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Interaction of $^{238}\text{PuO}_2$ Heat Sources with Terrestrial and Aquatic Environments

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QUARTERLY REPORT
ENVIRONMENTAL AND RADIOLOGICAL SAFETY STUDIES

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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 Office of the Assistant Director for Space Application of the ERDA Division of Nuclear Research and Application 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 details of the experimental testing are minimized, and they will be largely repetitive in succeeding reports to emphasize the main purpose, compilation of all usable data generated in each experiment. This compilation includes 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 thermoelectric generators (RTGs). Because the half-life of ^{238}Pu is relatively short (87.5 yr), the alpha-particle specific activity 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 Office of the Assistant Director for Space Application of the ERDA Division of Nuclear Research and Application continually seeks more information about the heat sources to improve their safety. As part of this program, LASL 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 LASL 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 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 m³ 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 as high as 30 μm/s (~ 4 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.

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). Pressure plate moisture measurements are also planned for these soils.

TABLE I
COMPOSITION OF SOILS USED
IN TERRESTRIAL EXPERIMENTS^a

	Loam	Clay	Silt Loam	Sand
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	6.3
C, organic (%)	0.6	0.12	0.03	0.01
Carbonate as CaCO ₃ (%)	trace	8	10	4
H ₂ O at 15 bars (%)	7.5	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

^aSoil analyses provided by the U. S. Soil Conservation Service Laboratory, Riverside, CA.

C. Test Conditions and Analyses Performed

The first three 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. The experiment in the chamber containing MHFT-27 was terminated (see Sec. II.D.2). Subsequently two additional chamber experiments were operated alternately under arid winter conditions (3 to 28°C, 21 to 87% RH) and arid summer weather (26 to 53°C, 7 to 33% RH). Three other experiments were started under humid winter conditions. All seven current chamber experiments are now operating under summer conditions, two in arid climate, the other five under humid conditions.

The fuel material sample is placed in the center of the tray on the soil surface. 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 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 is also analyzed to get a relative indication of the airborne plutonium in the chamber. Core samples of the soil are collected at approximately 6-month intervals. Each core is divided into 5 to 14 sections that are 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 -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 currently under way. They are summarized in Table II.

1. *MHFT-12*. This was a PPO test sphere for the Multi-Hundred Watt (MHW) program which shattered during a simulated reentry and impact. 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 in one environmental test chamber on June 21, 1973, and 28 g of 0.01- to 6-mm-diam material was placed in another on September 27, 1973. These experiments are now in their fifth summer 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

TABLE II
Terrestrial Environmental Chamber Experiments

Experiment Number	Description	Date Started
1	MHFT-12 chunks on loam; humid climate	6-21-75
2A	MHFT-25 in GIS on sand; humid climate	4-10-75
3	MHFT-12 fines on loam; humid climate	9-27-75
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

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

analysis in the dehumidifier condensates. Analyses for a period of 2 months are shown in Fig. 1. The explanation seems to be 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.⁴ 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 samples were taken continuously during a rain sequence. The average airborne plutonium concentration during the first 5 min of rain was 10^4 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 some of the 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

Fig. 1.
Effect of rain on plutonium content of dehumidifier condensates.

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 ($\mu\text{Ci}/\text{m}^3$)	Remarks
1	24	3	Before rain
2	5	3×10^4	First 5 min of rain
3	5	6×10^3	
4	10	4×10^3	
5	10	5×10^3	
6	15	9×10^2	
7	15	8×10^2	Last 15 min of rain
8	50	2×10^2	
9	180	3	
10	860	3	

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 2π 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 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 four 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 $1.8 \mu\text{Ci}$ of plutonium. This very rapid plutonium breakthrough indicated that some of the material that spalled from the fuel was in the form of very small particles or a colloid. 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 some ion-exchange type of mechanism.

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 liters of water. Under summer conditions, about 6 liters of rainwater percolated through the soil. This water contained an average of $0.6 \mu\text{Ci}$ 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 $18 \mu\text{Ci}$ 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. 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

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

Climate	Conditions	Average Percolated Water Per Rain (liters)	Average Pu (μCi)
Winter	0 - 17°C	11	9
	60 - 100% RH		
Summer	20 - 30°C	6	0.7
	85 - 97% RH		

TABLE V

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

Climate	Average Volume per Rain (liters)	Average Pu μCi
Winter	11	0.19
Summer	6	0.13

much smaller but also showed little change with climate (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 PuO₂. 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 were also measured using an air sampler. 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

TABLE VI

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

Filter No.	Collection Time (min)	Pu Concentration (μCi/m ³)	Remarks
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	

probably indicated that soil wetting inhibited resuspension of plutonium-bearing particles. The counting rates were so low that we had to combine the five filters used during the rain to obtain meaningful statistics. The same treatment was necessary for the first five filters used after the rain. There was very little seasonal effect on the plutonium content of the rainwater that percolated through the soil; it averaged about 0.2 μCi both winter and summer.

We analyzed eight soil cores from each of these two chambers for plutonium. The results and core locations 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 LASL for plutonium. Zeros indicated that less than 0.02 ng of plutonium was found.

The data in Figs. 2 and 3 indicate 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 con-

seems to have been caused by water released from a gate valve directly above core position 1 whenever summer humid conditions 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 were 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 experi-

ment 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 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.

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 particles, 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 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 inadver-

tently 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 25 pg 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.

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	0-5	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
			6	17-18	33.9
Total Pu		5.80			168.9

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 section, 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. 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.⁸ 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 1-3, 1-4, 1-12, and 1-13. (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 1-1, 1-5, 1-14, 3-1, 3-3, 3-5, 3-6, 3-7, and 3-8 have this characteristic. All of 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

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 WHFT-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	0.33	8	15-17	0.04
10	20-22	10.7	9	17-19	5.43
11	22-23	3.59	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
					2.15×10^5

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.

Analysis of soil cores 3-13 and 3-14 (Table XI) indicates that core 3-14 has a normal plutonium profile, while core 3-13 has an abnormal profile because it has two subsurface peaks. During the

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
Total Pu		332	11	19-21	0.45
Pu, omitting section 1		59.2	12	21-23	4.56
			13	23-24	6.1
			14	24-25	91.9
					4.19×10^5
					403

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.

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 one year. 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." *

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 in environmental chamber No. 10, and the fraction of the material with diameters between 0.01 and 6 mm (32 g) was placed 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 fourth summer. Each chamber has received monthly rains, except that there were no 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 showed 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 month of summer without rain, averaged 12 ng of plutonium. The effects of arid climate changes (Table XII) are qualitatively similar to those in humid conditions, except that with the fine material we find the water volume relatively unchanged, while under humid conditions, it is the amount of plutonium that is relatively unchanged.

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.

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 XIII and XIV. 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. Roughly half of each of the following core sections was sent to LFE En-

TABLE XII
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	12	0.20	9	0.10
Summer	4	0.02	7	0.04

TABLE XIII

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

TABLE XIV

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.

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 XV through XVIII). 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 a few 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 XIX and XX) showed the normal 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.

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.

TABLE XV

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	<0.030	1	0-11	693
7	25-26	0.053	2-10	11-28	< 0.030
8-10	26-28	<0.030			
Total Pu		0.053			693

TABLE XVI

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	0.245	1	0-2	86.43
2	2-6	0.038	2	2-8	0.102
3-12	6-28	<0.030	3	8-12	0.0266
			4-11	12-28	<0.030
Total Pu		0.28			86.56

TABLE XVII

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.	Pu (ng)
1	0-7	15.6	1	819
2	7-9	0.382	2	1009
3	9-13	107.6	3	2381
4	13-17	0.089	4	1384
5-10	17-28	<0.060	5	1.14
Total Pu		123.7		5594

TABLE XVIII

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	4.081	1	0-1	2768
2	1-4	0.042	2	1-4	0.462
3	4-7	0.038	3	4-8	0.108
4	7-10	0.046	4	8-10	0.045
5-9	10-27	<0.030	5	10-12	0.0276
10	27-28	15 800	6	12-14	0.032
			7	14-16	0.045
			8	16-19	0.030
			9	19-20	< 0.03
			10	20-23	0.028
			11	23-25	0.048
			12	25-27	0.080
			13	27-28	0.057
Total Pu					
		15 800			276^

TABLE XIX

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 (μg)	Section No.	Depth (cm)	Pu (μg)
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	0.000185	4	10-13	0.00006
5	11-14	0.0359	5	13-16	0.00009
6	14-18	0.000179	6	16-17	0.00003
7	18-21	0.000059	7	17-21	0.00004
8	21-23	0.000068	8	21-25	0.00025
9	23-25	0.000033	9	25-27	0.179
10	25-26	0.000057	10	27-28	0.0217
11	26-27	0.000073			
12	27-28	0.090			
Total Pu					
		5.92			1.82

TABLE XX
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 (μg)
1	0-3	71	1	0-8	1.25
2	3-10	0.90	2	8-15	0.00177
3	10-12	0.054	3	15-20	0.000971
4	12-17	0.126	4	20-24	0.000169
5	17-20	0.066	5	24-28	0.000284
6	20-22	0.048			
7	22-24	0.056			
8	24-26	<0.03			
9	26-28	0.354			
Total Pu		73			1.25

3. *MHFT-50*. The fragments of PPO from the test sphere *MHFT-50* are in two environmental chambers equipped with specially designed soil compartments. One chamber contains 186 g of pieces greater than 2 mm in diameter, and the other contains 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 in summer humid weather and winter for 6-month periods, and are now in their third summer.

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 rains. Both the volume of water per rain and the plutonium content for the inner compartment of the chamber containing the larger pieces have been very low most of the time because of par-

tial blockage of the outlet tube for this compartment. In the first five rains 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. Before the first summer, water overflowed from the inner compartment to the outer one in the fines 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 ratio has gradually decreased to approximately the average for the first winter.

The plutonium in the dehumidifier condensates from the large-piece experiment was 5 to 10 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. The analyses for these cores are given in Tables XXI and XXII. 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. The analytical results for these are listed in Tables XXIII and XIV. 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. Tables XXV and XXVI list the analysis data from

TABLE XXI

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

Inner Compartment			Outer Compartment		
Section No.	Depth (cm)	Pu (ng)	Section No.	Depth (cm)	Pu (ng)
1	0-3	143.8	1	0-3	253
2	3-5	0.106	2	3-7	0.083
3	5-7	0.054	3	7-11	49.4
4	7-8	<0.03	4	11-15	0.050
5	8-10	0.046	5	15-19	38.0
6-11	10-21	<0.07	6	19-22	0.076
			7	22-24	0.043
			8	24-27	0.042
			9	27-28	0.043
			10	28-30	0.041
Total Pu		144.0			341

TABLE XXII

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	1929.	1	0-3	214.9
2	2-3	73.7	2	3-7	0.10
3	3-4	0.72	3	7-9	0.041
4	4-7	1.7	4	9-13	0.10
5	7-8	0.66	5	13-18	0.034
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		2009			215.2

TABLE XXIII

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	9337	1	0-4	629.0
2	5-10	149.9	2	4-9	0.090
3	10-14	10.8	3	9-13	0.050
4	14-20	5.9	4	13-17	0.070
5	20-21	0.12	5-6	17-26	<0.03
			7	26-29	0.10
			8	29-30	0.67
Total Pu		9504			630.0

TABLE XXIV

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	0.031			
Total Pu		685			538

TABLE XXV

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	2.08×10^3	1	0-4	9.7×10^2
2	2-5	11	2	4-7	< 0.03
3	5-9	0.35	3	7-11	0.20
4	9-13	0.08	4-9	11-30	< 0.03
5-10	13-21	< 0.03			
Total Pu		2.09×10^3			9.7×10^2

TABLE XXVI
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	4.08x10 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		4.12x10 ³			280

these cores. At the end of the second summer, core samples 7 and 8 were taken in both chambers at positions 3 cm inside and 3 cm outside, respectively, of the position behind the plutonium dioxide. Plutonium analyses of these cores (Tables XXVII

and XXVIII) showed that 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.

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

Core 6-7			Core 8-8		
Section No.	Depth (cm)	Pu (μg)	Section No.	Depth (cm)	Pu (μg)
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

TABLE XXVIII

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	0.000163
3	5-8	0.0266	3	7-11	0.00061
4	8-11	0.0086	4	11-15	0.000130
5	11-16	0.00116	5	15-20	0.0181
6	16-20	0.00159	6	20-23	0.000119
7	20-21	0.00660	7	23-25	0.000209
			8	25-28	0.00030
			9	28-30	0.00071
Total Pu		89.9			6.77

4. *MHFT-23*. We are testing 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 conditions and humid winter for 6-month periods, and now is in its third summer. The Graphite Impact Shell (GIS) contains *MHFT-23*, an iridium-clad PPO sphere that has not been subjected to an impact test. At the time we placed the FSA in the chamber, we found a small amount of alpha-particle contamination on the outside of the iridium shell, but the outer surface of the GIS was nominally free of contamination. There was a small amount of plutonium contamination in this chamber that remained after cleanup from its use for testing the PISA that contained *MHFT-27*. Very small amounts of plutonium have been found in rainwaters and dehumidifier condensates from time to time, but until recently most of the analyses were indistinguishable from background. Now most of the rainwater samples contain picogram amounts of plutonium, with a few greater than 1 ng. Soil cores 1 and 2 were collected at the beginning of the first summer 25 cm to the left and to the right of the sphere, respectively. In each core there was one section with a very small amount of plutonium (0.1 and 0.03 ng). The plutonium in the other sections was not distinguishable from background. Cores 3 and 4

were collected 25 cm in front of and behind the sphere at the end of the first summer. Core 2A-4 was found to have no significant plutonium content, while the top two sections of core 2A-3 contained only small amounts (0.03 and 0.15 ng) of plutonium. At the end of the second winter, cores 5 and 6 were collected at positions about 25 cm to the front left and back right, respectively, of the sphere. Core 6 contained 0.31 ng of plutonium in the section just below the surface. No measurable plutonium was found in the other sections of this core. Core 5 contained 43 pg in five sections (Table XXIX). Cores 7 and 8 were removed from this chamber at the end of the second summer from positions 25 cm to the front right and back left, respectively, of the plutonium dioxide source. Core 7 was lost in sampling. Core 8 was found to contain over a thousand times as much plutonium as any previous core in this chamber (Table XXIX). This may indicate that the iridium shell is leaking. Soil cores 9 and 10 were collected from positions 12 cm to the left and right, respectively, of the FSA during the third winter.

A Chromel-Alumel thermocouple was attached to the side of the GIS with an epoxy adhesive. During the winter cycle the external temperature varied between 180 and 192°C. After the change to the summer cycle, the temperature increased to 200 to 210°C. During a rain the temperature dropped

TABLE XXIX

PLUTONIUM IN SOIL CORES FROM
CHAMBER CONTAINING FSA MHFT-23

2A-5			2A-8		
Section	Depth	Pu	Section	Depth	Pu
No.	(cm)	(pg)	No.	(cm)	(pg)
1	0-1	8	1	0-3	117 000
2	1-4	7	2	3-6	55 500
3	4-9	15	3	6-8	35
4	9-11	<5	4	8-9	50
5	11-13	6	5	9-10	21 800
6-8	13-18	<5	6	10-12	107 000
9	18-20	7	7	12-14	6
10-14	20-27	<5	8	14-16	7
			9	16-17	6
			10	17-20	12
			11	20-21	5
			12	21-24	8
			13	24-26	670
			14	26-27	<5
Total		43			302 000

below 50°C, and when the temperature and humidity controls were shut down, resulting in air stagnation, the temperature rose to 235°C.

5. Spallation 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's surface, resulting in the spallation 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 four 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 prerain level of 0.1 nCi/m³. 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 prior to the rain. It is possible that the thermal shock releases not only very fine particles of plutonia but also the radioactive gas ²²⁰Rn (a daughter of ²³⁸Pu) entrapped in the plutonium oxide crystal lattice structure. The activity ratio of ²²⁰Rn to ²³⁸Pu 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 prior to 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. 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 2 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 is now growing in this chamber.

III. SORPTION OF PLUTONIUM BY SOILS

(Gilbert B. Nelson and George M. Matlack)

We are continuing the experiments to measure the absorption of plutonium by soils. This is done by allowing dilute plutonium to flow through soil contained in glass columns. The feed solution fed to the

soil columns is a dilute, neutral solution of plutonium chloride, 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.

A preliminary requirement was to determine how the soil column diameter affected the specific flow rate, because a large particle diameter relative to column diameter might introduce random perturbations into the experiment. Accordingly, we determined specific flow rates for columns of three diameters packed with two lots of sand that differed in particle size distribution. We found that in 5-, 10-, and 25-mm-diam columns packed with 38- to 147- μ m sand particles on the one hand and 350- to 600- μ m sand particles on the other, the specific flow rates differed by no more than 10% relative standard deviation when calculated in terms of milliliters per second per square millimeter for the average particle cross-sectional area. These experiments also included 110- and 140-mm waterheads on the columns. We concluded that the specific flow of water through these columns was independent of the column waterhead and diameter, as long as the particle diameter did not exceed 10% of the column diameter and the waterhead did not exceed 50% of the soil height.

In the plutonium absorption experiments, we have five 25-mm-diam columns packed with silt loam that was sieved to remove particles larger than 0.8-mm diam. This soil is about 60% silt, 25% sand, and 15% clay. Its bulk density is 1.07 g/cm³; its particle density, 2.70 g/cm³. It has a total porosity, ϵ , of 60.3, as calculated from the bulk and particle densities using the relation

$$\epsilon = 100(1 - D_b/D_p).$$

Three columns, 100 mm high, retained 99.8% of the plutonium from the feed solutions (1 μ g/l in distilled water) during 556 days of operation before they were switched to plutonium-free, 0.1 M sodium bicarbonate solution. Differences in flow rates from 1.6 to 0.084 μ l/s among the three columns did not affect the retention of plutonium significantly. Since the change to the bicarbonate feed solution (630 days), the flow rate has decreased significantly so that it now ranges from 12 to 200 nL/s. This result was unexpected. The bicarbonate

eluting reagent will be used to measure the rate of plutonium transfer through the columns caused by carbonate and bicarbonate ions which are known to form stable complexes with plutonium. Bicarbonate is also normally found in groundwaters. The slowest column still retains 99.6% of the plutonium, but the retention of the fastest has decreased to 79.8%.

The other three columns are 250 mm high and contain soil sieved to remove particles smaller than 52 μm in order to achieve more uniform flow rates. They have been running for 1028 days, except for one which was lost at day 517 because of failure of the glass wool plug that retained the soil in the column. The average flow rate through this column prior to failure was 1.4 $\mu\text{l/s}$, and it retained 99.94% of the plutonium. The flow rate in the two remaining columns has averaged 2.2 and 1.6 $\mu\text{l/s}$, and they have retained 99.79% and 99.93% of the plutonium, respectively.

Three additional 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. The columns are being

eluted with distilled water (a rainwater simulant). The columns have been in operation for 313 days. Measurements made after 211 and 313 days of operation (Table XXX) show that the flow rates have not changed significantly, but that the quantity of plutonium released has. 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 still remain at the top of each column; radiation cannot be detected from any particles that may have moved downward into the soil columns.

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

(George M. Matlack, Gilbert B. Nelson, 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 liters 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.

TABLE XXX
Pu TRANSPORT FROM PuO_2 PARTICLES

Column	Elution Duration			
	123 days		313 days ^a	
	Total Vol.(l)	Total Pu(ng)	Total Vol.(l)	Total Pu(ng)
7	12.5	16.2	20.2	12.6
8	10.4	15.9	17.4	16.3
9	4.5	31.7	7.4	34.6

^aThe 313-day period includes the 211-day period; both periods started on November 4, 1976.

A. Seawater Aquarium Experiments

Each aquarium contains 75 liters 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.

C. PMC Dissolution or Release Rates

These experiments have been terminated. A topical report has been issued as an LA document.¹⁰

D. PPO Dissolution or Release Rates

These tests are to compare release rates from 2.5- and 25-W PPO pellets in both fresh water and seawater. Because of the remote chance that a heat source may drop into a warm tidal pool, seawater tests at 37°C are included. Five experiments are under way, two with 2.5-W pellets and three with 25-W pellets.

1. *In Seawater.* The plutonium release rate from the 2.5-W pellet HPZ-3 (Table XXXI) has decreased to 4.7 nCi, following its earlier drop from a maximum of 69 nCi/m²-s observed 30 months ago. During the first 400 days of immersion, its average release was fairly linear at 65 nCi/m²-s. Since then, the rate has decreased slowly. Despite the lengthy immersion, the pellet has not deteriorated visibly, pointing to the superior stability of PPO compared to PMC.

TABLE XXXI
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	1273	Fresh	10	180
HPZ-3	2.5	1843	Sea	10	4.7
HPZ-60-3	2.5	1032	Sea	37 ^a	3.0
HPZ-111-1	25	981	Fresh	10	390
HPZ-59-4	25	1066	Sea	10	12
HPZ-59-2	25	1066	Sea	29 ^b	1.5

^aTerminated January 24, 1977.

^bTemperature was 37°C until November 16, 1976, when the heater broke. Repair of heater would stop the experiment.

The plutonium release rate from the 2.5-W pellet HPZ-60-3, immersed in 37°C water typical of tropical near-shore shallows, averaged 3.0 nCi/m²-s for 1032 days. (This experiment was terminated January 24, because the aquarium developed an irreparable leak). This is the second lowest rate from any ²³⁹Pu fuel tested thus far. A possible explanation may be accelerated plutonium deposition on underwater surfaces as a result of faster chemical reactions at this warm temperature. Because release rates are based on plutonium found dissolved or suspended in the water, rapid deposition can reduce the apparent release rate. Another explanation is that the warm water causes rapid precipitation of gelatinous plutonium hydroxide on the pellet surface, forming a protective coating that retards further release of plutonium.

The two 25-W PPO pellets (Table XXXI) are HPZ-59-2 at 29°C and HPZ-59-4 at 10°C. After 1066 days, the release rate is 12 nCi/m²-s for the pellet at 10°C, down 8% from the previous quarter's rate. This is the same order of magnitude as the 4.7 nCi/m²-s release rate of the 2.5-W pellet HPZ-3 at 10°C. On the other hand, the amount of plutonium in the water containing the 25-W pellet at 29°C is very low, only 19 µg. This corresponds to a release rate of 1.5 nCi/m²-s, the lowest observed for any heat source in the program, and the same order of magnitude as the 3.0 nCi/m²-s rate observed for the 2.5-W pellet in warm seawater described above. There seems little doubt at the present time that plutonium is released much more slowly into warm seawater than it is into cold seawater.

2. In Fresh Water. Plutonium release from the 2.5-W pellet HPZ-60-2 to fresh water at 10°C has averaged 180 nCi/m²-s for 1273 days (Table XXXI). This is approximately 20 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 390 nCi/m²-s for 981 days, which is the highest value of the three 25-W pellets. It is clear (see Table XXXI) that the highest release rates are found in fresh water and the lowest in 37°C seawater. There is no significant difference that can be related to the difference in power level between the 25-W and the 2.5-W pellets.

3. In Ocean Water. Two PPO pellets, designated HPZ-19, were placed 23 m deep in the ocean off San Clemente Island in March 1973, for long-term exposure. The two pellets were right circular cylinders, 12.5 mm in diameter by 12.5 mm high, each containing 14.7 g of PuO₂. They were placed on sea bottom debris inside protective chambers that permit water circulation but keep out animals larger than 1 cm. The pellets were visually inspected at 3-month intervals, then reimplanted. After 1 yr, they began to be encrusted with fine sand-like grains. Where the pellet edges were still visible, no deterioration could be seen, in contrast to the crumbling of PMC pellets exposed in the ocean.

These pellets were removed from the ocean and shipped to LASL in December 1975. Electron microprobe examination of the crusts from the pellets showed the presence of three major phases, calcium and oxygen, iron and oxygen, and aluminum and oxygen, mixed together in a heterogeneous fashion. Significant amounts of silicon were also present. These findings are consistent with the elements present in the sea bottom debris in which the pellets were buried for 2 yr.

4. Total Release Rate (Source Term). The plutonium release rates in Table XXXI 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 ²³⁹PuO₂ source, YMZ-8, 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 pin-hole leak in the cladding. The water was sampled in duplicate periodically for plutonium analyses.

The analytical data confirm that the source is leaking; and demonstrate that small plutonium oxide particles are being released. The release rate is also erratic. Variations in the plutonium concentration in duplicate samples indicate that the source is 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 pin-hole 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.

A source term will be calculated when the experiment is terminated, from measurements of the total plutonium in the water and on the interior chamber surfaces.

5. Hydrostatic Test Facility. The GIS assembly, MHFT-259, was placed in the Hydrostatic Test Facility (HTF) in 5 liters of simulated seawater on February 11, 1977, and brought to a pressure of 63 MPa (9200 psi). Daily plutonium analyses showed small amounts of plutonium in the seawater (~ 10 ng), but there was no significant trend. This small amount of plutonium could have come from contamination on the inside of the GIS. No iridium was detected in the seawater.

Pressure was reduced gradually over a period of 100 minutes to ambient at the end of 31 days. Within the next 2 days the plutonium concentration in the water increased to nearly 700 ng/l, then gradually decreased (Table XXXII). We believe that following depressurization, water that had been forced into the space between the fuel and the cladding was gradually expelled through the helium vents into the water in the pressure vessel. The subsequent decrease in concentration probably resulted from deposition of the plutonium on the Monel metal surfaces inside the vessel. These surfaces were decontaminated to recover this deposited plutonium. The plutonium recovered corresponded to a release rate of 10 nCi/m²-s for the 31-day period. No visible changes were seen on the GIS, which is presently being disassembled for closer examination.

The fuel sphere was placed in a new GIS, and this assembly was placed in the HTF and pressurized to 70 MPa on May 27, 1977. This commences an experiment planned to last approximately one year.

E. Clad Sphere in Simulated Seawater

The plutonium release from a 100-W PISA, MHFT-13, was measured during immersion 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

TABLE XXXII
PLUTONIUM RELEASE IN HYDROSTATIC TEST FACILITY

Duration = 31 days

Pressure = 65 MPa

Simulated Ocean Depth = 6 km

Day	Pu Conc., ng/l	Day	Pu Conc., ng/l
0	~ 0.05	31-1	0.6
1	0.1	31-2	10
6	2.0	32	260
13	1.4	33	670
17	0.4	34	310
21	0.3	35	76
28	0.5	38	15

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 irradiation caused chemical attack and disintegration of the iridium cladding from within.

V. PLUTONIUM RELEASE AS A FUNCTION OF PARTICLE SIZE

(Gilbert B. Nelson and George M. Matlack)

Three sets of carefully sized $^{238}\text{PuO}_2$ particles have been prepared, and weighed amounts of these particles have been placed in 50-ml volumetric flasks containing 1 M HClO_4 that are agitated in a 37°C water bath. 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.

The data (Table XXXIII) have been collected for 677 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 44-61 μm . Distributions B and C represent con-

TABLE XXXIII
SUMMARY OF DISSOLUTION EXPERIMENTS IN 1M HClO_4

Experiment Number	PuO_2 , mg	Dissolution Rate	
		g/s	g/s-g
A1	2.02	7.0×10^{-13}	3.5×10^{-10}
A2	2.06	6.5×10^{-13}	3.2×10^{-10}
A3	10.19	3.5×10^{-12}	3.4×10^{-10}
A4	9.90	3.4×10^{-12}	3.4×10^{-10}
B1	2.02	1.5×10^{-12}	7.4×10^{-10}
B2	2.03	1.8×10^{-12}	8.9×10^{-10}
B3	10.25	7.9×10^{-12}	7.7×10^{-10}
B4	10.45	8.3×10^{-12}	7.9×10^{-10}
C1	2.23	4.6×10^{-12}	2.1×10^{-9}
C2	2.31	4.3×10^{-12}	1.9×10^{-9}
C3	9.89	1.7×10^{-11}	1.7×10^{-9}
C4	10.67	1.8×10^{-11}	1.7×10^{-9}

tinually smaller sizes. Data show that the dissolution rate, grams dissolved per second, changes in three different ways. The larger particles, A, show a slightly increasing rate with time; the intermediate particles, B, show a constant or slightly decreasing rate, while the smallest particles, C, show a decreasing rate. The dissolution rate per gram of oxide (g/s-g) is fairly uniform within each of the three sets of particle sizes. However, within a given particle size, the rate (g/s) is directly proportional to the mass of the particles.

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