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*Behavior of Plutonium Oxide Particulates
in a Simulated Florida Environment*

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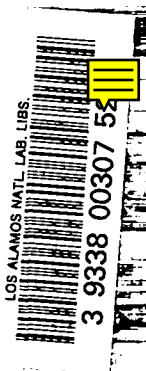
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BEHAVIOR OF PLUTONIUM OXIDE PARTICULATES IN A SIMULATED FLORIDA ENVIRONMENT

by

R. C. Heaton, J. H. Patterson, and K. P. Coffelt

ABSTRACT

The behavior of ^{238}Pu oxide particles (20 to 74 μm in diameter) deposited on a soil surface was studied by using an environmental test chamber. The soil was obtained from Florida orange groves, and the chamber was set up to simulate a Florida climate. After more than 9 months and more than 60 simulated rainfalls, the plutonium oxide particles remained on top of the soil and showed no evidence of having moved down into the soil column. Plutonium was released into the soil drainages at the rate of 18 $\text{ng}/\text{m}^2/\text{L}$. This release, which represents a minute portion of the source, appears to correlate with the volume of the drainage rather than with time and probably consists of plutonium attached to very fine soil particles. The average concentration of plutonium observed in the air was 7 fCi/L , which, on an absolute basis, represents $8 \times 10^{-12}\%$ of the source material. Thus the generation of airborne plutonium constitutes an insignificant release pathway in terms of the original source. However, the air concentration during, and especially at the beginning of, a rainfall was typically much higher (1400 fCi/L). This concentration decayed rapidly after the end of the rainfall. These results are compared with those from past experiments, and their implications are discussed.

INTRODUCTION

Radioisotope heat sources containing ^{238}Pu oxide are used in many space missions to provide power for instrument operation and data transmission. The heat source container is designed to withstand reentry from orbit and launch pad explosions so the source will be safely contained until it is recovered. Although existing designs have proven to be more than adequately safe, additional information about the heat sources and the plutonium contained therein is continually sought so that designs can be improved and reliable risk assessment analyses can be performed.

One concern that needs to be evaluated for the risk analysis is the possibility that a launch accident might

disperse PuO_2 heat sources into the area surrounding the Kennedy Space Center. To evaluate this concern, qualitative information on the behavior of plutonium and plutonium oxide particulates in a Florida-type agricultural ecosystem is required. In addition, some quantitative information is required. Particularly important is the vertical distribution of the plutonium in the soil column as a function of time and rainfall.

The primary objectives of this experiment are twofold: to determine the vertical distribution of plutonium in the soil column, as noted above, and to evaluate plutonium release pathways into the environment. A secondary objective is to identify parameters that may influence the mobilization and transport of plutonium.

EXPERIMENTAL WORK

Equipment

This experiment was conducted with an environmental test chamber manufactured by Standard Environmental Systems, Inc., of Totowa, New Jersey, model SLHH/64 (see Fig. 1). The chamber allows automatic control of daily temperature, humidity, and lighting cycles. A spray head was added so rainfalls could be conducted as needed. Use of this type of chamber permits the simulation of a wide variety of climatic conditions in the laboratory, while providing for the containment of radioactive materials.

Several substantial modifications to this chamber were required. The main door was modified to include glove ports, a bagout port, and two small auxiliary ports. In addition, a carriage system was placed inside the chamber so a cutter could be manipulated through the glove ports. These modifications were required so that vegetation within the chamber could be cut regularly without having to open the chamber and risk release of radioactive materials into the laboratory.

The soil tray used in this experiment was 91 cm (36 in.) square by 30 cm (12 in.) deep. The drain from this tray was routed to a collection vessel outside the chamber, so the drainages could be collected easily and analyzed periodically. We collected 25-kg (approximately 55-lb) samples of soil at various locations in orange groves adjacent to the Kennedy Space Center. A representative sample of this soil, obtained by using a series of sample splitters of decreasing sizes on half of the total sample, was sent to the National Soil Survey Laboratory in Lincoln, Nebraska, for characterization (see Table A-I in the Appendix).

Climate

Temperature and humidity ranges were determined by using meteorological data for Daytona Beach, Florida.¹ These data are shown in Figs. 2 and 3. We decided that using four "seasons" allowed a reasonably concise simulation of the climatic data. We shall subsequently refer to these seasons as winter, spring, summer, and fall, with winter being the coldest and summer the hottest. The temperature ranges for spring and fall were identical. However, the humidity ranges were quite different because Florida has a rainy season that occurs during the summer and early fall. Figure 4 shows the precipitation data from Ref. 1. We simulated these data by using two seasons, a rainy season running from the beginning of June to the end of October and a dry season covering the rest of the year. The dry season was simulated by a weekly rain of 0.610 in./rain; the wet

season was simulated by a twice weekly rain of 0.750 in./rain.

Diurnal temperature cycles were calculated for each season by scaling data from Fig. 38 in Ref. 2 to match the temperature range and average for the appropriate season in Fig. 2. In the case of an actual deposition of the source onto the ground, the source would be situated on or just under the ground surface. The temperature of the air just above the ground is controlled by the ground temperature, which, in turn, is a strong function of the insolation. In our environmental chambers, the insolation is not a significant factor in determining the soil temperature. Rather, the soil temperature is controlled entirely by the air temperature, which, in turn, is determined by the climate control machinery of the chamber. Thus, to simulate the environment that actually relates to the source at ground level, we must artificially adjust the temperature of the chamber to represent that at ground level, rather than that recorded by meteorologists (typically 2 m above ground). Accordingly, we extrapolated the scaled temperature data to 5 cm above ground level (as low as the data allow) to determine the hourly temperatures to use for the chamber. Results of these calculations are shown in Tables A-II through A-V.

Diurnal humidity curves were constructed by averaging the hourly data points from Ref. 1 over each season and plotting these on graphs (four points for each season) with relative humidity on the vertical axis and time of day on the horizontal axis. The other curves were filled in freehand, noting that the humidity should change rapidly with temperature and that the humidity should remain constant when the temperature is constant. The relative humidities determined and the corresponding wet bulb depressions are shown in Tables A-II through A-V.

At the latitude of central Florida, 29°N, daytime is approximately 2 h longer in the summer than in spring and fall and about 2 h shorter during the winter. Thus the lights were operated 14 h per day during the summer and 10 h per day during the winter. During the spring and fall seasons the lights were operated 12 h per day.

Source

Plutonium oxide fines from an impact-tested heat source fuel pellet (HF-160) were collected for use in this experiment. The plutonium was nominally 83% ²³⁸Pu; isotopic composition is shown in Table A-VI. The fines were wet sieved, and the material able to pass through a 74- μ m (200-mesh) screen but not through a 20- μ m electroformed sieve was used. The surface area of the source particles was calculated by assuming spherical particles with densities of 10.0 g/cm³ and a constant mass-to-diameter distribution between 20 and 74 μ m.

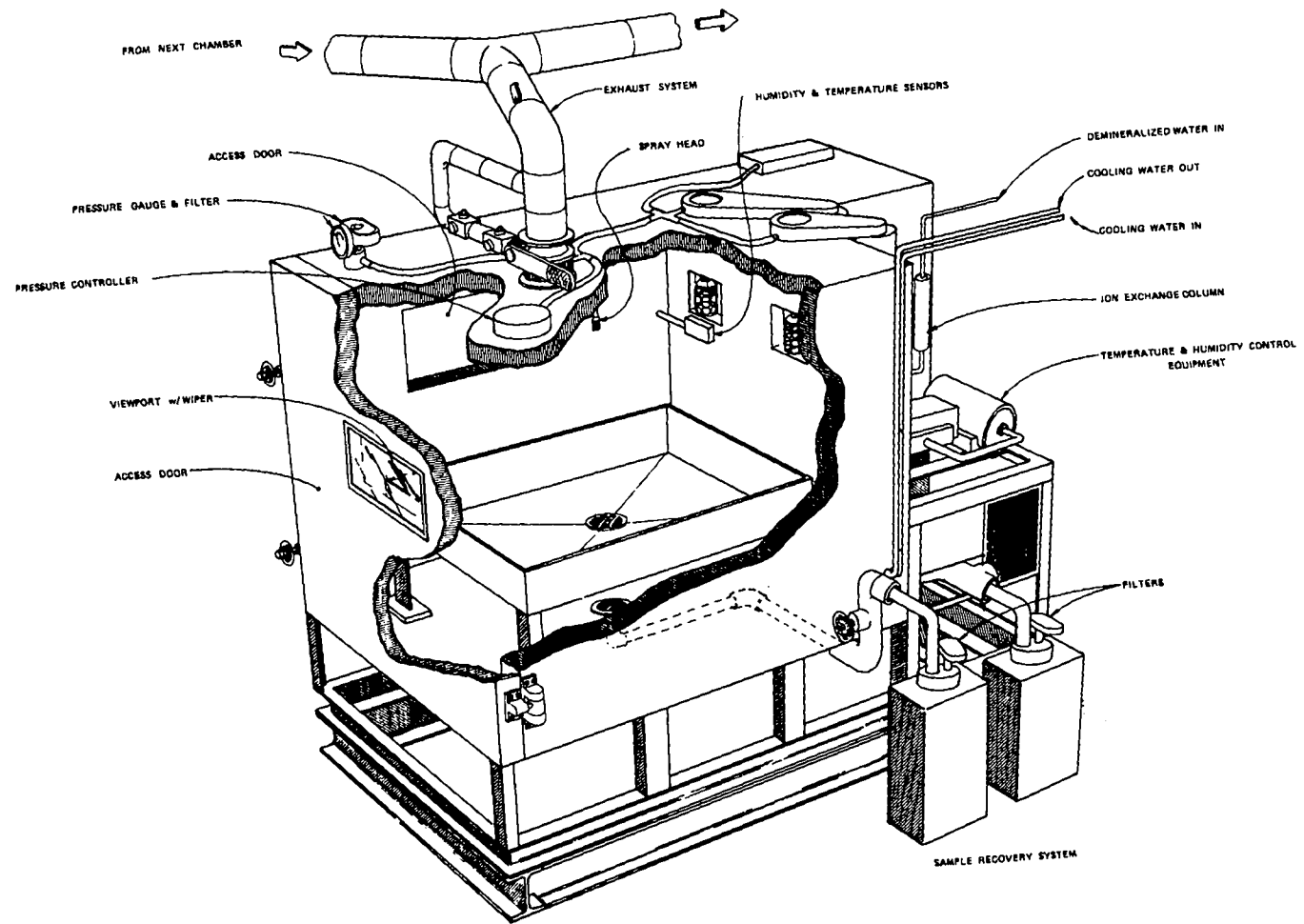


Fig. 1. Environmental simulation chamber (unmodified).

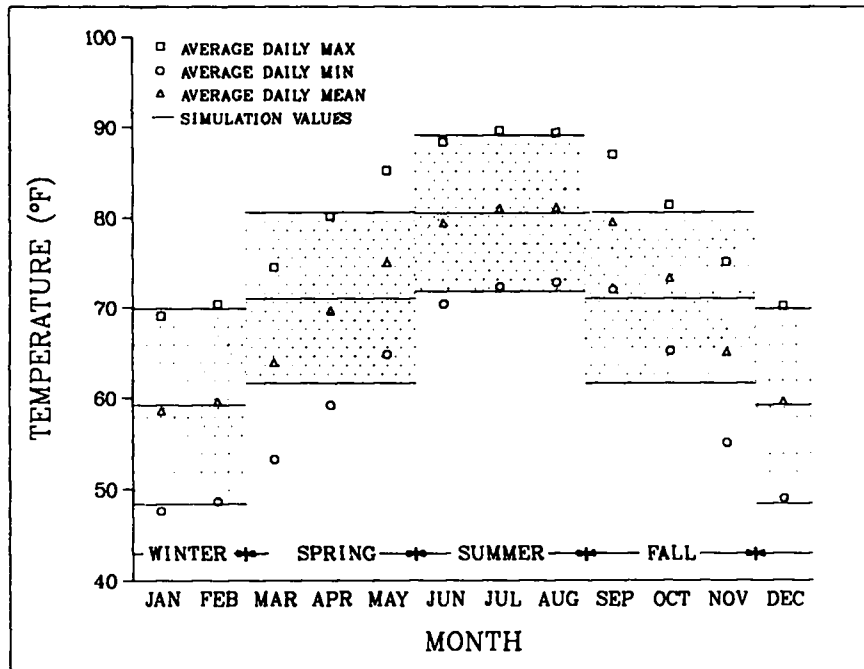


Fig. 2. Seasonal temperature ranges for Daytona Beach, Florida, and corresponding simulation chamber values.

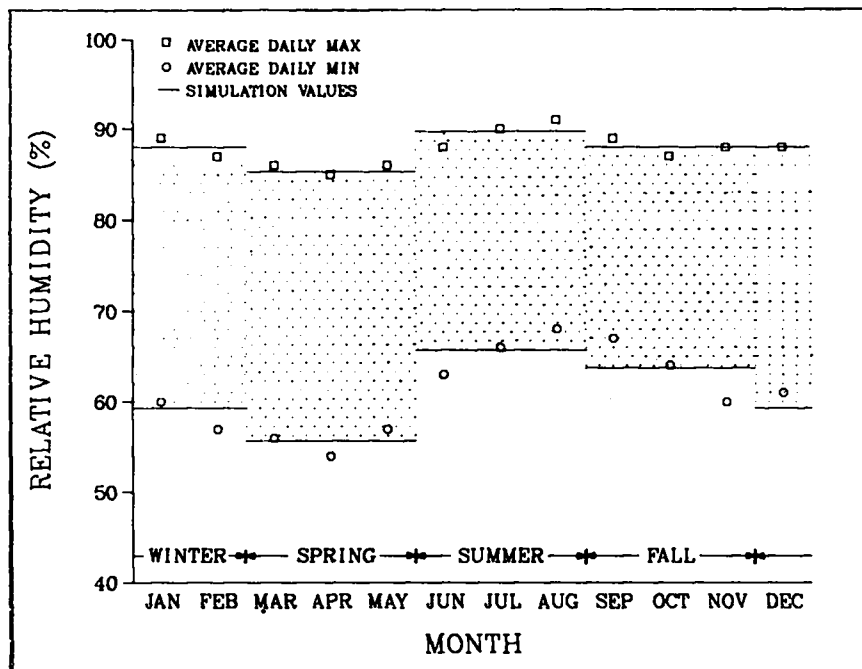


Fig. 3. Seasonal humidity ranges for Daytona Beach, Florida, and corresponding simulation chamber values.

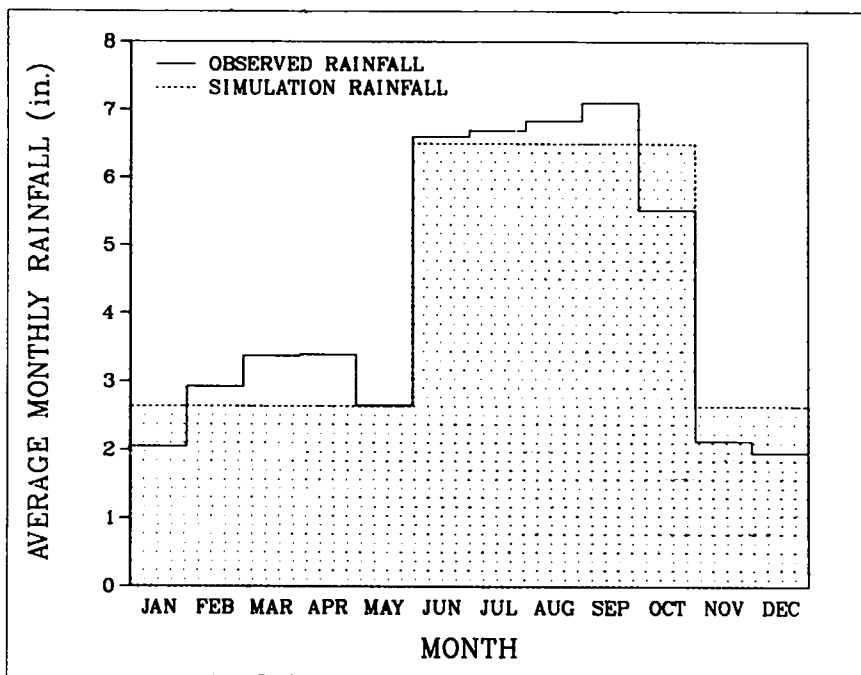


Fig. 4. Rainfall data for Daytona Beach, Florida, and corresponding simulation chamber values.

These particles, weighing 17.53 g (surface area of 0.255 m²), were mixed with 161.2 g of sea sand that had been ground to -325 mesh. This mixture was distributed with a flour sifter over the front half of the soil tray. In this way the plutonium oxide fines were somewhat uniformly scattered over the surface of the soil. We distributed the fines in the front of the chamber because that was the only part accessible through the glove ports. The source deposition was done immediately before the first rainfall cycle of the experiment. The source was deposited in the middle of the spring season of the chamber climate cycle.

The amount of plutonium deposited on the soil was quite large in order to facilitate detection of the plutonium by gamma counting. The most obvious effect of the plutonium was to kill all the plants in the front half of the chamber where the plutonium was deposited; the plants in the back half were apparently unaffected.

Samples

Percolates. The drainage from the soil tray was collected after each rain. Measurements of pH and specific conductance were made on each percolate collected.

Plutonium contents of the samples were determined by liquid scintillation counting.

Condensates. Dehumidifier condensates were collected after each rain, and the pH was measured in every case. The plutonium contents of the samples were determined by liquid scintillation counting.

Air Samples. Air samples were collected by drawing known volumes of air through filters and determining the plutonium collected on the filters. A number of sample series were obtained during the experiment, one very shortly after introduction of the source and several later in the experiment after 50 rain cycles. The 0.4- μ m Nuclepore filters were dissolved with commercially available tissue solubilizer, and their plutonium contents were determined by liquid scintillation counting.

Core Samples. Core samples were obtained by pushing short lengths of Plexiglas tubing (3/4-in. o.d. with a 1/16-in. wall) into the soil to the full depth of the soil tray and then withdrawing the tube and the soil. The remaining hole was filled with molten paraffin wax to prevent the passage of rainwater through the hole to the bottom of the soil tray. Plutonium distributions within

the cores were determined by gamma counting. A sodium iodide detector was placed underneath a 1/8-in.-thick lead sheet with a 1/8-in. slot cut in it. The core was placed on top of the sheet, and the count rate was recorded as a function of the position of the core relative to the slot. In this way the gamma activities at various depths within the soil were readily determined. Three of the cores were cut into six sections each. These sections were dissolved in acids and their plutonium contents were determined by liquid scintillation counting.

Depth Profiles. In addition to the core samples, another attempt to determine the plutonium depth profiles was made by measuring gamma activities *in situ*. Three plastic wells were placed in the soil before introducing the PuO₂ source. These were made of Plexiglas tubing, 1-1/4-in. o.d. with 1/16-in. walls, and designed to accept insertion of a sodium iodide detector (Geoline model .75MT1/.75L, equipped with an aluminum can instead of the usual stainless steel). Several layers of lead tape were wrapped around the barrel of the detector so only gamma rays coming from beneath the detector would be seen. Count rates as functions of depth were then obtained by lowering the detector to various depths in the sampling wells and measuring the count rates.

RESULTS AND DISCUSSION

Depth Profiles

Plutonium distribution in the soil column is best revealed by the soil cores. The gamma activities over the lengths of the cores are shown in Figs. 5 to 14. The resolution of the detector used was plus or minus 2.1 mm, which was determined by obtaining count rates on a thin americium source as a function of position relative to the entrance slit of the detector. Two conclusions

are readily apparent. Virtually all the gamma activity resides within the top centimeter of soil and the distribution remains unchanged over the course of the experiment. If one makes the reasonable assumption that the plutonium is located in the same place as the gamma activity, then plutonium particles were not observed to move down into the soil with rainfall in this experiment.

One of the cores was cut into six equal sections and the sections were dissolved and analyzed for plutonium. Results of these determinations (core number 6) are shown in Table I. More than 99% of the plutonium was found in the topmost section of the core, in agreement with the gamma counting results. However, the plutonium contents of the lower sections of the core were not zero, but ranged in the tens to hundreds of microcuries. With these data alone, it was not clear whether this activity was in these lower sections of the soil before the core was taken or if the lower parts of the core were contaminated during the act of pushing the coring tube into the soil. To resolve this issue, two cores were taken during the final coring operation; one was taken the normal way. However, before the other was taken, the top half-inch of soil was removed. Thus the coring tube was not pushed through the top, highly contaminated layer of soil before reaching the lower sections. These two cores were cut into six sections each and analyzed together. The results are shown in Table I (cores 10 and 10A). Core 10 is a virtual repeat of core 6, confirming those results. However, the plutonium concentrations in all sections of core 10A are drastically lower than either of the other two cores. Clearly the plutonium found in the lower sections of the earlier cores was deposited there during the coring operation. We may conclude that virtually all the plutonium resided in the top 1 to 2 cm (core sections were nominally 2 cm in length) of soil and that the PuO₂ did not migrate down into the soil as a result of simulated rainfall.

TABLE I. Soil Core Analyses

| Core 6 | | Core 10 | | Core 10A | |
|----------|-------------------|----------|-------------------|----------|-------------------|
| Section | Pu (μ Ci) | Section | Pu (μ Ci) | Section | Pu (μ Ci) |
| 1 Top | 85 400 | 1 Top | 73 300 | 1 Top | 1.37 |
| 2 | 561 | 2 | 110 | 2 | 0.295 |
| 3 | 168 | 3 | 53.2 | 3 | 0.0389 |
| 4 | 31.3 | 4 | 34.7 | 4 | 0.0208 |
| 5 | 32.3 | 5 | 59.2 | 5 | 0.0218 |
| 6 Bottom | 59.1 | 6 Bottom | 126 | 6 Bottom | 0.0350 |
| Total | 86 200 | Total | 73 700 | Total | 1.78 |

surface.

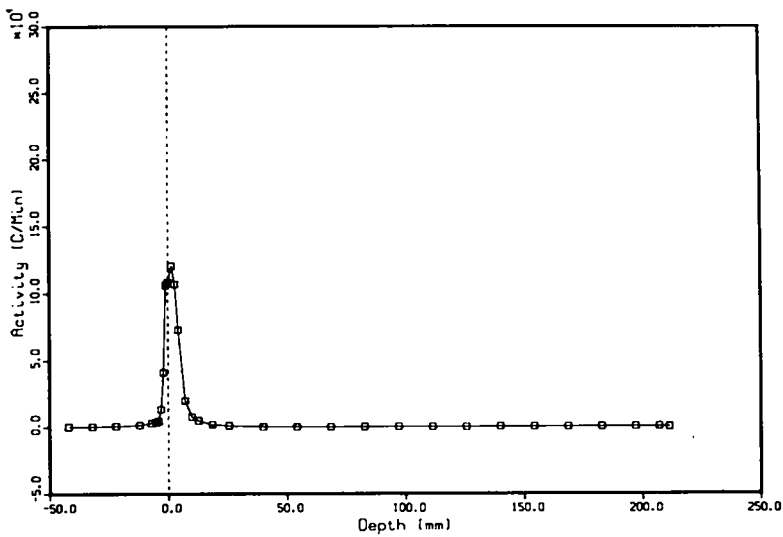
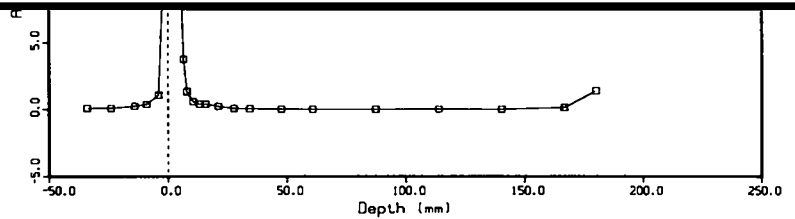


Fig. 7. Gamma activity profile for soil core number 3, which was collected 27 days after deposition of the plutonium oxide particles on the soil surface.

Fig. 8. Gamma activity profile for soil core number 4, which was collected 35 days after deposition of the plutonium oxide particles on the soil surface.

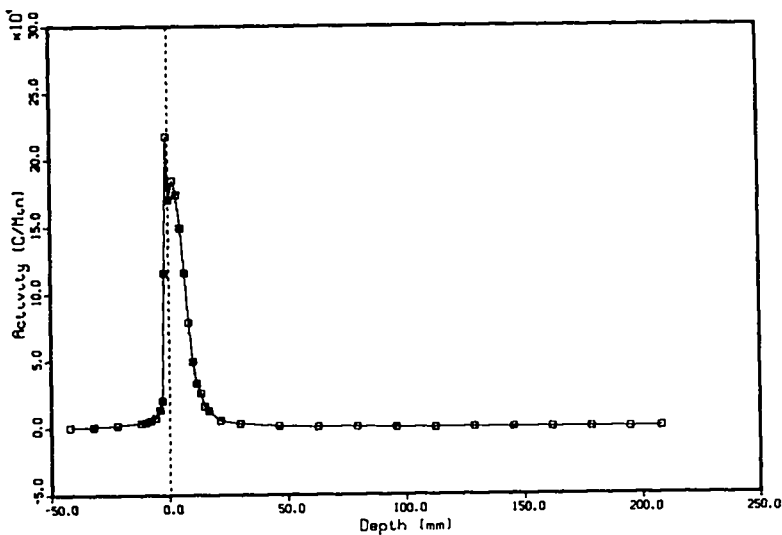
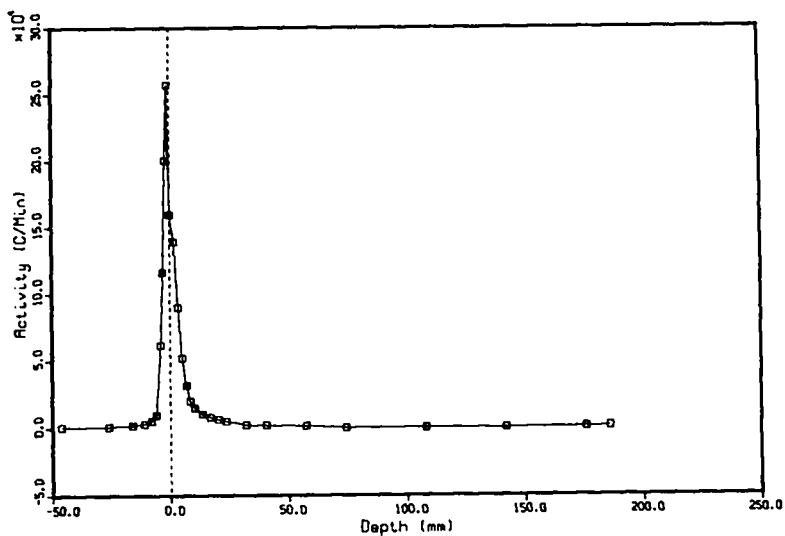
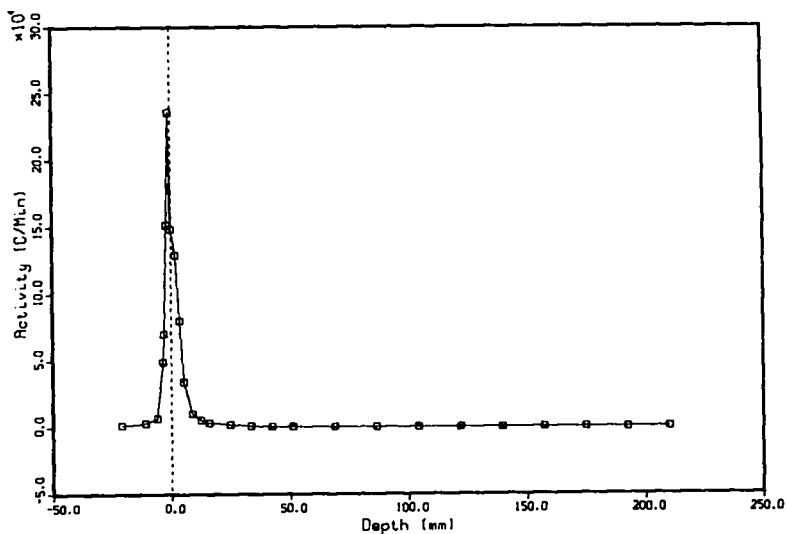


Fig. 9. Gamma activity profile for soil core number 5, which was collected 46 days after deposition of the plutonium oxide particles on the soil surface.

Fig. 10. Gamma activity profile for soil core number 6, which was collected 63 days after deposition of the plutonium oxide particles on the soil surface.



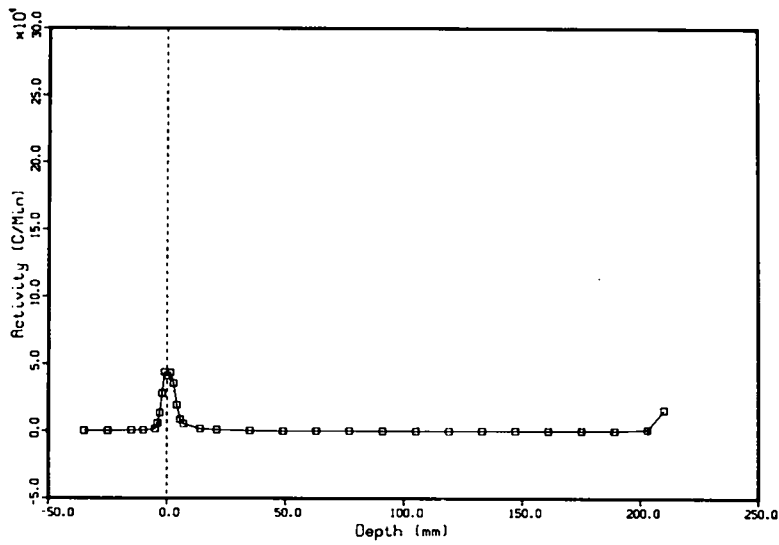


Fig. 11. Gamma activity profile for soil core number 7, which was collected 104 days after deposition of the plutonium oxide particles on the soil surface.

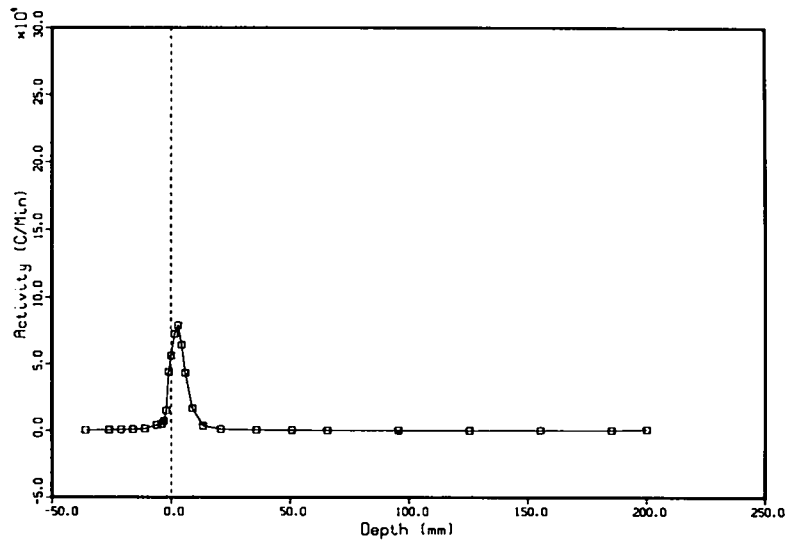


Fig. 12. Gamma activity profile for soil core number 8, which was collected 147 days after deposition of the plutonium oxide particles on the soil surface.

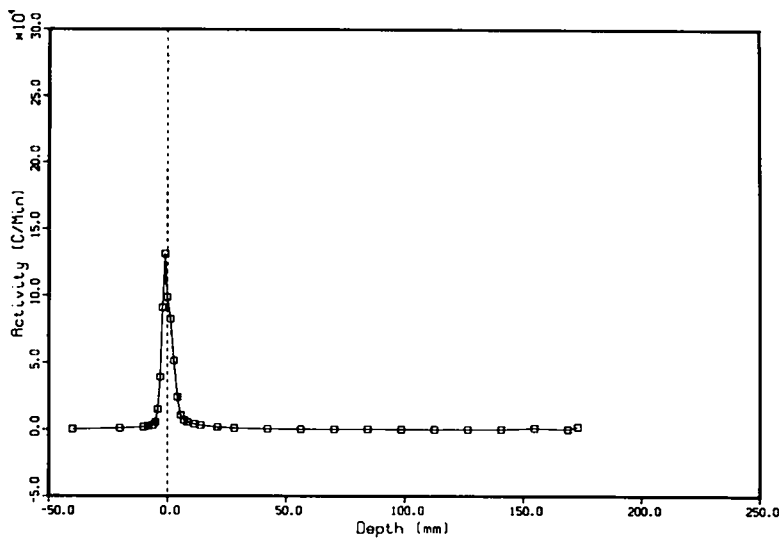


Fig. 13. Gamma activity profile for soil core number 9, which was collected 175 days after deposition of the plutonium oxide particles on the soil surface.

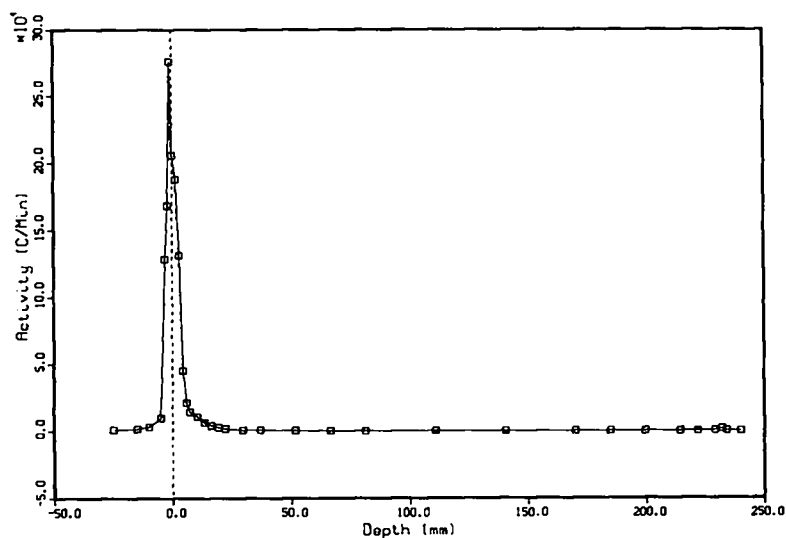


Fig. 14. Gamma activity profile for soil core number 10, which was collected 273 days after deposition of the plutonium oxide particles on the soil surface.

An additional attempt was made to determine the plutonium depth distribution by gamma counting. This involved lowering a sodium iodide detector into a plastic-walled well in the soil tray and measuring count rates as a function of depth. The detector was wrapped with lead tape in an attempt to restrict the detector response to activity located below the plane of the bottom of the detector. Results of several of the counting profiles are shown in Fig. 15. These exponential decay curves are highly suggestive of absorption processes. We suspect that the soft gamma rays emitted by the plutonium located on top of the soil are simply being scattered and absorbed by the soil, which is all around the detector. Thus we observe an exponential decay in count rate as the detector is moved deeper into the well and farther from the source of activity. This interpretation is consistent with the results discussed above, but these measurements do not add a great deal to our understanding of the plutonium behavior. Consequently, these measurements were discontinued early in the experiment.

Soil Percolates

Drainages from the soil tray were collected after each simulated rain and their pH and plutonium contents were determined. Results are summarized in Table A-VII. The pH averaged 8.01 and ranged from 7.70 to 8.63. There was no apparent trend in the pH values; they varied randomly about the mean and were clearly controlled by the soil. The pH did not vary enough during the experiment to cause any changes in plutonium mobility.

The cumulative plutonium contents of the percolates are shown versus time in Fig. 16. This curve shows two distinct inflections, one at 152 days and the other at 189 days. Until the first inflection point, there was no significant release of plutonium. We interpret this as an induction period, during which the soil becomes saturated with the plutonium that is released thereafter. We are not certain which form of plutonium this is. Whatever the form, it is a very minor constituent because the vast majority of the plutonium remains on the top of the soil. After the induction period but before the second inflection point, plutonium is released into the soil drainage at a constant rate of 5.3×10^{-4} ng/m²/s. After the second inflection point, the release rate changes to 4.6×10^{-4} ng/m²/s. The only other environmental chamber experiment for which the surface area of the source was known was the four-soil experiment.³ In this experiment, plutonium was released into the soil percolates at the rate of 6.2×10^{-4} to 7.3×10^{-4} ng/m²/s. In view of all the uncertainties involved, the agreement between these two experiments is remarkable. The percolates from the four-soil experiment do not include the plutonium washed directly off the source because the source was placed on a pedestal and the direct wash-off was collected separately. The agreement between the percolate release rates for these two experiments suggests that the plutonium washed directly off the PuO₂ is not the primary source of the plutonium that appears in the soil percolates.

The second inflection point in Fig. 16 coincides with the end of the rainy season in the chamber climate cycle. This finding suggests that the change in slope is simply due to a change in the flow rate of water through the soil. To test this hypothesis, the data were replotted versus

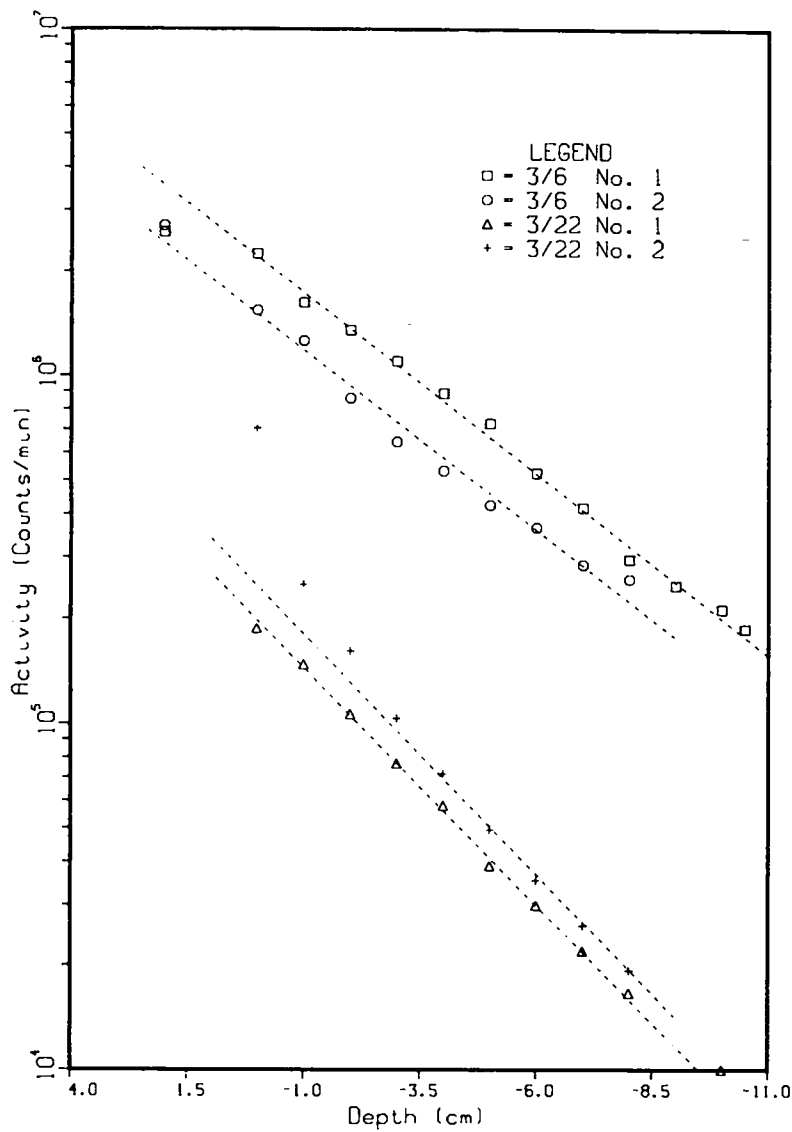
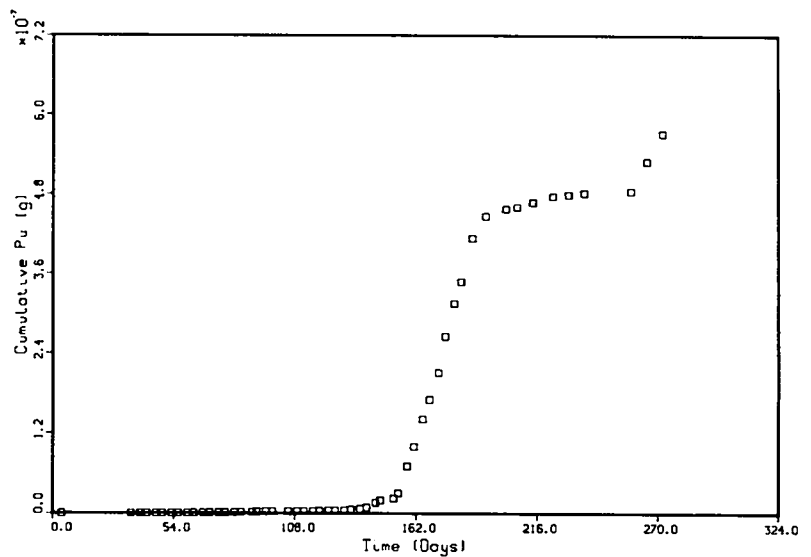


Fig. 15. Gamma activity profiles determined *in situ* with a movable detector within a sampling well.

Fig. 16. Cumulative plutonium released into the soil percolates versus time. The summer season and the rainy season began at 51 days. The summer season ended and the fall season began at 123 days. The rainy season ended at 187 days, and the winter season began at 235 days. The two curve inflections occur at approximately 152 days and 189 days.



the cumulative percolate volume (Fig. 17). When the data are plotted in this way, the inflection point vanishes, indicating that our hypothesis is correct. The slope of the latter part of the curve is $17.6 \text{ ng/m}^2/\text{L}$. When the data from the four-soil experiment were recalculated in this way, the results ranged from 25 to 29 $\text{ng/m}^2/\text{L}$.

The idea that the release of plutonium into the soil percolates may depend on the eluent volume and not on time has some interesting ramifications. For plutonium to emerge from the bottom of the soil, it must either be physically transported through the soil, as through cracks or channels, or the soil must be saturated with respect to the material emerging from the bottom. Because we did not attempt to determine the form of the plutonium in the effluent, these alternatives cannot be distinguished directly, but we can make some inferences. If the soil were at equilibrium with the plutonium emerging from the soil, the rate at which the plutonium emerges from the soil must equal the rate that it is being deposited in the soil. If we assume that this rate is constant, the soil loading for this form of plutonium can be calculated by multiplying the length of the induction period (in liters) times the release rate (in grams per liter) after the inflection point in Fig. 17. This gives a soil loading of 0.01 ng/g , which is orders of magnitude lower than the soil loadings observed in soil column experiments⁴ performed with soils similar to that used in this experiment. This alone suggests that the soil is not at equilibrium. We can also calculate a distribution coefficient by dividing the soil loading by the average concentration of the plutonium in the eluent, giving a value of 2.2 mL/g . This value is also

orders of magnitude lower than accepted values for natural waters and sediments.

These arguments suggest that the soil is not at equilibrium with any form of plutonium. Because the release rate depends on the volume of the eluent and not on time, the release process is not rate limited, at least on the time scale of this experiment. We think that the likeliest mode of transport is with the plutonium attached to very small soil particles and that the release rate is determined by how quickly these particles migrate through the soil column and are washed out into the soil drainage. If true, this supposition has some important consequences. First, a model of such a system must include particle migration processes and fracture flow calculations. Second, because the types and sizes of particles that can easily move through soil columns are severely limited, the plutonium transport through the soil is also limited. Finally, the released plutonium may not be in a readily soluble form and might not be readily assimilated by an organism that ingests it.

Air Samples

Data from the air samples are summarized in Table II. When studying this table, it is important to note not only the plutonium concentrations in the air, but also the duration of the sample. Samples collected during and immediately after a simulated rainfall typically covered only 15 min, whereas samples collected between rainfalls often encompassed a day or more. Table II shows that the plutonium concentrations in the air are

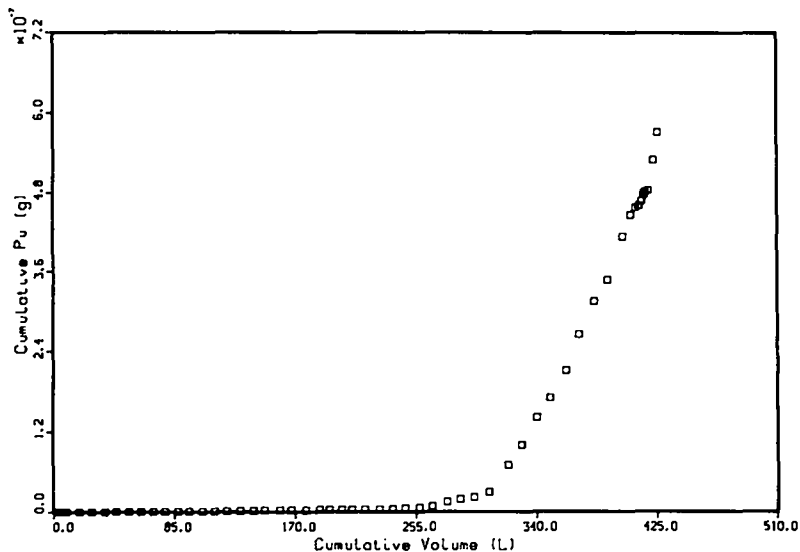


Fig. 17. Cumulative plutonium released into the soil percolates versus percolate volume.

TABLE II. Air Sample Results

| Sample ^{a,b} | Time from Rain (days) | Duration (h) | Average Concentration (fCi/L) | |
|-----------------------|-----------------------|--------------|-------------------------------|---------|
| 1-1 | -6.926 | 22.28 | 196 | |
| 1-2 | -5.997 | 91.60 | 18.4 | |
| 1-3 | -0.112 | 2.683 | 13.6 | |
| 1-4 | 0.000 | 0.283 | 4580 | Rain 2 |
| 1-5 | 0.012 | 18.95 | 237 | |
| 1-6 | 4.040 | 91.98 | 16.2 | |
| 2-1 | -5.027 | 24.33 | 1.84 | |
| 2-2 | -4.740 | 6.57 | 34.0 | |
| 2-3 | -1.037 | 24.88 | 2.55 | |
| 2-4 | 0.000 | 0.258 | 1420 | Rain 54 |
| 2-5 | 0.010 | 0.250 | 541 | |
| 2-6 | 0.021 | 3.508 | 15.6 | |
| 2-7 | 0.167 | 24.46 | 0.645 | |
| 2-8 | 1.186 | 21.04 | 0.403 | |
| 3-9 | -2.065 | 27.69 | 0.548 | |
| 3-10 | -0.911 | 21.86 | 3.98 | |
| 3-11 | 0.000 | 0.247 | 1390 | Rain 55 |
| 3-12 | 0.010 | 0.250 | 352 | |
| 3-13 | 0.978 | 23.24 | 2.75 | |
| 3-14 | 2.020 | 24.99 | 0.960 | |
| 3-15 | 2.239 | 5.267 | 7.01 | |
| 4-16 | -2.051 | 49.22 | 0.532 | |
| 4-17 | 0.000 | 0.250 | 1190 | Rain 56 |
| 4-18 | 0.010 | 0.250 | 558 | |
| 4-19 | 0.303 | 6.783 | 5.59 | |
| 4-20 | 2.181 | 45.07 | 0.917 | |

^aSamples 2-1 through 4-20 represent a continuous series.

^bThe time between Rains 54 and 55 was 7.008 days.

The time between Rains 55 and 56 was 6.988 days.

dramatically higher during the simulated rainfalls than at any other time. Samples collected immediately after the rainfall were substantially reduced in plutonium content, and those taken between rains contained the lowest concentrations of all.

In a previous environmental chamber experiment, a similar pattern was observed for airborne plutonium.⁵ In that experiment, which involved large fragments of PuO₂ fuel pellets, the concentration of plutonium in the air increased by several orders of magnitude during the first 5 min of the simulated rainfall and then decreased exponentially, with a half-time of less than 10 min until the end of the rainfall. After the rain ended, the concentration returned to its prerin value within 3 h. The rapid increase in concentration at the beginning of the

rainfall was attributed to spalling of the hot source fragments from thermal shock when struck by cold rainwater. The decay was attributed to washout of the airborne particles by the rain. When the same experiment was carried out with fines, the dramatic increase in plutonium concentration during rainfall was not observed, and the concentration after the rainfall was substantially lower than before the rainfall. The results of the present experiment resemble more closely those from the large source fragments in the past experiment; there is the large increase in airborne plutonium during the rainfall. However, the mechanism is clearly not spallation (because the source consisted of fines) but is probably a resuspension process. We should note that the rainfall used in the present experiment lasted for 15

min, whereas that in the older experiment lasted more than 1 h. If resuspension is the predominating mechanism for generating airborne plutonium when source fines are used, we would expect a more pronounced effect in the present experiment in which a shorter, more vigorous rainfall was used.

Over 13 days that included three rainfalls, the average air concentration was 7 fCi/L. More than 45% of the total activity observed in the air was collected during the rainfall periods, which constituted less than 1 h out of the 13 days. This average might represent a typical release from source fines deposited on Florida soil, but only to the extent that the frequency and intensity of the rainfall are typical of a Florida environment. The volume of air in the test chamber is approximately 2400 L. Thus the total amount of plutonium suspended in the air, on average, is 17 pCi. This amounts to $8 \times 10^{-12}\%$ of the original source; in terms of plutonium deposited on the soil, the quantity in the air is completely insignificant. The maximum plutonium concentration observed over this same time was 1420 fCi/L, during rain number 54. This represents 15 min during which approximately 3.4 nCi of plutonium was suspended in the air. This value, amounting to $1.6 \times 10^{-9}\%$ of the source, is also insignificant compared with the source, but does represent a significantly increased air concentration. For airborne exposure, the period during and especially at the beginning of a rainfall is the time of maximum risk. Of course this discussion does not apply to airborne plutonium during and immediately after deposition of

the source, at which time the airborne plutonium concentrations are very much higher.

Dehumidifier Condensates

Condensates from the refrigeration-type dehumidifier were collected after each simulated rain, and their pH and plutonium contents were determined. Results are summarized in Table A-VIII. The pH averaged 6.2 and ranged from 5.4 to 7.0. These solutions were unbuffered, and precise pH measurements were difficult to obtain. There were no observable trends in the pH values.

The plutonium released into the dehumidifier condensates versus time is shown in Fig. 18. As the volume of condensate released over time was virtually constant at 2 L/day, the same curve results if the data are plotted versus cumulative volume. Figure 18 shows three distinct regions: an induction period lasting for 30 days, a linear increase from 30 to 160 days, and another linear region lasting until the end of the experiment. The induction period probably represents the time required for all the piping in the test chamber to equilibrate with the contaminated effluents. The first linear region reflects a constant plutonium release rate of 0.23 pg/m²/s (0.12 pg/m²/L), and the second linear portion of the curve reflects a constant plutonium release rate of 0.03 pg/m²/s (0.015 pg/m²/L). The inflection point between these two slopes does not correspond with any

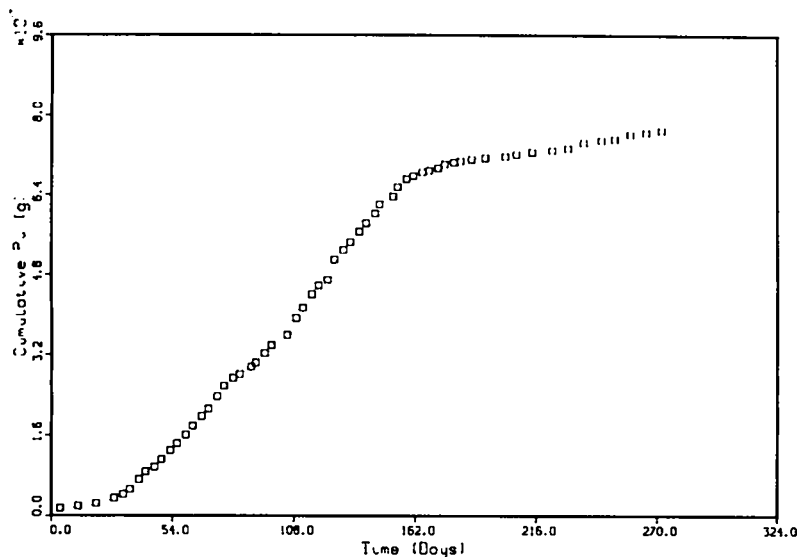


Fig. 18. Cumulative plutonium released into the dehumidifier condensates versus time. The summer season and the rainy season began at 51 days. The summer season ended and the fall season began at 123 days. The rainy season ended at 187 days, and the winter season began at 235 days. The inflection in the curve occurs at 160 days.

known event in the experiment or the chamber climate cycle. Thus the reason for the change in slope is unknown. There appear to be no seasonal effects on the plutonium released into the dehumidifier condensates.

Comparing the observed plutonium release rates with those from previous environmental chamber experiments yields some interesting results. In the four-soil experiment,³ the plutonium released into condensate samples that did not contain direct wash-off from the source amounted to 0.072 ng/m²/s, more than 300 times that in the present experiment. The releases into the soil percolates, discussed above, were comparable. The difference in condensate behaviors derives from the natures of the sources themselves. The source in the four-soil experiment was a single large piece of plutonium oxide that was subject to spalling when cold rainwater contacted the hot fuel pellet. This mechanism was not operative in the present experiment, which contained fines as the source. Similar behavior was observed in earlier experiments in which large fragments from impact-tested sources were placed in one environmental test chamber while the fines from the same source were placed in a second.⁵

The dehumidifier condensate data probably reflect trends in the airborne plutonium concentration, although they are not a quantitative measure of this. Quantitative data may be obtained from the air samples described above. However, Fig. 18 suggests that the airborne plutonium concentration was probably lower at the end of the experiment, after the inflection point at 160 days, than it was earlier. The air samples discussed earlier were mostly collected after this inflection, so the concentrations observed earlier may have been higher. In fact, the one set of data taken very early in the experiment does suggest a value 5 times higher, but these data are complicated by their proximity to the source introduction. Although of academic interest, the observed airborne plutonium levels would have to be many orders of magnitude higher for this to become a significant release pathway.

CONCLUSIONS

This experiment clearly demonstrates that plutonium oxide particulates (20 to 74 μm in diameter), when deposited on Florida soil in a simulated Florida environment, remain in the top 1 to 2 cm of the soil and do not migrate down into the soil, at least on the time scale of this experiment (1 yr).

Plutonium releases into the soil drainages amounted to 17.6 ng/m²/L and appeared to correlate with the volume of the drainage rather than with time. This release compares well with those observed in similar past experiments. Plutonium released in this way represents a very minor constituent of the source. The evidence suggests but does not prove that plutonium released into the soil drainages is associated with soil particles and that the rate of release may depend on the rate at which such particles can migrate through the soil column and be washed out of the bottom. Thus soil particle migration may be important in modeling this release pathway.

The plutonium concentration observed in the air was much higher during simulated rainfalls than at any other time. Air samples collected immediately after a rainfall contained substantially lower amounts of plutonium, whereas those collected between rainfalls contained the lowest amount of all. The average air concentration observed over 13 days near the end of the experiment was 7 fCi/L. The total amount of plutonium in the air during this time amounted to $8 \times 10^{-12}\%$ of the source. In terms of the source, the release of airborne plutonium is insignificant. However, during the brief period at the beginning of a rainfall the airborne concentration reached 1400 fCi/L. Thus, although the overall airborne plutonium release was small, it occurred as a series of sharp spikes during simulated rainfalls.

Plutonium was released into the dehumidifier condensates at a rate 300 times less than that observed in a previous experiment involving a single large piece of plutonium oxide as the source, even though the percolate release rates for the two experiments agree. This difference is attributed to the difference in the sources. The single large piece of plutonium oxide may have been subject to spalling of particles off the surface by cold water contacting the hot fuel surface, whereas the plutonium oxide fines were not subject to such spalling. Similar differences in behavior have been observed in the past. Condensate data suggest that air concentrations of plutonium were lower at the end of the experiment.

All the release pathways examined in this experiment (release into the soil drainages, release into the air, and migration of the plutonium oxide particles into the soil), were shown to be insignificant in relation to the particles themselves. Thus the major transport pathway for the plutonium must be movement of the particles themselves, probably by the actions of surface waters and erosion processes.

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APPENDIX. EXPERIMENTAL DATA TABLES

TABLE A-I. Florida Soil Data^{a,b}

| | |
|--|------|
| Total sand | 92 |
| Total silt | 4.9 |
| Total clay | 3.1 |
| Total organic carbon | 2.41 |
| Particle size analysis | |
| >2 mm | 2 |
| >0.1 mm | 88 |
| 1 mm < sand < 2 mm | 0.3 |
| 0.5 mm < sand < 1.0 mm | 4.9 |
| 0.25 mm < sand < 0.5 mm | 16.8 |
| 0.1 mm < sand < 0.25 mm | 65.4 |
| 0.05 mm < sand < 0.1 mm | 4.6 |
| 0.02 mm < silt < 0.05 mm | 1.7 |
| 0.002 < silt < 0.02 | 3.2 |
| clay < 0.0002 mm | 2.4 |
| Free iron (dith-cit extractable) | 0.2 |
| Aluminum (dith-cit extractable) | 0.1 |
| NH ₄ OAc extractable Mg (meq/100 g) | 1.5 |
| NH ₄ OAc extractable Na | - |
| NH ₄ OAc extractable K (meq/100 g) | 0.4 |
| Cation exchange capacity (pH 7)(meq/100 g) | 8.1 |
| Calcium carbonate equivalent | 2 |
| pH (1:2 0.01M CaCl ₂) | 6.7 |
| pH (1:1 H ₂ O) | 6.9 |
| Surface area (EGME) (m ² /g) | 35 |
| Moisture content at 15 bar | 6.2 |

^aAll values are in weight percent unless otherwise noted.

^bAnalyses were performed by the US Department of Agriculture Soil Conservation Service Laboratory, Lincoln, NE 68508.

TABLE A-II. Diurnal Temperature and Humidity Data for Spring Season

| Hour | Temperature (°F) | Relative Humidity (%) | Temperature at 5 cm (°F) | Wet Bulb Depression (°F) |
|------|---------------------|-----------------------------|--------------------------------|--------------------------------|
| 00 | 67.1 | 84.2 | 62.9 | 3.2 |
| 01 | 65.1 | 84.7 | 61.0 | 3.0 |
| 02 | 63.9 | 85.1 | 59.1 | 3.0 |
| 03 | 62.8 | 85.5 | 58.0 | 2.8 |
| 04 | 61.9 | 85.7 | 58.0 | 2.7 |
| 05 | 61.6 | 85.8 | 58.6 | 2.8 |
| 06 | 62.2 | 85.8 | 59.9 | 2.8 |
| 07 | 64.4 | 85.3 | 64.7 | 3.2 |
| 08 | 68.5 | 84.0 | 71.5 | 3.8 |
| 09 | 71.5 | 79.6 | 75.7 | 5.1 |
| 10 | 74.8 | 69.9 | 80.5 | 8.0 |
| 11 | 77.9 | 60.8 | 83.4 | 11.1 |
| 12 | 79.2 | 56.5 | 84.7 | 12.8 |
| 13 | 80.1 | 55.7 | 84.5 | 13.0 |
| 14 | 80.6 | 55.7 | 83.8 | 12.8 |
| 15 | 80.6 | 55.7 | 82.9 | 12.8 |
| 16 | 80.6 | 55.7 | 82.0 | 12.8 |
| 17 | 80.4 | 55.9 | 79.6 | 12.2 |
| 18 | 78.5 | 58.8 | 75.2 | 10.8 |
| 19 | 75.0 | 71.0 | 71.4 | 7.0 |
| 20 | 72.1 | 78.7 | 68.4 | 5.2 |
| 21 | 70.0 | 81.2 | 66.4 | 4.2 |
| 22 | 68.7 | 82.7 | 65.2 | 3.7 |
| 23 | 68.1 | 83.6 | 64.1 | 3.4 |
| 24 | 67.1 | 84.2 | 62.9 | 3.2 |

TABLE A-III. Diurnal Temperature and Humidity Data for Summer Season

| Hour | Temperature (°F) | Relative Humidity (%) | Temperature at 5 cm (°F) | Wet Bulb Depression (°F) |
|------|---------------------|-----------------------------|--------------------------------|--------------------------------|
| 00 | 76.8 | 89.5 | 73.0 | 2.4 |
| 01 | 75.0 | 89.7 | 71.2 | 2.3 |
| 02 | 73.9 | 90.0 | 69.6 | 2.2 |
| 03 | 72.9 | 90.2 | 68.5 | 2.2 |
| 04 | 72.0 | 90.2 | 68.5 | 2.2 |
| 05 | 71.8 | 90.0 | 69.0 | 2.2 |
| 06 | 72.3 | 89.7 | 70.2 | 2.3 |
| 07 | 74.4 | 88.7 | 74.6 | 2.6 |
| 08 | 78.1 | 86.8 | 80.8 | 3.3 |
| 09 | 80.8 | 82.8 | 84.6 | 4.6 |
| 10 | 83.8 | 75.3 | 89.0 | 7.1 |
| 11 | 86.6 | 69.0 | 91.6 | 9.3 |
| 12 | 87.8 | 66.7 | 92.8 | 10.1 |
| 13 | 88.6 | 65.4 | 92.6 | 10.6 |
| 14 | 89.1 | 65.0 | 92.0 | 10.8 |
| 15 | 89.1 | 65.0 | 91.2 | 10.5 |
| 16 | 89.1 | 65.1 | 90.4 | 10.3 |
| 17 | 88.9 | 66.0 | 88.1 | 10.0 |
| 18 | 87.2 | 71.3 | 84.2 | 7.9 |
| 19 | 84.0 | 78.9 | 80.8 | 5.5 |
| 20 | 81.3 | 83.8 | 78.0 | 4.0 |
| 21 | 79.4 | 86.3 | 76.1 | 3.4 |
| 22 | 78.3 | 87.8 | 75.1 | 2.8 |
| 23 | 77.8 | 88.8 | 74.0 | 2.6 |
| 24 | 76.8 | 89.5 | 73.0 | 2.4 |

TABLE A-IV. Diurnal Temperature and Humidity Data for Fall Season

| Hour | Temperature (°F) | Relative Humidity (%) | Temperature at 5 cm (°F) | Wet Bulb Depression (°F) |
|-------------|-----------------------------|--------------------------------------|---|---|
| 00 | 67.1 | 85.8 | 62.9 | 2.9 |
| 01 | 65.1 | 86.3 | 61.0 | 2.8 |
| 02 | 63.9 | 86.8 | 59.1 | 2.6 |
| 03 | 62.8 | 87.3 | 58.0 | 2.4 |
| 04 | 61.9 | 87.7 | 58.0 | 2.3 |
| 05 | 61.6 | 88.0 | 58.6 | 2.3 |
| 06 | 62.2 | 88.1 | 59.9 | 2.3 |
| 07 | 64.4 | 88.0 | 64.7 | 2.5 |
| 08 | 68.5 | 87.0 | 71.5 | 3.0 |
| 09 | 71.5 | 83.3 | 75.7 | 4.1 |
| 10 | 74.8 | 72.7 | 80.5 | 7.2 |
| 11 | 77.9 | 66.3 | 83.4 | 9.5 |
| 12 | 79.2 | 64.2 | 84.7 | 10.2 |
| 13 | 80.1 | 63.7 | 84.5 | 10.3 |
| 14 | 80.6 | 63.7 | 83.8 | 10.3 |
| 15 | 80.6 | 63.6 | 82.9 | 10.2 |
| 16 | 80.6 | 63.6 | 82.0 | 10.1 |
| 17 | 80.4 | 64.1 | 79.6 | 9.7 |
| 18 | 78.5 | 68.3 | 75.2 | 8.0 |
| 19 | 75.0 | 79.0 | 71.4 | 5.0 |
| 20 | 72.1 | 82.0 | 68.4 | 4.0 |
| 21 | 70.0 | 83.5 | 66.4 | 3.6 |
| 22 | 68.7 | 84.5 | 65.2 | 3.4 |
| 23 | 68.1 | 85.2 | 64.1 | 3.1 |
| 24 | 67.1 | 85.8 | 62.9 | 2.9 |

TABLE A-V. Diurnal Temperature and Humidity Data for Winter Season

| Hour | Temperature (°F) | Relative Humidity (%) | Temperature at 5 cm (°F) | Wet Bulb Depression (°F) |
|------|---------------------|-----------------------------|--------------------------------|--------------------------------|
| 00 | 54.6 | 84.8 | 49.9 | 2.0 |
| 01 | 52.4 | 85.6 | 47.7 | 2.4 |
| 02 | 51.0 | 86.2 | 45.6 | 2.1 |
| 03 | 49.8 | 86.8 | 44.3 | 2.0 |
| 04 | 48.7 | 87.2 | 44.3 | 2.0 |
| 05 | 48.4 | 87.6 | 45.0 | 1.9 |
| 06 | 49.0 | 87.8 | 46.4 | 1.9 |
| 07 | 51.6 | 88.0 | 51.9 | 2.2 |
| 08 | 56.2 | 87.1 | 59.6 | 3.0 |
| 09 | 59.6 | 84.2 | 64.4 | 3.3 |
| 10 | 63.4 | 78.5 | 69.7 | 4.9 |
| 11 | 66.8 | 68.8 | 73.1 | 7.8 |
| 12 | 68.3 | 61.3 | 74.5 | 9.9 |
| 13 | 69.3 | 59.3 | 74.3 | 10.5 |
| 14 | 69.9 | 58.8 | 73.5 | 10.6 |
| 15 | 69.9 | 58.7 | 72.5 | 10.4 |
| 16 | 69.9 | 58.8 | 71.5 | 10.2 |
| 17 | 69.7 | 59.2 | 68.7 | 9.7 |
| 18 | 67.5 | 65.5 | 63.8 | 7.6 |
| 19 | 63.5 | 77.0 | 59.5 | 4.6 |
| 20 | 60.3 | 80.3 | 56.1 | 3.7 |
| 21 | 57.8 | 82.0 | 53.8 | 3.4 |
| 22 | 56.4 | 83.2 | 52.5 | 3.0 |
| 23 | 55.8 | 84.2 | 51.2 | 2.8 |
| 24 | 54.6 | 84.8 | 49.9 | 2.5 |

TABLE A-VI. Source Isotopic Composition^a

| Pu Isotope | Percent (01-07-81) | Percent (04-07-84) |
|---------------|-----------------------|-----------------------|
| 238 | 83.13 | 82.81 |
| 239 | 14.22 | 14.53 |
| 240 | 2.08 | 2.13 |
| 241 | 0.41 | 0.35 |
| 242 | 0.17 | 0.17. |

^aSpecific activity (04-07-84) = 14.19 Ci/g.

TABLE A-VII. Percolate Data

| Sample | Date (mcdvyr) | Time (Days) | Perc Mass (kg) | Perc Cum Mass (kg) | pH | Pu Conc (g/L) | Total Pu (g) | Cum Pu (g) | Solution Resistance (ohm cm) |
|--------|---------------|-------------|----------------|--------------------|------------|---------------|--------------|------------|------------------------------|
| a01r | 030584 | 0.4000e+01 | 0.3040e+01 | 0.3040e+01 | 0.8630e+01 | 0.1640e-10 | 0.4986e-10 | 0.4986e-10 | |
| b02r | 031384 | 0.1200e+02 | 0. | 0.3040e+01 | | | 0. | 0.4986e-10 | |
| a03r | 032184 | 0.2000e+02 | 0. | 0.3040e+01 | | | 0. | 0.4986e-10 | |
| c04r | 032984 | 0.2800e+02 | 0. | 0.3040e+01 | | | 0. | 0.4986e-10 | |
| b05r | 040284 | 0.3200e+02 | 0. | 0.3040e+01 | | | 0. | 0.4986e-10 | |
| b06r | 040584 | 0.3500e+02 | 0.5950e+01 | 0.8990e+01 | 0.8190e+01 | 0.1540e-10 | 0.9162e-10 | 0.1415e-09 | 0.2350e+04 |
| a07r | 040984 | 0.3900e+02 | 0.8990e+01 | 0.1798e+02 | 0.8010e+01 | 0.5000e-11 | 0.4495e-10 | 0.1864e-09 | |
| c08r | 041284 | 0.4200e+02 | 0.8850e+01 | 0.2683e+02 | 0.7880e+01 | 0.5000e-11 | 0.4425e-10 | 0.2307e-09 | 0.1850e+04 |
| a09r | 041684 | 0.4600e+02 | 0.9260e+01 | 0.3609e+02 | 0.7900e+01 | 0.5000e-11 | 0.4630e-10 | 0.2770e-09 | |
| b10r | 041984 | 0.4900e+02 | 0.7850e+01 | 0.4394e+02 | 0.8100e+01 | 0.5680e-11 | 0.4459e-10 | 0.3216e-09 | |
| a11r | 042384 | 0.5300e+02 | 0.9080e+01 | 0.5302e+02 | 0.8170e+01 | 0.5000e-11 | 0.4540e-10 | 0.3670e-09 | 0.1830e+04 |
| b12r | 042684 | 0.5600e+02 | 0.8400e+01 | 0.6142e+02 | 0.7750e+01 | 0.1090e-10 | 0.9156e-10 | 0.4585e-09 | |
| a13r | 043084 | 0.6000e+02 | 0.8670e+01 | 0.7009e+02 | 0.7950e+01 | 0.5000e-11 | 0.4335e-10 | 0.5019e-09 | |
| b14r | 050384 | 0.6300e+02 | 0.8260e+01 | 0.7835e+02 | 0.7940e+01 | 0.2190e-10 | 0.1809e-09 | 0.6828e-09 | 0.1690e+04 |
| a15r | 050784 | 0.6700e+02 | 0.9120e+01 | 0.8747e+02 | 0.7820e+01 | 0.1380e-10 | 0.1259e-09 | 0.8086e-09 | 0.1830e+04 |
| b16r | 051084 | 0.7000e+02 | 0.7880e+01 | 0.9535e+02 | 0.8030e+01 | 0.5000e-11 | 0.3940e-10 | 0.8480e-09 | 0.1830e+04 |
| c17r | 051484 | 0.7400e+02 | 0.9310e+01 | 0.1047e+03 | 0.7800e+01 | 0.5000e-11 | 0.4655e-10 | 0.8346e-09 | 0.1710e+04 |
| a18r | 051784 | 0.7700e+02 | 0.8490e+01 | 0.1132e+03 | 0.7830e+01 | 0.5000e-11 | 0.4245e-10 | 0.9370e-09 | 0.1750e+04 |
| a19r | 052184 | 0.8100e+02 | 0.8540e+01 | 0.1217e+03 | 0.7960e+01 | 0.2860e-10 | 0.2442e-09 | 0.1181e-09 | 0.1790e+04 |
| b20r | 052484 | 0.8400e+02 | 0.9490e+01 | 0.1312e+03 | 0.8260e+01 | 0.3460e-10 | 0.3284e-09 | 0.1510e-09 | 0.1890e+04 |
| a21r | 052984 | 0.8900e+02 | 0.9350e+01 | 0.1405e+03 | 0.8170e+01 | 0.3280e-10 | 0.3067e-09 | 0.1816e-08 | 0.2280e+04 |
| c22r | 053184 | 0.9100e+02 | 0.9100e+01 | 0.1483e+03 | 0.7820e+01 | 0.5870e-10 | 0.4555e-09 | 0.2272e-08 | 0.1910e+04 |
| b23r | 060484 | 0.9500e+02 | 0.1090e+02 | 0.1592e+03 | 0.7850e+01 | 0.1640e-10 | 0.1788e-09 | 0.2451e-08 | 0.1890e+04 |
| a24r | 060784 | 0.9800e+02 | 0.7950e+01 | 0.1671e+03 | 0.7750e+01 | 0.9620e-11 | 0.7648e-10 | 0.2527e-09 | 0.2050e+04 |
| b25r | 061484 | 0.1050e+03 | 0.1026e+02 | 0.1774e+03 | 0.7940e+01 | 0.1210e-10 | 0.1241e-09 | 0.2651e-08 | 0.1910e+04 |
| c26r | 061884 | 0.1090e+03 | 0.9670e+01 | 0.1871e+03 | 0.8070e+01 | 0.1090e-10 | 0.1054e-09 | 0.2757e-08 | 0.2000e+04 |
| a27r | 062184 | 0.1120e+03 | 0.6900e+01 | 0.1940e+03 | 0.8050e+01 | 0.2990e-10 | 0.2063e-09 | 0.2963e-08 | 0.2005e+04 |
| b28r | 062584 | 0.1160e+03 | 0.8630e+01 | 0.2026e+03 | 0.8310e+01 | 0.2330e-10 | 0.2011e-09 | 0.3164e-08 | 0.2001e+04 |
| a29r | 062884 | 0.1190e+03 | 0.7130e+01 | 0.2097e+03 | 0.7960e+01 | 0.2900e-10 | 0.2068e-09 | 0.3371e-08 | 0.2050e+04 |
| b30r | 070284 | 0.1230e+03 | 0.9080e+01 | 0.2188e+03 | 0.8120e+01 | 0.5930e-10 | 0.5384e-09 | 0.3909e-08 | 0.2020e+04 |
| a31r | 070584 | 0.1260e+03 | 0.1031e+02 | 0.2291e+03 | 0.8300e+01 | 0.5000e-11 | 0.5155e-10 | 0.3961e-08 | 0.2011e+04 |
| b32r | 070984 | 0.1300e+03 | 0.9260e+01 | 0.2384e+03 | 0.7830e+01 | 0.4630e-10 | 0.4287e-09 | 0.4390e-08 | 0.2020e+04 |
| a33r | 071284 | 0.1330e+03 | 0.8780e+01 | 0.2472e+03 | 0.7900e+01 | 0.1120e-09 | 0.9834e-09 | 0.5373e-08 | 0.2040e+04 |
| b34r | 071684 | 0.1370e+03 | 0.1006e+02 | 0.2572e+03 | 0.7750e+01 | 0.1270e-09 | 0.1278e-08 | 0.6650e-08 | 0.2120e+04 |
| a35r | 071984 | 0.1400e+03 | 0.8990e+01 | 0.2662e+03 | 0.7930e+01 | 0.1910e-09 | 0.1717e-08 | 0.8368e-08 | 0.2280e+04 |
| b36r | 072384 | 0.1440e+03 | 0.1049e+02 | 0.2767e+03 | 0.7850e+01 | 0.6850e-09 | 0.7186e-08 | 0.1555e-07 | 0.2350e+04 |
| a37r | 072584 | 0.1460e+03 | 0.9030e+01 | 0.2857e+03 | 0.8010e+01 | 0.4030e-09 | 0.3639e-08 | 0.1919e-07 | 0.2350e+04 |
| c38r | 073184 | 0.1520e+03 | 0.9850e+01 | 0.2956e+03 | 0.8240e+01 | 0.2890e-09 | 0.2947e-08 | 0.2204e-07 | 0.2410e+04 |
| a39r | 080284 | 0.1540e+03 | 0.1058e+02 | 0.3062e+03 | 0.7840e+01 | 0.7430e-09 | 0.7861e-08 | 0.2990e-07 | 0.2450e+04 |
| b40r | 080684 | 0.1580e+03 | 0.1348e+02 | 0.3196e+03 | 0.7700e+01 | 0.2990e-08 | 0.4031e-07 | 0.7021e-07 | 0.2810e+04 |

TABLE A-VII. (cont)

| | | | | | | | | | |
|------|--------|------------|------------|------------|------------|------------|------------|------------|------------|
| c41r | 080984 | 0.1610e+03 | 0.9620e+01 | 0.3293e+03 | 0.7820e+01 | 0.3080e-08 | 0.2963e-07 | 0.9983e-07 | 0.2700e+04 |
| b42r | 081384 | 0.1650e+03 | 0.1067e+02 | 0.3399e+03 | 0.7700e+01 | 0.3860e-08 | 0.4119e-07 | 0.1410e-06 | 0.2650e+04 |
| a43r | 081684 | 0.1680e+03 | 0.9350e+01 | 0.3493e+03 | 0.8030e+01 | 0.3140e-08 | 0.2936e-07 | 0.1704e-06 | 0.2850e+04 |
| b44r | 082084 | 0.1720e+03 | 0.1098e+02 | 0.3603e+03 | 0.8080e+01 | 0.3710e-08 | 0.4074e-07 | 0.2111e-06 | 0.2700e+04 |
| a45r | 082384 | 0.1750e+03 | 0.9350e+01 | 0.3696e+03 | 0.7770e+01 | 0.5780e-08 | 0.5404e-07 | 0.2652e-06 | 0.2775e+04 |
| b46r | 082784 | 0.1790e+03 | 0.1078e+02 | 0.3804e+03 | 0.8030e+01 | 0.4600e-08 | 0.4959e-07 | 0.3147e-06 | 0.2770e+04 |
| c47r | 083084 | 0.1820e+03 | 0.9350e+01 | 0.3897e+03 | 0.7830e+01 | 0.3470e-08 | 0.3244e-07 | 0.3472e-06 | 0.2680e+04 |
| a48r | 090484 | 0.1870e+03 | 0.1100e+02 | 0.4007e+03 | 0.7370e+01 | 0.5970e-08 | 0.6567e-07 | 0.4129e-06 | 0.2850e+04 |
| b49r | 091084 | 0.1930e+03 | 0.5650e+01 | 0.4064e+03 | 0.8170e+01 | 0.5910e-08 | 0.3339e-07 | 0.4463e-06 | 0.2720e+04 |
| c50r | 091984 | 0.2020e+03 | 0.3500e+01 | 0.4099e+03 | 0.8210e+01 | 0.3160e-08 | 0.1106e-07 | 0.4573e-06 | 0.2770e+04 |
| b51r | 092484 | 0.2070e+03 | 0.2450e+01 | 0.4123e+03 | 0.8490e+01 | 0.1150e-08 | 0.2818e-08 | 0.4601e-06 | 0.2700e+04 |
| c52r | 100184 | 0.2140e+03 | 0.1820e+01 | 0.4142e+03 | 0.8390e+01 | 0.3990e-08 | 0.7262e-08 | 0.4674e-06 | 0.3240e+04 |
| a53r | 101084 | 0.2230e+03 | 0.1450e+01 | 0.4156e+03 | 0.8290e+01 | 0.6250e-08 | 0.9063e-08 | 0.4765e-06 | 0.3000e+04 |
| c54r | 101784 | 0.2300e+03 | 0.2300e+00 | 0.4158e+03 | 0.8440e+01 | 0.9560e-08 | 0.2199e-08 | 0.4797e-06 | 0.2840e+04 |
| b55r | 102484 | 0.2370e+03 | 0.6600e+00 | 0.4165e+03 | 0.8360e+01 | 0.4270e-08 | 0.2818e-08 | 0.4815e-06 | 0.3040e+04 |
| a56r | 110184 | 0.2450e+03 | 0. | 0.4165e+03 | | | 0. | 0.4815e-06 | |
| c57r | 110784 | 0.2510e+03 | 0. | 0.4165e+03 | | | 0. | 0.4815e-06 | |
| b58r | 111484 | 0.2580e+03 | 0.2360e+01 | 0.4189e+03 | 0.8140e+01 | 0.1080e-08 | 0.2549e-08 | 0.4840e-06 | 0.4400e+04 |
| a59r | 112184 | 0.2650e+03 | 0.3500e+01 | 0.4224e+03 | 0.8180e+01 | 0.1290e-07 | 0.4515e-07 | 0.5292e-06 | 0.3680e+04 |
| c60r | 112884 | 0.2720e+03 | 0.3400e+01 | 0.4258e+03 | 0.8200e+01 | 0.1230e-07 | 0.4182e-07 | 0.5710e-06 | 0.4600e+04 |

TABLE A-VIII. Condensate Data

| Sample | Date (modyyr) | Time (Days) | Cond Mass (Kg) | Cond Cum Mass (Kg) | pH | Pu Conc (g/L) | Total Pu (g) | Cum Pu (g) |
|--------|------------------|----------------|----------------------|--------------------------|------------|---------------------|--------------------|------------------|
| a01c | 030584 | 0.4000e+01 | 0.9920e+01 | 0.9920e+01 | 0.7000e+01 | 0.1500e-08 | 0.1488e-07 | 0.1488e-07 |
| b02c | 031384 | 0.1200e+02 | 0.1071e+02 | 0.2063e+02 | 0.6600e+01 | 0.4060e-09 | 0.4348e-08 | 0.1923e-07 |
| a03c | 032184 | 0.2000e+02 | 0.1037e+02 | 0.3100e+02 | 0.6700e+01 | 0.5010e-09 | 0.5195e-08 | 0.2442e-07 |
| c04c | 032984 | 0.2800e+02 | 0.1634e+02 | 0.4734e+02 | 0.6200e+01 | 0.6380e-09 | 0.1042e-07 | 0.3485e-07 |
| b05c | 040284 | 0.3200e+02 | 0.6720e+01 | 0.5406e+02 | 0.6940e+01 | 0.1120e-08 | 0.7526e-08 | 0.4237e-07 |
| a06c | 040584 | 0.3500e+02 | 0.6310e+01 | 0.6037e+02 | 0.6200e+01 | 0.1610e-08 | 0.1016e-07 | 0.5253e-07 |
| c07c | 040984 | 0.3900e+02 | 0.8900e+01 | 0.6927e+02 | 0.6400e+01 | 0.2210e-08 | 0.1967e-07 | 0.7220e-07 |
| b08c | 041284 | 0.4200e+02 | 0.7580e+01 | 0.7685e+02 | 0.6200e+01 | 0.2010e-08 | 0.1524e-07 | 0.8744e-07 |
| a09c | 041684 | 0.4600e+02 | 0.8630e+01 | 0.8548e+02 | 0.6200e+01 | 0.1070e-08 | 0.9234e-08 | 0.9667e-07 |
| b10c | 041984 | 0.4900e+02 | 0.8080e+01 | 0.9356e+02 | 0.6800e+01 | 0.1900e-08 | 0.1535e-07 | 0.1120e-06 |
| a11c | 042384 | 0.5300e+02 | 0.8720e+01 | 0.1023e+03 | 0.6400e+01 | 0.2100e-08 | 0.1831e-07 | 0.1303e-06 |
| b12c | 042684 | 0.5600e+02 | 0.8170e+01 | 0.1105e+03 | 0.6240e+01 | 0.1660e-08 | 0.1356e-07 | 0.1439e-06 |
| a13c | 043084 | 0.6000e+02 | 0.9120e+01 | 0.1196e+03 | 0.6730e+01 | 0.1900e-08 | 0.1733e-07 | 0.1612e-06 |
| b14c | 050384 | 0.6300e+02 | 0.7720e+01 | 0.1273e+03 | 0.6240e+01 | 0.2280e-08 | 0.1760e-07 | 0.1788e-06 |
| a15c | 050784 | 0.6700e+02 | 0.9760e+01 | 0.1371e+03 | 0.6070e+01 | 0.1990e-08 | 0.1942e-07 | 0.1983e-06 |
| b16c | 051084 | 0.7000e+02 | 0.7260e+01 | 0.1443e+03 | 0.6700e+01 | 0.2030e-08 | 0.1474e-07 | 0.2130e-06 |
| c17c | 051484 | 0.7400e+02 | 0.9490e+01 | 0.1538e+03 | 0.6150e+01 | 0.2590e-08 | 0.2458e-07 | 0.2376e-06 |
| a18c | 051784 | 0.7700e+02 | 0.9170e+01 | 0.1630e+03 | 0.6200e+01 | 0.2260e-08 | 0.2072e-07 | 0.2583e-06 |
| a19c | 052184 | 0.8100e+02 | 0.7170e+01 | 0.1701e+03 | 0.5970e+01 | 0.2190e-08 | 0.1570e-07 | 0.2740e-06 |
| b20c | 052484 | 0.8400e+02 | 0.6950e+01 | 0.1771e+03 | 0.6170e+01 | 0.1090e-08 | 0.7576e-08 | 0.2816e-06 |
| a21c | 052984 | 0.8900e+02 | 0.1060e+02 | 0.1877e+03 | 0.6400e+01 | 0.1410e-08 | 0.1495e-07 | 0.2965e-06 |
| c22c | 053184 | 0.9100e+02 | 0.5170e+01 | 0.1929e+03 | 0.6290e+01 | 0.1520e-08 | 0.7858e-08 | 0.3044e-06 |
| b23c | 060484 | 0.9500e+02 | 0.9670e+01 | 0.2025e+03 | 0.6100e+01 | 0.1980e-08 | 0.1915e-07 | 0.3235e-06 |
| a24c | 060784 | 0.9800e+02 | 0.6720e+01 | 0.2093e+03 | 0.5710e+01 | 0.2400e-08 | 0.1613e-07 | 0.3396e-06 |
| b25c | 061484 | 0.1050e+03 | 0.1330e+02 | 0.2226e+03 | 0.6100e+01 | 0.1520e-08 | 0.2022e-07 | 0.3599e-06 |
| c26c | 061884 | 0.1090e+03 | 0.1235e+02 | 0.2349e+03 | 0.6100e+01 | 0.2710e-08 | 0.3347e-07 | 0.3933e-06 |
| a27c | 062184 | 0.1120e+03 | 0.7490e+01 | 0.2424e+03 | 0.6150e+01 | 0.2770e-08 | 0.2075e-07 | 0.4141e-06 |
| b28c | 062584 | 0.1160e+03 | 0.9350e+01 | 0.2517e+03 | 0.5950e+01 | 0.2800e-08 | 0.2618e-07 | 0.4403e-06 |
| a29c | 062884 | 0.1190e+03 | 0.7990e+01 | 0.2597e+03 | 0.6100e+01 | 0.2170e-08 | 0.1734e-07 | 0.4576e-06 |
| b30c | 070284 | 0.1230e+03 | 0.5180e+01 | 0.2649e+03 | 0.5900e+01 | 0.2220e-08 | 0.1150e-07 | 0.4691e-06 |
| a31c | 070584 | 0.1260e+03 | 0.8670e+01 | 0.2736e+03 | 0.5980e+01 | 0.4670e-08 | 0.4049e-07 | 0.5096e-06 |
| b32c | 070984 | 0.1300e+03 | 0.9080e+01 | 0.2827e+03 | 0.6160e+01 | 0.2150e-08 | 0.1952e-07 | 0.5291e-06 |
| a33c | 071284 | 0.1330e+03 | 0.7290e+01 | 0.2900e+03 | 0.6260e+01 | 0.2190e-08 | 0.1597e-07 | 0.5451e-06 |
| b34c | 071684 | 0.1370e+03 | 0.9120e+01 | 0.2991e+03 | 0.6040e+01 | 0.2290e-08 | 0.2088e-07 | 0.5660e-06 |
| a35c | 071984 | 0.1400e+03 | 0.6720e+01 | 0.3058e+03 | 0.6000e+01 | 0.2490e-08 | 0.1673e-07 | 0.5827e-06 |
| b36c | 072384 | 0.1440e+03 | 0.8580e+01 | 0.3144e+03 | 0.5670e+01 | 0.2290e-08 | 0.1965e-07 | 0.6023e-06 |
| a37c | 072584 | 0.1460e+03 | 0.7850e+01 | 0.3222e+03 | 0.5880e+01 | 0.2380e-08 | 0.1868e-07 | 0.6210e-06 |
| c38c | 073184 | 0.1520e+03 | 0.9220e+01 | 0.3314e+03 | 0.5700e+01 | 0.1690e-08 | 0.1558e-07 | 0.6366e-06 |
| a39c | 080284 | 0.1540e+03 | 0.6810e+01 | 0.3383e+03 | 0.5930e+01 | 0.2760e-08 | 0.1880e-07 | 0.6554e-06 |
| b40c | 080684 | 0.1580e+03 | 0.1030e+02 | 0.3486e+03 | 0.6000e+01 | 0.1570e-08 | 0.1617e-07 | 0.6716e-06 |

TABLE A-VIII. (cont)

| | | | | | | | | |
|------|--------|------------|------------|------------|------------|------------|------------|------------|
| c41c | 080984 | 0.1610e+03 | 0.6450e+01 | 0.3550e+03 | 0.5750e+01 | 0.1030e-08 | 0.6644e-08 | 0.6782e-06 |
| b42c | 081384 | 0.1650e+03 | 0.8440e+01 | 0.3634e+03 | 0.5820e+01 | 0.8480e-09 | 0.7157e-08 | 0.6854e-06 |
| a43c | 081684 | 0.1680e+03 | 0.6450e+01 | 0.3699e+03 | 0.6550e+01 | 0.5590e-09 | 0.3606e-08 | 0.6890e-06 |
| b44c | 082084 | 0.1720e+03 | 0.9940e+01 | 0.3798e+03 | 0.6190e+01 | 0.4950e-09 | 0.4920e-08 | 0.6939e-06 |
| a45c | 082384 | 0.1750e+03 | 0.6400e+01 | 0.3862e+03 | 0.5490e+01 | 0.1290e-08 | 0.8256e-08 | 0.7022e-06 |
| b46c | 082784 | 0.1790e+03 | 0.8310e+01 | 0.3945e+03 | 0.6170e+01 | 0.4040e-09 | 0.3357e-08 | 0.7055e-06 |
| c47c | 083084 | 0.1820e+03 | 0.5950e+01 | 0.4005e+03 | 0.6000e+01 | 0.3510e-09 | 0.2088e-08 | 0.7076e-06 |
| a48c | 090484 | 0.1870e+03 | 0.8670e+01 | 0.4092e+03 | 0.5700e+01 | 0.4590e-09 | 0.3980e-08 | 0.7116e-06 |
| b49c | 091084 | 0.1930e+03 | 0.1135e+02 | 0.4205e+03 | 0.5850e+01 | 0.2500e-09 | 0.2838e-08 | 0.7144e-06 |
| c50c | 091984 | 0.2020e+03 | 0.1220e+02 | 0.4327e+03 | 0.6200e+01 | 0.2560e-09 | 0.3123e-08 | 0.7175e-06 |
| b51c | 092484 | 0.2070e+03 | 0.1220e+02 | 0.4449e+03 | 0.5400e+01 | 0.3600e-09 | 0.4392e-08 | 0.7219e-06 |
| c52c | 100184 | 0.2140e+03 | 0.1330e+02 | 0.4582e+03 | 0.6100e+01 | 0.3100e-09 | 0.4123e-08 | 0.7261e-06 |
| a53c | 101084 | 0.2230e+03 | 0.1460e+02 | 0.4728e+03 | 0.6300e+01 | 0.2830e-09 | 0.4132e-08 | 0.7302e-06 |
| c54c | 101784 | 0.2300e+03 | 0.1200e+02 | 0.4848e+03 | 0.6000e+01 | 0.3290e-09 | 0.3948e-08 | 0.7341e-06 |
| b55c | 102484 | 0.2370e+03 | 0.1250e+02 | 0.4973e+03 | 0.6440e+01 | 0.8980e-09 | 0.1123e-07 | 0.7454e-06 |
| a56c | 110184 | 0.2450e+03 | 0.1360e+02 | 0.5109e+03 | 0.5950e+01 | 0.3690e-09 | 0.5018e-08 | 0.7504e-06 |
| c57c | 110784 | 0.2510e+03 | 0.9940e+01 | 0.5209e+03 | 0.6130e+01 | 0.2660e-09 | 0.2644e-08 | 0.7530e-06 |
| b58c | 111484 | 0.2580e+03 | 0.1044e+02 | 0.5313e+03 | 0.6400e+01 | 0.9370e-09 | 0.9782e-08 | 0.7628e-06 |
| a59c | 112184 | 0.2650e+03 | 0.1071e+02 | 0.5420e+03 | 0.6400e+01 | 0.3170e-09 | 0.3395e-08 | 0.7662e-06 |
| c60c | 112884 | 0.2720e+03 | 0.9990e+01 | 0.5520e+03 | 0.6630e+01 | 0.4140e-09 | 0.4136e-08 | 0.7703e-06 |

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