XV). The profiles of cores 3-17 and 3-18 are normal in shape (Table XVI), but there is very little plutonium below the surface in core 3-18. Thus chamber 3 again shows a reversal of the previously seen summer and winter differences.

2. *MHFT-27*. As part of the MHW safety sequential test (SST) program, MHFT-27, a postimpact sphere assembly (PISA), was subjected to a humid environment on sand in an environmental test chamber for 1 yr. The results of this experiment were reported in a topical report entitled "Interaction of an Iridium-Clad RTG Heat Source Unit with a Simulated Terrestrial Environment." ¹⁰

After this experiment was completed the plutonium oxide material in the PISA was sorted by particle size. One fraction of the material, consisting of all of the pieces greater than 6 mm in diameter (221 g), was placed on sand in environmental chamber No. 10, and the fraction of the material with diameters between 0.01 and 6 mm (32 g) was placed on sand in environmental chamber No. 7. These chambers were initially programmed for winter arid conditions. They have been operated in alternate 6-month periods of arid winter and arid summer and are now in their seventh winter. Each chamber has received monthly rains, except that there were no

TABLE XI
PLUTONIUM IN SOIL CORES 3-13 AND 3-14

Core 3-13			Core 3-14		
Section No.	Depth (cm)	Pu Found (ng)	Section No.	Depth (cm)	Pu Found (ng)
1	0-3	273	1	0-3	4.19×10^5
2	3-6	0.8	2	3-5	225
3	6-9	0.41	3	5-7	40.2
4	9-12	28.2	4	7-9	16.9
5	12-15	0.04	5	9-11	6.97
6	15-17	0.09	6	11-13	4.11
7	17-20	22.9	7	13-15	2.45
8	20-23	< 0.03	8	15-17	1.97
9	23-24	5.6	9	17-18	0.48
10	24-25	1.2	10	18-19	1.59
			11	19-21	0.45
			12	21-23	4.56
			13	23-24	6.1
			14	24-25	91.9
Total Pu		332			4.19×10^5
Pu, omitting section 1		59.2			403

TABLE XII
PLUTONIUM IN SOIL CORES 1-15 AND 1-16

Core 1-15			Core 1-16		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-3	378	1	0-1	187
2	3-6	56.9	2	1-6	2.3
3	6-9	3.62	3	6-10	13.5
4	9-11	1.12	4	10-13	4.49
5	11-13	1.22	5	13-16	2.27
6	13-15	2.85	6	16-18	0.64
7	15-17	0.08			
8	17-18	0.21			
Total Pu		444			210

TABLE XIII

PLUTONIUM IN SOIL CORES 1-17 AND 1-18

Core 1-17			Core 1-18		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-6	1.53×10^3	1	0-3	5
2	6-10	0.11	2-4	3-14	<0.03
3	10-14	0.06	5	14-17	0.04
4-5	14-18	<0.03	6	17-18	0.24

TABLE XIV

PLUTONIUM IN SOIL CORES 3-15 AND 3-16

Core 3-15			Core 3-16		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-2	1.05×10^4	1	0-3	1.24×10^5
2	2-4	0.87	2	3-6	24.9
3	4-7	0.36	3	6-10	20.8
4	7-10	0.35	4	10-13	1.05
5	10-13	0.04	5	13-16	0.14
6	13-17	0.03	6	16-19	0.59
7	17-19	<0.03	7	19-22	0.11
8	19-22	24.1	8	22-24	1.09
9	22-24	<0.03	9	24-25	2.00×10^3
10	24-25	0.27			
Total Pu		10.5 μg			126 μg

TABLE XV

PLUTONIUM IN SOIL CORES 1-19 AND 1-20

Core 1-19			Core 1-20		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-1	<0.2	1	0	0.2
2	1-7	1.32	2	0-3	2.32
3-4	7-15	<0.03	3	3-7	0.31
5	15-18	0.03	4	7-10	0.07
			5	10-13	0.11
			6	13-16	0.04
			7-8	16-18	<0.03
Total Pu		1.35			3.0

TABLE XVI

PLUTONIUM IN SOIL CORES 3-17 AND 3-18

Core 3-17			Core 3-18		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-3	34.8	1	0-2	74.1
2	3-5	1.16	2	2-5	0.11
3	5-8	0.33	3	5-8	0.07
4	8-10	0.09	4-9	8-25	<0.03
5	10-11	0.05			
6	11-13	0.04			
7	13-15	0.03			
8	15-16	0.05			
9	16-18	0.03			
10-14	18-25	<0.03			
Total Pu		36.6			74.3
Total Below Surface		1.78			0.18

TABLE XVII
CLIMATIC EFFECTS ON AMOUNT OF PLUTONIUM CARRIED
BY WATER PERCOLATING THROUGH SOIL
(Coarse and Fine Material from MHFT-27 Under Arid Conditions)

Climate	Coarse		Fine	
	Volume(l)	Pu(μ Ci)	Volume(l)	Pu(μ Ci)
Winter	10	0.19	9	0.08
Summer	4	0.04	6	0.2

rains during the first summer. The water that percolated through the soil after the first rain contained about 24 ng of plutonium in the chamber with the large pieces and 2.5 ng in the chamber with the fine material. The water aliquots from the chamber containing the large pieces show a wide variation in plutonium content, indicating the presence of relatively large particles of plutonium. The plutonium content decreased for the next three rains in the first winter, so that there was less than 1 ng in the fourth rain. The six rains in the second winter, after 6 months of summer without rain, averaged 12 ng of plutonium. The effects of arid climate changes (Table XVII) are qualitatively similar to those in humid conditions, except that with the fine material we find the average summer plutonium content is greater than the average for winter. In all other experiments, arid or humid, the winter plutonium averages are greater than the summer ones.

The plutonium contents of the dehumidifier condensates were similar to the contents of the early condensates from the two chambers containing material from MHFT-12. The condensates from the fine material decreased in plutonium content after the first rain, while the condensates from the large pieces that were collected for periods that included rains had much more plutonium than the other condensates from this chamber. This effect has decreased recently, partly because of an artifact in chamber operation. The rate of collection of plutonium in the dehumidifier condensates from the chamber containing the large pieces is 3 times that for the fine material.

Two soil cores were taken in each of these chambers at positions about 25 cm to the left and to the right of the PPO material before the first change to summer climate. The analyses for these cores are shown in Tables XVIII and XIX. Soil samples 3 and

4 were taken at the end of summer at positions 25 cm in front of and behind the PPO material, respectively, for each of these chambers. After four rains in the second winter, soil cores 5 and 6 were taken at positions approximately 25 cm to the front left and back right, respectively, of the PPO sources in the two chambers. The sand at the position of core 10-4 was so dry that, despite special precautions, a large fraction of it fell back into the hole while it was being sampled. For this reason only the top few centimeters of the soil were sampled, and no depth measurements could be made. About half of each of the following core sections was sent to LFE Environmental Analysis Laboratories for PuO_2 particle size analysis: sections 1, 3, 5, 7, and 9 of core 10-3 and sections 1, 3, and 5 of core 10-4. The rest of the material in the cores was dissolved and analyzed for plutonium (Tables XX - XXIII). The cores from the chamber containing the fine material had very little plutonium below the surface, before or after the rain. For the cores from the chamber containing the large pieces, the concentration of plutonium in the top few sections indicated that the plutonium oxide penetrated the soil several centimeters during the dry period. The analyses of the cores taken after rains began showed that the fine plutonia particles were washed through the soil by the rainwater.

Soil cores 7 and 8 were taken at the end of the second summer from each of these chambers, from positions approximately 25 cm to the front right and back left of the sources, respectively. Plutonium analyses from these cores (Tables XXIV and XXV) showed the normal summer pattern of nearly all the plutonium on the surface, with small but measurable amounts in all but one of the other sections and a slight increase in the bottom sections. There was much less plutonium in core 10-7 than in the other cores.

TABLE XVIII

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING FINE MATERIAL FROM MHFT-27

Left Core			Right Core		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-3	9.45	1	0-4	412.
2	3-7	0.19	2	4-10	0.16
3	7-10	<0.03	3	10-13	0.03
4	10-14	0.03	4-5	13-17	<0.03
5-10	14-28	<0.05	6	17-19	0.07
			7-9	19-23	<0.03
			10	23-25	0.04
			11	25-27	<0.03
			12	27-28	0.13
Total Pu		9.67			412.2
Total Subsurface Pu		0.22			0.43

TABLE XIX

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING LARGER PIECES FROM MHFT-27

Left Core			Right Core		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-6	3.30	1	0-1	3.97
2	6-13	<0.03	2	1-10	1087.
3	13-22	19.26	3	10-16	0.19
4-5	22-28	<0.03	4	16-21	0.05
			5-7	21-28	<0.05
Total Pu		22.56			1091.

TABLE XX

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING FINE MATERIAL FROM MHFT-27
AFTER 6-MONTH DROUGHT

Core 7-3			Core 7-4		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1-6	0-25	<0.03	1	0-11	693
7	25-26	0.05	2-10	11-28	<0.03
8-10	26-28	<0.03			
Total Pu		0.05			693

TABLE XXI

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING FINE MATERIAL FROM MHFT-27
AFTER RAINS RESUMED

Core 7-5			Core 7-6		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-2	0.24	1	0-2	86.4
2	2-6	0.04	2	2-8	0.10
3-12	6-28	<0.03	3	8-12	0.03
			4-11	12-28	<0.03
Total Pu		0.28			86.56

TABLE XXII

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING LARGER PIECES FROM MHFT-27
AFTER 6-MONTH DROUGHT

Core 10-3			Core 10-4		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-7	15.6	1		820
2	7-9	0.38	2		1010
3	9-13	108	3		2380
4	13-17	0.09	4		1380
5-10	17-28	<0.06	5		1
Total Pu		123.7			5591

TABLE XXIII

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING LARGER PIECES FROM MHFT-27
AFTER RAINS RESUMED

Core 10-5			Core 10-6		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-1	4.08	1	0-1	2770
2	1-4	0.04	2	1-4	0.46
3	4-7	0.04	3	4-8	0.11
4	7-10	0.05	4	8-10	0.04
5-9	10-27	<0.03	5	10-12	0.03
10	27-28	15 800	6	12-14	0.03
			7	14-16	0.04
			8	16-19	0.03
			9	19-20	<0.03
			10	20-23	0.03
			11	23-25	0.05
			12	25-27	0.08
			13	27-28	0.06
Total Pu		15 800			2771
Omitting first and last sections		0.13			0.90

TABLE XXIV

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING FINE MATERIAL FROM MHFT-27
DURING THE SECOND SUMMER

Core 7-7			Core 7-8		
Section No.	Depth (cm)	Pu (μ g)	Section No.	Depth (cm)	Pu (μ g)
1	0-2	5.73	1	0-2	1.62
2	2-6	0.0645	2	2-6	0.00014
3	6-8	0.00045	3	6-10	0.00011
4	8-11	0.00018	4	10-13	0.00006
5	11-14	0.0359	5	13-16	0.00009
6	14-18	0.00018	6	16-17	0.00003
7	18-21	0.00006	7	17-21	0.00004
8	21-23	0.00007	8	21-25	0.00025
9	23-25	0.00003	9	25-27	0.179
10	25-26	0.00006	10	27-28	0.0217
11	26-27	0.00007			
12	27-28	0.090			
Total Pu		5.92			1.82
Omitting first and last sections		0.1015			0.18

TABLE XXV

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING LARGER PIECES FROM MHFT-27
DURING THE SECOND SUMMER

Core 10-7			Core 10-8		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-3	71	1	0-8	1250
2	3-10	0.90	2	8-15	1.77
3	10-12	0.05	3	15-20	0.97
4	12-17	0.13	4	20-24	0.17
5	17-20	0.07	5	24-28	0.28
6	20-22	0.05			
7	22-24	0.06			
8	24-26	<0.03			
9	26-28	0.35			
Total Pu		73			1253
Omitting first and last sections		1.26			2.91

During the third winter, soil core samples 9 and 10 were collected from both of these chambers at positions to the left and right, respectively, of the plutonium dioxide sources, 30 cm from the fine material and 10 cm from the large pieces. The plutonium profiles of cores 10-9 and 10-10 (Table XXVI) both have peaks below the surface, and the total plutonium below the surface is quite low in core 10-9. The plutonium profile of core 7-9 has a large peak below the soil surface, while the subsurface plutonium content of core 7-10 is low (Table XXVII).

Core samples 11 and 12 were taken during the third summer in both of these chambers from positions 13 cm to the front and rear, respectively, of the plutonium oxide source. All of these cores showed subsurface peaks in their plutonium profiles, and in cores 7-12 and 10-11 most of the plutonium was below the surface (Tables XXVIII and XXIX).

3. *MHFT-50*. The fragments of PPO from the test sphere *MHFT-50* were placed in two environmental chambers equipped with specially designed soil compartments. One chamber contained 186 g of pieces greater than 2 mm in diameter, and the other contained 68 g of fine material, with particle diameters

between 0.01 and 2 mm. The soil in each chamber is a loam, and the soil tray is divided into two sections by a circular partition 45 cm in diameter in the center of the tray. The PPO material was positioned in the center of the circular section. The water that percolates through the soil in the two sections is collected separately for plutonium analysis. Initially the two chambers were on a winter cycle with humid conditions. They were changed alternately to summer and winter humid weather for 6-month periods until their seventh winter. The large pieces gradually crumbled until they were indistinguishable from the small stones in the soil.

Early in the seventh winter, the sources and the top 2 cm of soil were removed from both of these chambers. The alpha activity level of the soil was lowered in this process. The operation of the chambers is continuing in order to simulate conditions after a partial cleanup of an accident.

The volume of water that percolated through the soil was much lower in the inner compartment than in the outer compartment, especially in the chamber that contained the large pieces, because the outer compartment had three times the area and because the heat from the plutonium dioxide dried the soil in the inner compartment more extensively between

TABLE XXVI
PLUTONIUM IN SOIL CORES 10-9 AND 10-10

Core 10-9			Core 10-10		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-1	4.0×10^2	1	0-3	49.2
2	1-2	0.4	2	3-5	0.5
3	2-5	0.03	3	5-8	1.0
4	5-7	0.04	4	8-10	0.17
5-7	7-15	<0.03	5	10-11	0.08
8	15-17	0.35	6	11-12	0.04
9-14	17-28	<0.03	7	12-14	0.06
			8	14-17	0.13
			9	17-18	0.04
			10	18-20	0.04
			11	20-21	<0.03
			12	21-23	0.03
			13	23-26	0.07
			14	26-28	0.04
Total Pu		0.40 μ g			51.4 ng
Total below the surface		0.8 ng			2.2 ng

TABLE XXVII
PLUTONIUM IN SOIL CORES 7-9 AND 7-10

Core 7-9			Core 7-10		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-1	0.25	1	0-3	727
2	1-5	0.09	2	3-6	0.17
3	5-7	<0.03	3	6-9	0.27
4	7-10	30.8	4	9-12	0.04
5-11	10-26	<0.03	5	12-14	0.11
12	26-28	0.11	6-11	14-28	<0.03
Total Pu		31.2			728
Subsurface Pu		31.0			0.66

TABLE XXVIII
PLUTONIUM IN SOIL CORES 10-11 AND 10-12

Core 10-11			Core 10-12		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-5	124	1	0-2	6.3×10^3
2	5-14	266	2	2-7	71.4
3	14-17	0.63	3	7-15	2.29×10^3
4	17-20	0.03	4	15-20	248
5-6	20-27	<0.03	5	20-24	100
7	27-28	0.05	6	24-28	566
Total		391			9.6×10^3
Subsurface		267			3.28×10^3

TABLE XXIX
PLUTONIUM IN SOIL CORES 7-11 AND 7-12

Core 7-11			Core 7-12		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-4	4.3×10^2	1	0	0.54
2	4-6	0.33	2	0-2	5.7×10^2
3	6-10	0.06	3	2-5	507
4	10-13	<0.03	4	5-8	23.1
5	13-16	0.15	5	8-10	0.07
6	16-19	<0.03	6	10-12	0.07
7	19-21	0.03	7	12-15	0.06
8	21-25	2.18	8	15-18	0.05
9-10	25-28	<0.03	9	18-20	<0.03
			10	20-24	0.04
			11-12	24-28	<0.03
Total Pu		4.3×10^2			1.10×10^3
Subsurface Pu		2.75			1.10×10^3

rains (Table XXX). Both the volume of water per rain and the plutonium content for the inner compartment of the chamber containing the larger pieces were very low much of the time because of partial blockage of the outlet tube for this compartment. After the blockage was removed, the plutonium contents in the two compartments have been approximately equal, and the water volumes were roughly proportional to the surface areas.

Early in the present quarter the drainage of the inner compartment of chamber 6 (fine particles) became very slow. Forcing air backwards through the drainage system caused a greater flow but also a 100-fold increase in the plutonium. If there was a plug in the pipes, plutonium may have collected behind it, or more likely the soil was pulled away from the sides of the tray, permitting channelling of the water.

Before the first summer, water overflowed from the inner compartment to the outer one in the fine particle chamber because of a valve malfunction. This carried plutonium to the outer compartment, resulting in a much higher outer-to-inner ratio in the first summer for this chamber. This increase in the ratio was caused by a decrease in the plutonium content of the inner compartment, not by an increase in the outer compartment. The average outer compartment plutonium decreased by a factor of 2 for that summer, whereas inner compartment plutonium

decreased by a factor of 14. The flooding may have carried away the fine material that would have been washed through by the rains. A similar flooding incident during the fourth winter in this chamber did not affect this ratio. Evidently the plutonium had been "weathered" into the soil where it was not moved by surface water.

The rate of plutonium collection in the dehumidifier condensates from the large-piece experiment was 4 times that from the fine particle experiment. Two soil cores were taken from each chamber during the first humid winter cycle, core 1 just inside and core 2 just outside the circular isolating partition on the left side of the chamber.

Cores 3 and 4 were collected just inside and just outside, respectively, of the partition on the right side of the tray during the first summer. At the end of the second winter, soil cores 5 and 6 were collected just inside and just outside, respectively, of the partition at the front of the trays in both of these chambers. In the first pair of cores taken in the chamber containing the larger pieces from MHFT-50 (Table XXXI), the inner core is low in plutonium, while the outer one has a profile that shows peaks below the surface. The analyses of the other 10 cores during the first three seasons in chambers 6 and 8 (Tables XX-XII through XXXVI) showed normal profiles for the inner cores and low plutonium contents in the outer cores. At the end of the second summer core samples

TABLE XXX
EFFECT OF CLIMATE ON VOLUME AND PLUTONIUM CONTENT OF RAINWATER
PERCOLATES FROM CHAMBERS WITH PARTITIONED SOIL

Climate	Compartment	Coarse		Fine	
		Volume(ℓ)	Pu(μCi)	Volume(ℓ)	Pu(μCi)
Winter	Inner	1.5	0.06	2.	0.25
	Outer	7.7	0.12	8.	0.22
Summer	Inner	0.9	0.05	2.	0.09
	Outer	9.1	0.08	9.	0.16

TABLE XXXI
PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING LARGER PIECES FROM MHFT-50

Inner Compartment			Outer Compartment		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-3	144	1	0-3	253
2	3-5	0.11	2	3-7	0.08
3	5-7	0.05	3	7-11	49.4
4	7-8	<0.03	4	11-15	0.05
5	8-10	0.05	5	15-19	38.0
6-11	10-21	<0.07	6	19-22	0.08
			7	22-24	0.05
			8	24-27	0.04
			9	27-28	0.04
			10	28-30	0.04
Total Pu		144			341
Omitting first and last sections		0.21			87.8

TABLE XXXII

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING FINE MATERIAL FROM MHFT-50

Core 6-1			Core 6-2		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-2	1930	1	0-3	215
2	2-3	73.7	2	3-7	0.10
3	3-4	0.72	3	7-9	0.04
4	4-7	1.70	4	9-13	0.10
5	7-8	0.66	5	13-18	0.03
6	8-11	1.12	6-10	18-30	<0.03
7	11-12	0.58			
8	12-15	0.96			
9	15-18	0.42			
10	18-21	0.35			
Total Pu		2010			215
Subsurface Pu		80.2			0.27

TABLE XXXIII

PLUTONIUM IN SOIL CORES
(Fine Material from MHFT-50)

Core 6-3			Core 6-4		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-5	9340	1	0-4	629
2	5-10	150	2	4-9	0.09
3	10-14	10.8	3	9-13	0.05
4	14-20	5.9	4	13-17	0.08
5	20-21	0.12	5-6	17-26	<0.03
			7	26-29	0.12
			8	29-30	0.67
Total Pu		9506			630.0
Omitting first and last sections		167			0.34

TABLE XXXIV

PLUTONIUM IN SOIL CORES
(Larger Pieces from MHFT-50)

Core 8-3			Core 8-4		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-5	684	1	0-3	538
2	5-8	1.1	2-7	3-30	<0.03
3	8-10	0.20			
4-8	10-20	<0.03			
9	20-21	0.03			
Total Pu		685			538
Subsurface Pu		1.34			<0.03

TABLE XXXV

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING LARGER PIECES
FROM MHFT-50 COLLECTED DURING THE SECOND WINTER

Core 8-5			Core 8-6		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-2	2080	1	0-4	970
2	2-5	11	2	4-7	<0.03
3	5-9	0.35	3	7-11	0.20
4	9-13	0.08	4-5	11-30	<0.03
5-10	13-21	<0.03	6	21-25	0.03
Total Pu		2090	7-9	25-30	<0.03
Subsurface Pu		11			9.7×10^2
					0.23

TABLE XXXVI

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING FINE MATERIAL
FROM MHFT-50 COLLECTED DURING THE SECOND WINTER

Core 6-5			Core 6-6		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-3	4.08×10^3	1	0-5	280
2	3-5	25.6	2	5-10	0.12
3	5-8	7.14	3	10-16	0.03
4	8-10	4.11	4-6	16-30	<0.03
5	10-13	0.64			
6	13-15	0.10			
7	15-17	0.06			
8	17-20	0.03			
9	20-21	0.007			
Total Pu		4.12×10^3			280
Subsurface Pu		37.7			0.15

7 and 8 were taken in both chambers at positions behind the plutonium dioxide, 3 cm inside and 3 cm outside, respectively, of the partition. Plutonium analyses of these cores (Tables XXXVII and XXXVIII) showed that again the inner cores contained much more plutonium than the outer cores, and that most of the plutonium was in the first two sections of the inner cores and the first section of the outer cores. During the third winter, soil cores 9 and 10 were collected in both of these chambers from positions 3 cm inside and outside of the partition, respectively, to the front and left of the PPO sources. The plutonium profiles (Tables XXXIX and XL) had abnormally high fractions of the plutonium below the surface as well as subsurface peaks.

Soil cores 11 and 12 were collected during the third summer in both of these chambers from positions behind the plutonium dioxide 3 cm inside and outside, respectively, of the partition to the back right of the sources. The inner cores in both of these chambers have high subsurface concentrations of plutonium and subsurface peaks in the profile (Tables XLI and XLII). The subsurface plutonium concentrations in the outer cores are rather low, and the profiles are normal.

4. *MHFT-23.* We tested this fuel sphere assembly (FSA) in an environmental chamber on a sandy soil under humid conditions. Initially the system was

programmed for winter temperatures and humidities. The chamber was changed alternately to humid summer and humid winter conditions for 6-month periods until the FSA was removed from the chamber on April 17, 1978. The details of this experiment are described in a topical report.^{11>}

5. *Spalling Induced by Liquid Nitrogen.* Earlier work has demonstrated that the airborne plutonium concentration increases dramatically during a rain on large, thermally hot PPO pieces. This has always been presumed to be the result of thermal shock to the fuel surface, resulting in the spalling of tiny fragments of plutonia. We decided to find out if this cooling mechanism was a general phenomenon, or if water played a significant role in the process by means of a chemical reaction with the hot fuel.

We conducted an experiment where liquid nitrogen drops fell on chunks of PPO fuel in the environmental test chamber of Experiment 10. This liquid nitrogen rain continued for 5 min. Air samples were taken before, during, and after the rain. We found that the airborne plutonium activity increased by 4 orders of magnitude at the onset of the cooling period. About an hour after the end of the liquid nitrogen rain, the airborne plutonium concentration had dropped back to its prerin level of 0.1 nCi/m³.

TABLE XXXVII

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING FINE MATERIAL FROM MHFT-50
DURING THE SECOND SUMMER

Core 6-7			Core 6-8		
Section No.	Depth (cm)	Pu (μ g)	Section No.	Depth (cm)	Pu (μ g)
1	0-1	32.4	1	0-6	9.89
2	1-4	184	2	6-12	0.00262
3	4-6	12.7	3	12-18	0.00038
4	6-8	0.657	4	18-23	0.00035
5	8-10	1.71	5	23-27	0.00035
6	10-12	0.981	6	27-30	0.126
7	12-15	0.500			
8	15-17	0.655			
9	17-20	0.0955			
10	20-21	0.201			
Total Pu		234			10.02
Omitting first and last sections		201			0.0037

TABLE XXXVIII

PLUTONIUM IN SOIL CORES FROM CHAMBER CONTAINING LARGER PIECES FROM MHFT-50
DURING THE SECOND SUMMER

Core 8-7			Core 8-8		
Section No.	Depth (cm)	Pu (μ g)	Section No.	Depth (cm)	Pu (μ g)
1	0-2	39.0	1	0-4	6.75
2	2-5	50.9	2	4-7	0.00016
3	5-8	0.0266	3	7-11	0.00061
4	8-11	0.0086	4	11-15	0.00013
5	11-16	0.00116	5	15-20	0.0181
6	16-20	0.00159	6	20-23	0.00012
7	20-21	0.00660	7	23-25	0.00021
			8	25-28	0.00030
			9	28-30	0.00071
Total Pu		89.9			6.77
Subsurface Pu		50.9			20.3

TABLE XXXIX
PLUTONIUM IN SOIL CORES 6-9 AND 6-10

Core 6-9			Core 6-10		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-1	1.53×10^3	1	0-2	178
2	1-4	6.15×10^3	2	2-5	<0.05
3	4-8	234	3	5-8	4.31
4	8-11	20.5	4	8-13	1.83
5	11-14	16.4	5	13-15	23.7
6	14-16	11.1	6	15-19	0.17
7	16-19	2.31	7-9	19-26	<0.03
8	19-21	157	10	26-28	27.4
			11	28-30	<0.03
Total Pu		8121			235

TABLE XL
PLUTONIUM IN SOIL CORES 8-9 AND 8-10

Core 8-9			Core 8-10		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-2	630	1	0-3	2.3
2	2-5	631	2	3-6	0.53
3	5-10	17.5	3-6	6-18	<0.03
4	10-14	3.90	7	18-20	0.05
5	14-17	2.15	8	20-22	<0.03
6	17-21	19.9	9	22-24	0.30
7	21	0.06	10	24-26	0.38
			11	26-28	1.15
			12	28-30	<0.03
Total Pu		1305			4.7

TABLE XLI

PLUTONIUM IN SOIL CORES 6-11 AND 6-12

Core 6-11			Core 6-12		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-4	2.19×10^4	1	0-4	9.77×10^3
2	4-8	247	2	4-9	0.16
3	8-10	1.12×10^3	3	9-12	0.08
4	10-12	8.6	4	12-15	0.04
5	12-16	6.07	5	15-19	0.05
6	16-18	102	6	19-23	0.06
7	18-19	0.12	7	23-27	0.13
8	19-20	0.89	8	27-28	0.42
9	20-21	1.74	9	28-30	139
Total Pu		2.34×10^4			9.91×10^3
Without Sections 1 and 9		1.48×10^3			0.94

TABLE XLII

PLUTONIUM IN SOIL CORES 8-11 AND 8-12

Core 8-11			Core 8-12		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0	1.05×10^3	1	0-3	4.34×10^3
2	0-2	1.43×10^4	2	3-8	0.40
3	2-4	220	3	8-11	0.04
4	4-6	5.38	4-6	11-21	<0.03
5	6-7	2.22	7	21-24	0.06
6	7-9	0.51	8	24-27	<0.03
7	9-10	0.17	9	27-29	0.05
8	10-12	0.15	10	29-30	0.37
9	12-13	0.20			
10	13-15	0.06			
11	15-17	11.9			
12	17-19	5.11			
13	19-20	0.10			
14	20-21	3.70			
Total Pu		1.56×10^4			4.34×10^3
Omitting first and last sections		1.45×10^4			0.55

These results are quantitatively similar to those found with aqueous rains and suggest that hydrolysis reactions are not involved in the spallation process during an aqueous rain.

The air filters on which the plutonium was collected contained a short-lived alpha activity, in addition to the long-lived plutonium activity. This short-lived activity was not present in samples collected before the rain. It is possible that the thermal shock releases not only very fine particles of plutonia but also the radioactive gas ^{220}Rn (a daughter of ^{238}Pu) entrapped in the plutonium oxide crystal lattice structure. The activity ratio of ^{220}Rn to ^{238}Pu in this heat source material is 1×10^{-6} , which would account for finding only small amounts of the short-lived activity.

During this experiment we also measured the adsorption of the airborne plutonia on four different types of surfaces: glass, Teflon, stainless steel, and Lucite. Glass was the most effective collector of the airborne plutonia, adsorbing approximately 1000 times more than the Teflon or Lucite surfaces during the liquid nitrogen rain. Very little was found on the stainless steel surface.

We also made background measurements on a separate set of the same kinds of surfaces by exposing them to the chamber air for 48 h before the liquid nitrogen rain. The fact that significant amounts of alpha activity were found on the glass surface suggests that spontaneous spallation occurs continually, most likely caused by radiation damage in the plutonium oxide. This is probably the source of the low-level airborne plutonium found between rains in all chambers that contain bare plutonia.

A third set of adsorber measurements was made 1 day after the liquid nitrogen rain. These results showed far lower adsorption rates than existed before the liquid nitrogen rain. Apparently the thermal shock of the rain not only causes a great increase in spallation, but also frees loose particles created by radiation damage, so that at the end of the rain a fresh, undamaged surface is exposed.

Significant soil contamination was observed after the liquid nitrogen rain. The radioactivity, however, was found only on the soil surface and not beneath the surface. This is unlike soil contamination in the case of aqueous rains, where some of the plutonia particles are transported downward through the soil by the rainwater that drains through the soil. Thus hydrodynamic transfer is a significant transport mechanism under aqueous rain conditions.

6. *Terrarium Experiment.* A new chamber was fitted with special fluorescent lamps that should provide adequate light for growing plants in order to

make measurements of plutonium uptake. The lamps consist of four banks of two each, 40-W daylight fluorescent lamps. Although the photon flux will be somewhat lower than found in unobstructed sunlight, a variety of plants should grow readily in the artificial lighting. Blue grass was planted and has now reached maturity.

Another chamber has been set up as a terrarium to include simple animal life, such as earthworms, as well as vegetation. The grass planted in this one received a setback from a temperature excursion in the chamber. New grass has been planted and is now reaching maturity. Controls have been added to shut off the heat and refrigeration if a temperature excursion begins.

7. *Multiple-Soil Experiment.* This is an experiment with four different types of soil in one test chamber. The four soils are sand, silt loam, loam, and humus. The rainwater that percolates through each soil is collected separately for plutonium analysis. The source, ET1-1, is made from GROG-type fuel in the shape of a right circular cylinder, with a power rating of 15 W. The source is placed in a cup at the center of the tray containing the four soils. The cup drains into the dehumidifier condensate bottle.

Only 2 of the 24 samples of percolated rainwater collected from the first 6 rains contained plutonium. Both samples were from the loam soil sector, were collected after the second and fourth rain, and contained 2 to 3 μg of plutonium. This might indicate that if there is release of plutonium from this GROG-type source, the particles may be too large to be washed through the soils.

Since the first rain, all of the dehumidifier condensate collections have contained plutonium. The rates of plutonium collection during periods that included rains averaged 25 times as great as those for periods without rains. The higher value during rains may have some contribution from airborne spalling, but there is no doubt that most of the plutonium found in the condensates after rains has been washed off by the rainwater after being loosened by radioactive recoil processes at the surface of the source.

III. SORPTION OF PLUTONIUM BY SOILS

(Evangeline M. Hodge and George M. Matlack)

Three soil columns are operating to test the transport of plutonium derived from plutonium oxide particles on the soil surface. The soil columns are made of silt loam soil, approximately 14 cm high and 2.4 cm in diameter. These columns are identified as 7, 8, and 9. Column 7 has 3.5 mg of PuO_2 particles on its soil surface, with a particle size distribution mainly in the 40- to 60- μm range. Column 8 contains 2.2 mg of PuO_2 particles with an average diameter of 12 μm , and column 9 contains 2.6 mg with an average diameter of 7 μm . These three size ranges are from the same three batches of plutonia particles that we are using in the experiments for measuring dissolution rate as a function of particle size (Sec. V). The columns are being eluted with distilled water (a rainwater simulant). The columns have been in operation for 1596 days. Measurements made after 1506 and 1596 days of operation (Table XLIII) show that the flow rates and the quantity of plutonium released have not changed significantly. The flow rates range from 0.4 to 0.8 $\mu\text{l/s}$. The variation in flow rate between columns is unexpected, inasmuch as they all contain soil from the same source. Radiation measurements show that at least 99% of the PuO_2 particles remain at the top of each column; radiation cannot be detected from any particles that may have moved downward into the soil columns.

Three additional columns have been underway for 415 days to measure the absorption of plutonium in rainwater by sandy soils. The feed solution to the columns is a dilute, neutral solution of plutonium chloride (15 $\mu\text{g/l}$), known to be unstable as it ages. This type of feed solution is necessary, however, to simulate rainwater contaminated with plutonium as it percolates through soils. Instead of using a gravity flow of solution through the columns, we are using a reverse flow upwards through the columns, with the flow rate controlled by a peristaltic pump. This ensures better reproducibility of experimental conditions between columns. The flow rates are all 2.12 $\mu\text{l/s}$, and the columns have retained 99.36 to 99.46% of the plutonium applied to them (1048 μg) so far.

IV. BEHAVIOR OF ^{238}Pu IN AQUATIC ENVIRONMENTS

(George M. Matlack, Gilbert B. Nelson, Frederick Steinkruger, and James H. Patterson)

The behavior of ^{238}Pu fuels in fresh water and seawater, especially rates of plutonium release, is being investigated in aquaria and in natural sea environments. Although the solubility of hydrated PuO_2 is extremely low, corresponding to about one atom of plutonium in 100 000 l of water at pH 7, other interactions between water-solvent systems and typical ^{238}Pu fuel can release plutonium at a low but finite rate. Measurements of these release rates from various fuels in differing circumstances provide data for assessing the risk to the biosphere from fuel container failure in the ocean or in fresh water and for devising means to prevent unacceptable risks.

A. Seawater Aquarium Experiments

Each aquarium contains 75 l of simulated seawater, made by dissolving a commercial mixture of salts in distilled water, and maintained at pH 8.1 to 8.3. A refrigeration system keeps the water at 10°C when a 40-W heat source is immersed in the water and the room temperature is 25°C. Water is circulated by an air bubble stream behind a baffle in each rear corner. Each aquarium has a cover sealed at the sides with silicone rubber cement to contain airborne plutonium. Water samples are removed through a small hole in the cover.

B. Ocean Test Facility

Samples in specially designed steel chambers are submerged in 25 m of seawater off San Clemente Island, 100 km west of San Diego, at a Naval Ocean Systems Center facility. Each chamber is approximately 50 cm in diameter and 25 cm deep, and the inside is covered with an asphaltum-like coating. The chamber is weighted on the bottom by a circular slab of concrete. The open top is protected by a conical metal canopy about 60 cm high. Water can enter only through the perforations in the canopy.

The sea floor in the test area, approximately 500 m from shore, is sandy, with rocks and abundant patches of seaweed. Each test chamber is filled with local sand and seawater, and the sample is placed about 2.5 cm beneath the sand surface before the chamber is lowered to the ocean floor.

No samples are currently under test.

TABLE XLIII
Pu TRANSPORT FROM PuO₂ PARTICLES

Column	Elution Duration			
	1506 days		1596 days	
	Total Vol. (ℓ)	Total Pu (ng)	Total Vol. (ℓ)	Total Pu (ng)
7	106.5	36.6	112.0	38.2
8	93.0	36.2	97.9	38.2
9	51.9	45.8	55.2	46.9

C. PMC Dissolution or Release Rates

These experiments have been terminated. A topical report has been issued as an LA document.¹²

D. PPO Dissolution or Release Rates

These tests are to compare release rates from PPO pellets in both fresh water and seawater. Five experiments are under way with various sizes of test pellets.

1. In Seawater The 25-W PPO pellet (Table XLIV) HPZ-59-4 was removed from the water in March 1981. The water shows a release rate after 2350 days of 8 nCi/m²-s, down 25% from the previous quarter's rate.

The 18-W pellet HPZ-174 has been immersed in seawater at 10°C for 1245 days. The release rate is 5 nCi/m²-s, unchanged in the last 9 months. This is the same system in which we maintained a live sea cucumber for 230 days.

A second experiment, this time with 3 sea cucumbers, has been underway for 257 days. The animals are in 10°C seawater that contains a broken GROG

TABLE XLIV
SUMMARY OF
Pu RELEASE RATES FROM PPO IN AQUATIC ENVIRONMENTS

Sample	Power (W)	Time (Days)	Water	Temperature (°C)	Release (nCi/m ² -s)
HPZ-60-2	2.5	2551	Fresh	10	144
HPZ-111-1 *	25	2256	Fresh	10	440
HPZ-59-4	25	2350	Sea	10	6
HPZ-174	18	1245	Sea	10	5
HPZ-186-4	19	1241	Sea (tidal)	10	52
HPZ-186-2	15	257	Sea	10	2

* Fuel pellet removed from the water in March 1981.

pellet, HPZ-186-2, that weighs 32 g, equivalent to 13 W of thermal power. The fuel piece is supported on a glass pedestal that prevents direct contact of the animals with the fuel. The sea cucumbers will be removed sequentially at yearly intervals for analysis of the plutonium each has ingested from the seawater. After 257 days immersion of the fuel pellet, the release rate was 2 nCi/m²-s, a rate which is unusually low for cold seawater, but unchanged from the previous quarter.

A tidal simulation aquarium, using the 19-W pellet HPZ-186-4, has been under way for 1241 days. The source is on sand in a stainless steel tray that previously was lowered into the water and raised above the water twice every 24 h. Seventeen months ago, however, the elevator mechanism broke, so that the tray and source are now continually immersed in the water. The plutonium release rate dropped since the end of February 1978 from 440 to a minimum of 25 nCi/m²-s by February 1979. The pellet has gradually cracked and broken into 30 or more pieces since its initial immersion, probably the result of the thermal shock that occurred when it entered the water each time. As a result of this breakup, the release rate is 52 nCi/m²-s, up 27% from last quarter's value.

2. *In Fresh Water.* Plutonium release from the 2.5-W pellet HPZ-60-2, which was removed from the water in March 1981 to fresh water at 10°C has averaged 144 nCi/m²-s for 2557 days (see Table XLIV). This is up unchanged from last quarter and is approximately 15 times greater than the rate typical of the PPO pellets in 10°C seawater.

The release rate from the 25-W pellet HPZ-111-1 has averaged 440 nCi/m²-s for 2256 days, which is the highest value of the test pellets. This rate is up 7% from last quarter's value. The pellet was removed from the water in March 1981. This high rate is consistent with the previous observation (see Table XLIV) that the highest release rates are found in fresh water and the lowest in warm seawater.

3. *Total Release Rate (Source Term).* The plutonium release rates in Table XLIV describe only the plutonium suspended in the water, and not that deposited on underwater surfaces. A total release rate, or source term, is not obtainable in an aquarium experiment. Therefore, a glass chamber was designed and constructed to permit such measurements.

A 30-g, iridium-clad ²³⁸PuO₂ source, HPZ-31-2, was placed in the glass chamber on July 25, 1977, and covered with 2.3 l of simulated seawater. The water was kept at 10°C. The source, previously used in an ocean floor experiment, was known to have a pinhole leak in the cladding. The water was sampled in duplicate periodically for plutonium analyses.

The analytical data confirm that the source was leaking and demonstrated that small plutonium oxide particles were being released. The release rate was also erratic. Variations in the plutonium concentration in duplicate samples indicated that the source was releasing particulate material of varying sizes.

Initially the plutonium concentration in the seawater increased steadily. From day 21 to day 31 the concentration was essentially constant. From day 32 to day 39 the release rate increased sharply and from day 40 to day 60 the concentration exhibited a slow and steady decrease. On day 63 the plutonium concentration was 60% higher than on day 60. The erratic nature of the release rate can be explained by alternate sealing and venting of the pinhole leak in the iridium clad. During the period of slow decrease in the plutonium concentration, the leak was sealed, and deposition processes were removing plutonium from the water. During periods of increasing plutonium concentration, the leak was venting although the degree of venting varied.

This experiment was terminated after 101 days, and the source was removed for metallurgical examination. The simulated seawater was removed and the walls and bottom of the chamber were gently

TABLE XLV
COMPARISON OF Pu LEVELS IN VARIOUS FRACTIONS OF THE
SOURCE TERM EXPERIMENT

<u>Sample</u>	<u>Pu(μg)</u>	<u>Comment</u>
16-54	10.9	Routine Water Sample
Seawater	14.5	Seawater and Rinse Combined
Leach	0.60	Acid Leach of Walls

rinsed. The seawater and the rinse water were combined, acidified with nitric and hydrofluoric acids, and digested to produce a homogeneous solution. The interior of the glass chamber was leached with acid to remove any plutonium adhering to the walls. Table XLV compares the results from the various fractions to those from sample 16-54, taken immediately before removal of the source. Combination of the seawater and leach fractions yields a total of 15.1 μg of plutonium, 38% higher than the quantity indicated by the routine water sample. Most of the additional plutonium appears to be material that settled to the bottom of the chamber or was loosely adhering to the walls, since a gentle rinse was sufficient to recover the material.

Metallurgical examination revealed that the fuel source had broken into four major pieces, two small pieces, and a small quantity of fine material. Thus the total area is unknown, and a reliable release rate cannot be calculated.

A 48.6-g, bare $^{238}\text{PuO}_2$ source, HPZ-186-2, was positioned in an empty glass chamber on December 20, 1977, and the chamber was placed in an aquarium at 10°C for temperature control. This experiment will provide base-line data for an unclad source exposed to air only. Such conditions occur in some of the environmental chambers. A 0.05- μm pore-size filter is changed weekly and analyzed for plutonium.

This experiment was terminated on day 454. Only 2 filters out of 63 had shown any plutonium, 6 pg on one and 2 pg on the other. These two results were not confirmed by subsequent filters and thus are attributed to contamination from an unknown source external to the experiment. The heat source was transferred to a second chamber and the first chamber was resealed and transferred for analysis. The source in the 2nd chamber was submerged in deionized water for 30 min and then removed. At this point the heat source fractured into three pieces and further experiments with this source were cancelled. The second chamber has been resealed in preparation for analysis.

Analysis of the first chamber consisted of the quantitative recovery of the plutonium on the interior surfaces. This was accomplished by using a nitric-hydrofluoric acid leach, followed by a wash with a decontamination solution. No residual activity could be detected on the surfaces that were accessible for monitoring.

The total plutonium recovered was 1.41 μg , which represents a release rate of 0.022 $\text{ng}/\text{m}^2\text{-s}$. This number is much lower than the release rates observed in the aquatic environmental experiments, which range from 0.6 to 36 $\text{ng}/\text{m}^2\text{-s}$. The analysis of the second

part of the experiment in which the source was submerged indicated a total release of 49 μg . Calculations based on the same surface area and a 30-min submergence yielded a release rate of 17 $\mu\text{g}/\text{m}^2\text{-s}$. This calculation may be invalid because the initial release may be that from loosed material on the "old" surface of the source. The next experiment, designed to measure the release from a "new" surface of the same source, could not be performed, because the source broke into three pieces of indeterminable total surface area.

Four experiments in glass chambers are under way using GROG sources containing approximately 40 g of plutonium in each. The four experiments consist of a fresh water and a simulated seawater environment at 10°C and at 35°C. The water is aerated and stirred by bubblers. The filters used on the 35°C experiments are 3.0 μm , and those on the 10°C experiments are 0.6 and 5.0 μm (seawater).

The plutonium concentration versus time for the four experiments are illustrated in Fig. 5, which shows that the highest plutonium concentration occurs in the cold fresh water system. This trend agrees with previous aquarium results. The warm fresh water system exhibits a plutonium concentration that is a factor of 5 lower than the cold fresh water results. This is the first time we have run release rate experiments in warm fresh water.

Also shown in Fig. 5 are the results of hot and cold simulated seawater experiments. In these systems, the cold experiment exhibits a higher plutonium concentration than the warm seawater experiment. This trend is consistent with previous seawater experiments in aquariums.

Four new aquatic experiments were started during the last quarter. These experiments, using 15-W GROG fuel pellets, duplicate the conditions in the sealed glass chambers. Insufficient data are available at this time to establish trends. In both of the present seawater systems the plutonium concentration is lower than that of the cold fresh water experiment, which is consistent with previous results in aquariums.

4. Hydrostatic Test Facility. The results of the 300-day immersion of the GIS assembly, MHF-259, in the 69 MPa (10 000 psi) seawater test experiment, have been published in a topical report.¹³

A second high pressure test facility, capable of operating at 103 MPa (15 000 psi) with a test volume of 8 ℓ , has been received from Autoclave Engineers, Inc. This will be used for tests at simulated ocean depths of 9100 m.

E. Clad Sphere in Simulated Seawater

The plutonium release from a 100-W PISA, MHFT-13, was measured during immersion in simulated seawater for 375 days. Before immersion in the test aquarium, the PISA was cooled in a beaker of seawater at room temperature to eliminate the hazard of handling a hot object outside a glove box. During its immersion in 20°C seawater, we found essentially no plutonium (less than 10 ng) in the water. The PISA gradually became a spotty brown, probably because of deposition of iron or manganese hydroxides, but no gross defects were visible. However, detailed metallography after the PISA was removed from the water revealed cladding defects that we believe were caused by seawater that entered the helium release vents when the sphere was first submerged. The defects appeared to be ruptures adjacent to the external closure weld. The inside of the cladding showed corrosive attack along grain boundaries, sometimes penetrating to 30% of the wall thickness, presumably caused by the seawater, but metallography of typical specimens showed no evidence of sodium chloride. The plutonia sphere showed no sign of corrosion. Probably, sudden immersion of the hot PISA caused water

penetration through the vents in the cladding and the water and the salt residue, together with the high temperature and intense alpha and gamma radiation, caused chemical attack and disintegration of the iridium cladding from within.

V. PLUTONIUM RELEASE AS A FUNCTION OF PARTICLE SIZE

(Gilbert B. Nelson, Frederick J. Steinkruger and George M. Matlack)

Three sets of sized $^{238}\text{PuO}_2$ particles were prepared, and weighed amounts of these particles were placed in 50-ml volumetric flasks containing 1 M HClO_4 in a 37°C water bath. (During this report period the heater system failed.) Periodically samples are removed, centrifuged to remove suspended particles, and then analyzed for plutonium. These data are needed to obtain information about dissolution under carefully controlled solution conditions and to determine the effects of particle size on dissolution rates.

TABLE XLVI
SUMMARY OF DISSOLUTION EXPERIMENTS IN 1M HClO_4

Experiment No.	PuO_2 (mg)	Per Cent Dissolved	Dissolution Rate	
			(g/s)	(g/s)
A1	2.02	6.3	7.6×10^{-13}	3.7×10^{-10}
A2	2.05	5.7	7.0×10^{-13}	3.4×10^{-10}
A3	10.19	5.8	3.6×10^{-12}	3.5×10^{-10}
A4	9.90	5.6	3.3×10^{-12}	3.3×10^{-10}
B1	2.02	10.1	1.2×10^{-12}	6.1×10^{-10}
B2	2.03	12.0	1.5×10^{-12}	7.2×10^{-10}
B3	10.25	9.1	5.6×10^{-12}	5.4×10^{-10}
B4	10.45	10.5	6.5×10^{-12}	6.3×10^{-10}
C1	2.23	21.7	2.9×10^{-12}	1.3×10^{-9}
C2	2.31	20.4	2.8×10^{-12}	1.2×10^{-9}
C3	9.89	21.0	1.2×10^{-11}	1.3×10^{-9}
C4	10.67	21.0	1.3×10^{-11}	1.3×10^{-9}

The data (Table XLVI) were collected for 1960 days for the three particle size distributions. Each of these particle size distributions was used to prepare four experiments, two containing 2 mg of particles and two containing 10 mg. Distribution A consists of the largest particles, having a size range of 1-120 μm . Distributions B and C represent continually smaller sizes.

A comprehensive review of this experiment, including a more detailed characterization of the three particle distributions, has been completed. The review revealed that the large particles in distribution A were actually aggregates of smaller particles. The suspension technique used in the particle sizing produced a combination of "large particles" (did not suspend) and small particles thought to be individual members of aggregates that had broken apart. It is not possible to determine a quantitative estimate for the amount represented by the large fraction or the small fraction; therefore, the calculation of an available surface area is not possible.

Distribution B contained both large and small particles. In this case the large particles consisted of a preponderance of single large particles with some fines adhering to them, whereas the large particles in distribution A consisted of aggregates of similar size particles. Again it is not possible to determine a total surface area because the sizing technique does not produce a quantitative estimate for the amount represented by each fraction.

Distribution C behaved normally in that all particles could be suspended and there were not anomalous large particles. This distribution lends itself to a calculation of total surface area, but there is no assurance that the initial area has remained constant. In fact, the surface area has probably decreased during the experiment.

An examination of the grams dissolved as a function of time for experiments A-1, B-1, and C-1, shows no discrepancies in the three experiments. Dissolution is fastest for the C distribution and slowest for the A distribution. In addition, the three experiments show different slope changes in their three dissolution curves. The experiment A-1 (coarse particles) shows a shallow minimum at about 350 days;

experiment B-1 (medium particles) shows a low maximum at about 230 days; and experiment C-1 (fine particles) shows a strong maximum at about 230 days, which has since decreased by 30%. The maximum is 3 times that of B-1. We believe that the initial rapid increase in the dissolution rates for C-1 represents the solubilization of the smallest particles. The rate is now decreasing because of the reduction in surface area, since only "larger" particles remain.

The third column in Table XLVI lists the percent of original material that has dissolved. Since a substantial part of the original material has dissolved, especially in the C experiments (20%), the size distributions may have changed considerably. We therefore plan to terminate these experiments.

An extraction procedure, designed to extract only Pu(IV), is being applied to these experiments. Preliminary results indicate that the plutonium in solution is 90%, or greater, in the ionic form. Work is continuing in this area.

VI. OCEAN WATER SAMPLING

(Frederick J. Steinkruger and Gilbert H. Nelson)

Two water samplers were placed in the ocean at San Clemente Island on May 15, 1979. Each sampler was designed to collect 1.5 ℓ of water over a period of 3 months to ascertain the level of plutonium contamination in the seawater at the site of previous ocean immersion tests of ^{238}Pu heat sources. The samplers were removed from the ocean and received at Los Alamos in late September 1979.

Sampler No. 1 collected a total of 0.44 ℓ . The less-than-maximum collection was caused by corrosion and clogging of the automatic collecting mechanism. Preliminary analyses of the collected water give an average plutonium level of 0.12 nCi/ ℓ . This is about 3% of the MPC for public drinking water, 5 nCi/ ℓ .

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