RECEIVED BY TIC FEB 28 1974

PLUTONIUM IN SURFACE SOIL IN THE
HANFORD PLANT ENVIRONS

J. P. Corley
D. M. Robertson
F. P. Brauer

August 1972

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

BNWL-SA-3967

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Rec

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

PLUTONIUM IN SURFACE SOIL IN THE
HANFORD PLANT ENVIRONS*

J. P. Corley
D. M. Robertson
F. P. Brauer

Analyses for plutonium in air, water, and foodstuffs have long been part of the routine surveillance program at Hanford. In addition, we have surveyed the ground and other surfaces for plutonium where there was possible deposition from stack emissions, waste spills, etc., using direct instrument measurements; detectable plutonium deposition from the few such incidents has been confined to restricted areas. The surface contamination level that can be detected with our portable instruments is approximately $0.007 \mu\text{g Pu per } 100 \text{ cm}^2$.

The desire to obtain additional information regarding any spread of plutonium beyond the restricted areas, as well as to distinguish between any plutonium in soil resulting from plant activities and that resulting from fallout led to a screening survey for plutonium in surface soils both on and off-site in February 1970.

The results to date and the procedures followed are discussed in this paper. Although some additional samples have been taken, the major part of my discussion will be on the initial survey. The limited amount of subsequent data has given results within the same range of plutonium concentrations.

* Presented orally by J. P. Corley at the Environmental Plutonium Symposium, Los Alamos Scientific Laboratory, Los Alamos, New Mexico (August 5, 1971).

Sampling Procedure

In order to minimize the variables associated with the sampling, an attempt was made to select uniform sampling sites. At Hanford, this means desert soils as free as possible from rocks and standing vegetation. Emphasis was placed on the sampling of undisturbed soils and only minimum amounts of rootmat or vegetative litter were accepted. Since the primary objective was to determine the current distribution of plutonium rather than to make a total environmental inventory, the sampling depth was kept to a practical minimum.

Initial sampling was done with a flat-bottomed scoop approximately 18 inches by 12 inches. An attempt was made to take only the top one-half inch of soil. Subsequent sampling has been done with a closed-top sampler to minimize variation in sample depth. An ordinary cellulose tape container gives a neat, sharp-edged, reproducible cut in our desert soils, 9 cm in diameter by 1.6 cm deep, provided no larger gravels are present. A trowel was used to make a clean cut across the bottom edge of the container and to retain the entire sample for transfer to a tared polystyrene sample jar. Repeated cuts within the selected sampling area give a total sample weight of 150 grams or more. Samples of known depth can be taken by removing soil from the side of the implanted container just deep enough to expose its bottom edge, removing the sample, and repeating the procedure.

Analytical Procedures

Plutonium analyses on the samples of February, 1970, were performed by two laboratories; Battelle-Northwest and U. S. Testing Company. The Battelle-Northwest Laboratory procedure used aliquots of 10 grams (dry weight) of soil for plutonium analysis. Each aliquot was spiked with a

nominal 1 dpm of ^{236}Pu and heated for 2-3 hours at 750°C . The soil was then leached with both dilute and concentrated hydrochloric acid followed by concentrated nitric acid. The total acid contact time was 3-4 days. The leach solution (10M in HCl) was loaded on Dowex-1 anion exchange resin and the resin washed with 10M HCl. The plutonium was reduced and eluted with 0.1M ammonium iodide in 5M HCl. The plutonium-bearing effluent was converted to 8M HNO_3 and again loaded onto Dowex-1; the resin was then washed with 8M HNO_3 , and eluted with 1.2N HCl. A final purification was accomplished with a thenoyltrifluoroacetone (TTA) extraction. The plutonium-bearing organic phase was evaporated on a platinum disc, and counted with a 150 mm^2 silicon surface-barrier detector for 8-10 days. Process-blank counting rates were less than one-tenth of the lowest sample counting rate. The detection level by this procedure is estimated to be 0.01 dpm per sample for a 10 day count or about 0.001 dpm/g $^{239+240}\text{Pu}$ of soil. Use of the silicon detector permitted distinction of ^{238}Pu from the $^{239+240}\text{Pu}$.

The ^{137}Cs content of the samples was measured by gamma-ray spectrometry and used to normalize the plutonium results for differences in the fallout content of the various samples. Several hundred grams of sample were placed in a five-inch diameter by three-quarter-inch deep plastic container. The samples were counted for at least 1000 minutes each between a pair of six-inch diameter by five-inch thick NaI(Tl) detectors operated in anticoincidence with a plastic phosphor annulus for Compton suppression and background reduction. A weighted least squares method was used to calculate ^{137}Cs estimates from the spectral data. The calculations gave a precision estimate for the ^{137}Cs analyses of better than $\pm 5\%$.

For a few of the February 1970 samples and for all subsequent samples, a somewhat different plutonium analytical procedure was used by the U. S. Testing Company.* Samples were weighed, oven-dried at 125°C for 24 hours, and manually stirred to mix the sample and break up any clods. Five grams of dried soil were used for hot leaching. One-hundred ml of 8N HNO₃ plus 2 drops of concentrated HF were applied under reflux. The resulting mixture was filtered and washed with hot 1N HNO₃. After evaporating the leach solution to dryness and resolubilizing, a lanthanum fluoride coprecipitation was carried out. TTA extraction and electrolytic deposition were used to purify and mount the plutonium for counting. Counting was performed by exposing NTA film to the plated disc for approximately 1 week. Alpha tracks in the film were counted and converted to dpm as total plutonium. Yield by this procedure was nominally 65%, with an expected detection level of about 0.007 dpm/g of soil. The procedure described following the leach step is the Hanford standard bioassay procedure for plutonium.

Analytical Results

Figure 1 shows the Hanford reservation, the chemical separations areas, the reactor areas, and the laboratory areas, as well as nearby communities. Sampling sites are indicated.

The distance to the nearest chemical separations area has been listed in Table I for all samples outside these areas. Both areas have, in the past, included facilities for liquid processing of irradiated fuels, while the West area also has facilities for processing plutonium to metal and metal fabrications. Much smaller quantities of plutonium have been

* U. S. Testing Company, Richland Branch - A contractor to the Atomic Energy Commission

handled in the 300 (Laboratory) Area. For the analytical data presented in Table I, U. S. Testing Company data are identified with an asterisk; the remainder are Battelle-Northwest data.

Bulk density measurements of the dried soil ranged from 1.35 to 1.65 g/ml, and an average value of 1.5 g/ml was used to convert concentration by weight to surface deposition per unit area.

The right hand column is labeled "Multiple Analyses." The entry "aliquot" in this column indicates analysis of more than one portion of one sample and "sample" indicates analysis of different samples taken at that one site.

The analytical results obtained on several samples taken from one sample site generally showed about the same variation as replicate analyses on one sample. For the analyses performed by the Battelle-Northwest Laboratory, the results were generally within statistically expected range. Values from the other laboratory compared less closely. The variations between sites and between replicate samples are believed due largely to non-uniform distribution, but other sources of inconsistency cannot be ruled out. However, the values obtained are generally in the expected range for plutonium from fallout, based on other reported results.

Summary and Conclusions

The primary intent of the work done was the identification of plutonium from plant releases within the restricted areas and to determine if this plutonium had migrated to areas outside the restricted areas.

Figure 2 shows the $^{239+240}\text{Pu}$ activity concentration as a function of

distance from plutonium processing facilities. The variability is apparent, permitting no positive conclusions. Figure 3 is a plot of $^{239+240}\text{Pu}$ activity concentration versus that activity normalized to ^{137}Cs , attributed to fallout. As is readily seen, the samples marked "W," those from the restricted area containing the plutonium-handling facilities, are distinct from all other samples. These values have a Pu/Cs activity ratio which can be attributed to plant releases. The remaining data, both on and off the reservation, have the same nominal Pu/Cs ratio, a ratio characteristic of regional surface fallout at the time of sampling.

During the operation of the Hanford plant, small localized releases of plutonium have taken place. The samples from the restricted West area conform this fact and indicate that a different Pu/Cs activity ratio is to be expected from that due to fallout. The samples from both outside the restricted area but within the plant boundaries, and outside the plant boundaries, have the same Pu/Cs ratio indicating that the plutonium found is due to fallout and not plant operation.

TABLE I
PLUTONIUM IN SURFACE SOIL AT HANFORD - FEBRUARY-MARCH, 1971
(Laboratory A except as indicated by *)

Distance from Separations Area, Miles	Pu-(239+240) dpm/g	Pu-(239+240) nCi/m ²	Fraction Pu-238/Total Pu	Ratio Pu-(239+240) /CS-137	Replicate
A. Inside Chemical Separations Areas					
	0.13 ± 0.01	1.11	0.23 ± 0.010	0.020	
	0.057 ± 0.004	0.49	<0.03	0.030	
	0.057 ± 0.003	0.49	<0.02	0.024	
	1.23 ± 0.06		0.029 ± 0.003	0.20	Aliquots
	1.54 ± 0.16	11.9	0.033 ± 0.004	0.25	
	0.85 ± 0.04	7.3	0.026 ± 0.003	0.11	
	0.28 ± 0.01	2.41	0.042 ± 0.005	0.07	
B. Within Hanford Site Boundary					
2	0.27 ± 0.01		0.033 ± 0.010	0.029	Aliquots
	0.24 ± 0.02	2.10	0.036 ± 0.007	0.026	
	*0.10	1.63			
2	0.039 ± 0.003		<0.09	0.023	Samples
	0.036 ± 0.004		<0.05	0.041	
	0.033 ± 0.002		<0.22	0.022	
	0.022 ± 0.002	0.28	0.018 ± 0.36	0.026	
2	0.064 ± 0.006		<0.08	0.025	Samples
	0.053 ± 0.003	0.503	<0.18	0.021	
4	0.064 ± 0.003		<0.07	0.040	Aliquots
	0.031 ± 0.005		<0.10	0.019	
	0.035 ± 0.002	0.37	0.06 ± 0.020	0.022	
	*0.092	0.79			
8	0.112 ± 0.006	0.96	0.052 ± 0.008	0.019	
9	0.041 ± 0.003		0.089 ± 0.018	0.023	Samples
	0.080 ± 0.004		0.056 ± 0.012	0.018	
	0.040 ± 0.002	0.46	0.06 ± 0.014	0.020	
9	0.081 ± 0.006		0.043 ± 0.010	0.017	Aliquots
	0.095 ± 0.005	0.76	0.039 ± 0.012	0.019	
	*0.205	1.76			
11	0.092 ± 0.004	0.79	0.041 ± 0.010	0.023	
	*0.031	0.27			
12	0.016 ± 0.002	0.14	0.023 ± 0.010	0.023	
16	0.023 ± 0.002		<0.09	0.016	
	0.022 ± 0.002		<0.16	0.016	
C. Off-Site					
12	0.061 ± 0.005	0.52		0.017	
17	0.008 ± 0.001		0.028 ± 0.056	0.020	Samples
	0.168 ± 0.007	0.76	0.027 ± 0.006	0.024	
	*0.076	0.65			
20	0.025 ± 0.002	0.22	0.014 ± 0.027	0.025	Aliquots
	*0.005	<0.04			
21	0.024 ± 0.003	0.21	<0.06	0.013	Aliquots
	*0.025	0.21			

SAMPLING SITES AT HANFORD FOR PLUTONIUM IN SURFACE SOILS

Figure 2

PLUTONIUM IN TOP LAYER OF SOIL vs DISTANCE FROM SEPARATIONS AREAS

Figure 3

PLOT OF PLUTONIUM ACTIVITY vs Pu/Cs ACTIVITY RATIO*