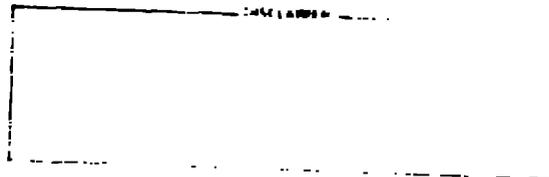


**TITLE:** EXPERIMENTS INVOLVING <sup>238</sup>PLUTONIUM DIOXIDE AND THE ENVIRONMENT

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EXPERIMENTS INVOLVING  $^{238}\text{PuO}_2$  AND THE ENVIRONMENT

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ABSTRACT

The interactions of  $^{238}\text{PuO}_2$  with terrestrial and aquatic environments have been examined. Samples were removed periodically and analyzed for plutonium to determine dispersion. The samples from terrestrial experiments were acidified, reduced in volume, and analyzed by liquid scintillation counting while aquatic experiment samples were diluted and counted by liquid scintillation. Results from the aquatic experiments indicated that the release rate was independent of the thermal power of the source. More plutonium was released in cold fresh water systems than in warm or cold seawater. Plutonium migration through soils appeared to be independent of soil type. Seasonal variations in plutonium migration appeared to result from the different quantity of water passing through the soil. Large pieces of source material released more plutonium to the environment which may be a thermal spallation effect. A simulated tidal experiment in which the source cycled into and out of the seawater twice a day resulted in severe fracturing of the source.

INTRODUCTION

Plutonium enriched 80% in  $^{238}\text{Pu}$ , an alpha emitter with a half-life of 87.7 years, is used in the dioxide form as the heat source in radioisotope thermoelectric generators that provide power for instruments and data transmission in many space missions. Because of the relatively short half-life and the high alpha energy the thermal specific power is 0.56 W/g of  $^{238}\text{Pu}$ . Although the heat source container is designed with a large safety factor to withstand

both reentry from orbit and impact with the earth, additional information is continuously sought concerning the interaction of the heat source with the environment.

Since there are two general environmental conditions into which a heat source may be deposited on return from orbit, namely, on land or in water, several variations of each condition have been utilized in experiments at Los Alamos Scientific Laboratory. The discussion of these variations will include the conditions, the samples obtained, the radiochemical processing, and results.

#### EXPERIMENTAL

Terrestrial experiments are conducted in sealed environmental chambers. These chambers, with an internal volume of 1.8 m<sup>3</sup>, contain a soil tray that is 0.3 meters deep by 0.9 by 0.9 meters in area. The chambers are equipped with environment control equipment including heaters and refrigeration for temperature control, and humidifier and dehumidifier coils for humidity control. Each chamber is equipped with a spray head that can provide a relatively uniform rain over the surface of the soil tray.

The samples obtained from a terrestrial experiment include rainwater that has percolated down through the soil, condensate from the dehumidifier coils, and soil core samples. Rainwater and condensate samples are collected in 20 liter polyethylene bottles. Core samples are obtained by driving a hollow tube down through the soil to the bottom of the tray. A core is approximately 27 cm long by 13 mm in diameter.

Two types of soils and two source configurations have been used in these experiments. A sandy soil containing approximately 90% sand and a loam soil containing 50% sand and 38% silt are the types used. Each soil is used in conjunction with two configurations from a particular source, large pieces and fines (from 10  $\mu$ m to 6000  $\mu$ m in diameter). This material is obtained from heat sources that have undergone impact testing as part of the overall source containment safety evaluation program.

Aquatic experiments are conducted in aquariums that have been outfitted with sealed lids and filter systems. These aquariums have heaters and refrigeration systems for temperature control. Fresh water has been utilized in experiments at 10°C and 37°C. Sources used in aquatic

experiments were usually cylindrical in shape and weighed about 6 g for low source power experiments to 60 g for high source power experiments. Samples obtained from these experiments were aliquots of the aqueous phase.

A variation of the aquatic experiments involved placing the source on a tray equipped to cycle into and out of the water twice a day. This experiment, conducted in simulated seawater represents a source lying in a tidal basin. In addition to aliquots of the aqueous phase, samples of sand from the tray on the bottom of the aquarium and a piece of the source were removed for analyses.

Other experiments being conducted include aquatic conditions in glass chambers, a high pressure seawater experiment, particle size solubility studies in 1M HClO<sub>4</sub>, ionic and particulate plutonium on soil columns, and tests to determine ionic versus particulate form.

Radiochemical processing and analyses of the samples are rather straightforward. Rainwater samples are quantitatively transferred to beakers, acidified with HNO<sub>3</sub> and a trace of HF, and reduced in volume by heating. The resulting small sample is quantitatively transferred to a volumetric flask and diluted to volume with 0.5M HNO<sub>3</sub>. An aliquot is then counted by liquid scintillation using a commercial instrument. A commercial cocktail, to which 0.1% D-2-EEPA is added, is used in a routine manner. A condensate sample is treated in the same manner except that only a 4 liter aliquot of the total 20 liter sample is processed. Soil cores are cut into segments, dried, weighed, and dissolved using HNO<sub>3</sub>-HF followed by HNO<sub>3</sub> - HClO<sub>4</sub>. The final solution is treated in the same manner as a rainwater or condensate. Aliquots of aquarium waters are pipetted directly into the cocktail and counted.

## RESULTS

### Terrestrial Experiments

Two types of soils, sand and loam, were used in the terrestrial studies. Large pieces and fines of the same source were placed on each type of soil. Experiments using loam were conducted on a humid winter-summer cycle, whereas experiments using sand were on an arid winter-summer cycle. These experiments are still in progress but some preliminary results are available at this time.

There appears to be a significant release of plutonium from the large pieces of source material upon contact with

rain. These releases are attributed to thermal spallation because the surface temperature of a large piece is higher than that of fines. Several air filter tests have been conducted covering the time interval before, during, and after a rain. In a chamber containing large pieces the airborne plutonium concentration increased by four orders of magnitude during the first 5 minutes of the rain and then decreased until it reached prerain levels about 1 to 2 h after the rain. In a chamber containing fines there was no significant increase in airborne plutonium when the rain commenced, but there was a reduction of a factor of 10 after the rain, which attributed to a cleansing effect of the rain.

In all cases plutonium was seen in the percolated rainwater collected during and after the first rain subsequent to source implanation. This result suggests that plutonium migrates in the particulate form and the migration rate is independent of soil type.

In a majority of the core samples most of the plutonium was present in the top layer of the soil (0-5 cm). The other layers contained measurable amounts of plutonium but usually significantly less than the top layer. There were a number of cores that contained a substantial amount of plutonium at some intermediate depth, but no consistent trend could be discerned.

Three soil columns, containing silt loam (840  $\mu\text{m}$  to 1650  $\mu\text{m}$  in diameter) with approximately 3 mg of  $^{238}\text{Pu}^{1/2}$  on the surface of each soil, have been in operation for about 1000 days. These columns are being eluated with distilled water. There is a continuous low level release of plutonium, 20 pg/l, but almost 90% of the released plutonium was collected during the first 5 days of the experiment. These results again suggest that plutonium migrates in the particulate form.

#### Aquatic Experiments

Table I is a summary of the aquatic environment experiments. Several observations can be made concerning the data. First, the lowest release rate is seen in warm seawater, a condition that may occur in an isolated tidal pool or in the tropics. The highest release rates are observed in the cold fresh water systems. Third, at this time no significance is attached to the variation in release rates resulting from the different thermal power of the sources. The substantial difference in the release rates between fresh water and seawater experiments indicates that removal reactions are occurring in the seawater

# TABLE I

## SUMMARY OF RELEASE RATES FROM PPO IN AQUATIC ENVIRONMENTS

WATER	POWER (W)	TIME (days)	TEMP. (°C)	RELEASE RATE (nCi/m <sup>2</sup> -s)
FRESH	2.5	1633	10	171
FRESH	25	1610	10	460
SEA	2.5	1660	10	7.7
SEA	25	1615	10	12
SEA	2.5	993	37	2.7
SEA	25	968	37	2.3

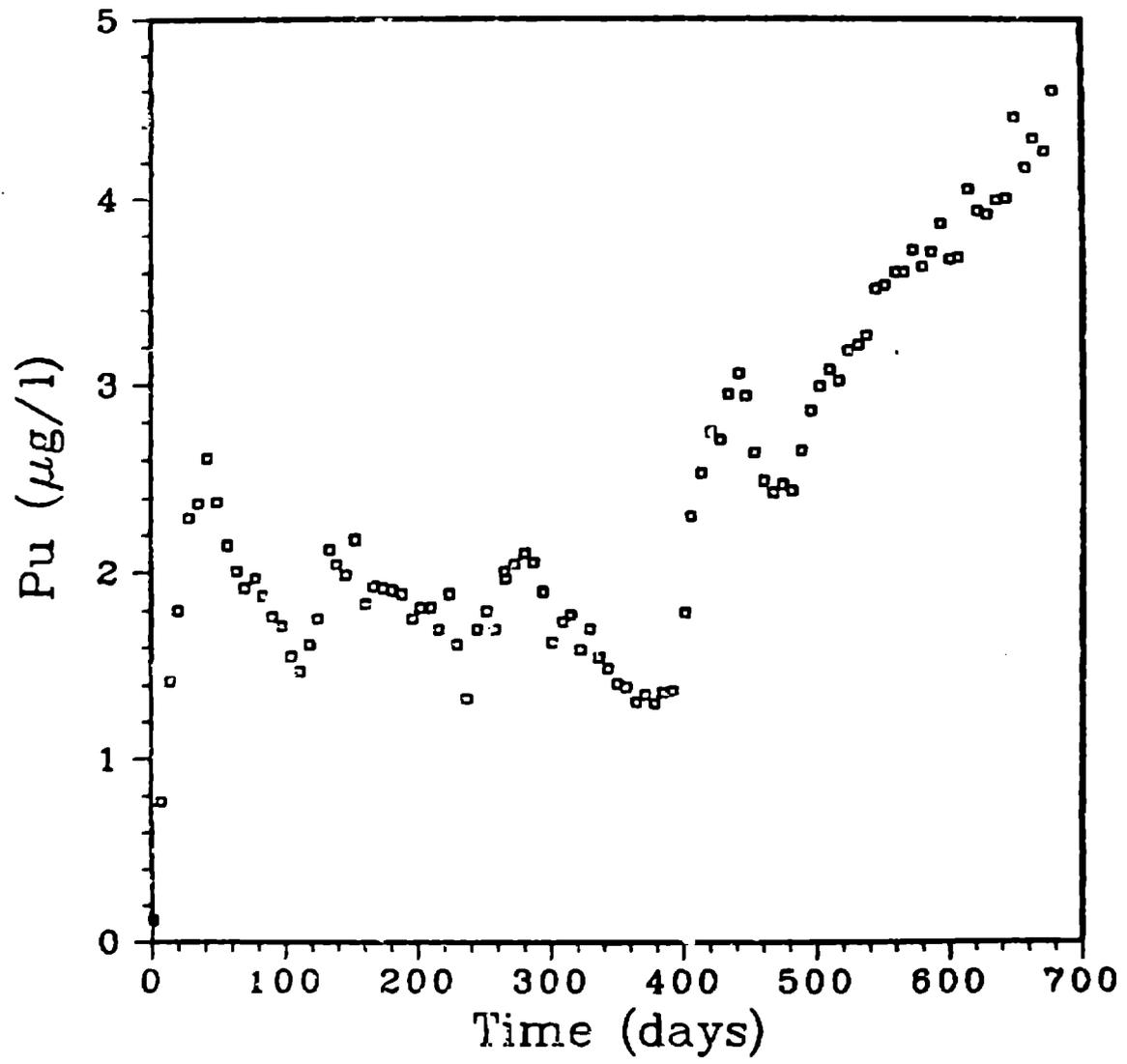
experiments that do not occur in fresh water, or are occurring at a faster rate, or that the release rate is substantially reduced in seawater, or a combination of all three.

A variation of the aquatic experiment was the tidal basin. In this aquarium the source was placed on a tray that was cycled into and out of the water twice a day. Figure 1 is a graph of the plutonium concentration in the water. Note the initial increase of plutonium to a peak at about day 40. After that time there appears to be a gradual decrease. On day 103 a crack was noted in the face of the source and on day 393 the source had split into two pieces. By day 404 there were 7 pieces, and at the present time there are over 30 pieces. The sharp increase in plutonium concentration starting about day 400 is attributed to the exposure of new surface. Again a peak followed by a decrease is seen. On day 451 the elevator motor failed and the source was left submerged. The cause of the increasing plutonium level to the present time is not known.

Because of the possibility of plutonium removal by plating out on the walls or settling to the floor of the aquarium, a glass chamber was designed which would permit total recovery of plutonium after termination of the experiment. An indication of the magnitude of this problem may be seen in the tidal experiment. Sand samples from the tray on the bottom of the aquarium were removed and analyzed for plutonium. The results, averaged over the area of the tray, imply that there are 28 mg of plutonium on the bottom of the aquarium. A total of 0.35 mg is estimated to be in the aqueous phase.

An initial experiment in the glass chamber designed to measure a magnitude of the plate-out error, revealed that a determination of plutonium released based on a measurement of the plutonium level in the aqueous phase would be low by 38%. This error is not as large as expected on the basis of the tidal experiment, but the source configurations differ significantly.

Additional experiments to determine a "total source" term have been conducted in the glass chambers. In one case a bare source was sealed in a dry chamber for 454 days. The total plutonium recovered from the chamber, excluding the source itself, was 1.41  $\mu\text{g}$ . This source was then submerged in deionized water for 30 min. Again the chamber was processed to determine the total plutonium released. In this case 49  $\mu\text{g}$  were recovered. The first



experiment indicated that a small amount of plutonium is released from the surface of a dry source by recoil. The second experiment was in agreement with a proposal by R. L. Fleischer. This proposal suggests that the application of a solvent to the surface will remove a significant quantity of plutonium that has been loosened by alpha decay process near the surface.<sup>1</sup> The next experiment in the sequence would be to allow the source to reach equilibrium in air and then resubmerge it. This test would permit a determination of the contribution of thermal spallation to the total release. Before this test could be accomplished the source split into three pieces. Since there was no longer a good estimate of surface area the experiment was terminated.

#### SUMMARY

Many of the experiments described above and others not described are still in progress at this time. The tentative conclusions mentioned are subject to change as more data become available. Additional experiments in progress are designed to determine the state of the plutonium in the aqueous phase, ionic or particulate. Additional terrestrial experiments are in the planning stages as are experiments in the glass chambers.

#### REFERENCES

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