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RETENTION OF PLUTONIUM IN WASTE SEEPAGE BEDS AT LOS ALAMOS

By E. R. Hermann

I. Introduction:

With the rapid advances in the production and utilization of radioisotopes over the past ten years, those engaged in preventing disease by controlling the environment have become more and more concerned with the problems attending the release of these carcinogens to the air we breathe, the water we drink, and the earth from which we derive our food. In connection with sanitary studies to determine the behavior of various radioactive substances following their discharge to the general environment, this report deals empirically with the fate of plutonium contained in liquid wastes discharged onto or just below the surface of the earth. This brief study is limited to Los Alamos, New Mexico, and was designed to provide pilot information as to the need for further investigation. The study may best be considered as a preliminary probing.

II. Experimental Procedure:

Two sites were chosen for collection of samples: the seepage bed in back of the TA-21 (DP) "hot" laundry and the tile field in back of Building 35, TA-21. The laundry site was chosen because more was known about the chemical characteristics, concentrations of plutonium, and discharge quantities than at any other site for either burial of solid wastes or discharge of liquid wastes. The tile field near Building 35, TA-21, was chosen because past information indicated that this particular location has probably received more plutonium

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contamination from liquid wastes than any other area.

A. Sampling:

Samples of the sand, gravel, boulders and rock encountered at points designated on Figures 1 and 2 were obtained variously by use of pick and shovel, driven pipe, and an air-motor driven rotary drilling machine. The core barrel available for use with the drilling rig was designed for use in solid rock or concrete. Unfortunately, the earth media encountered in all instances was a conglomeration of soil, sand, and rock; this hindered drilling and yielded only short fragmentary cores or no core at all. Thus nearly all of the samples were obtained by backing the drill out of the hole and scraping the bottom of the hole with an auger or driving a small open pipe into the bottom and then retrieving the plug of earth, sand or gravel. Since samples were desired at one foot intervals and the drill stem could be raised and lowered only very slowly, the above procedure was very time consuming. Removing the drill from the hole to obtain a sample had additional disadvantages in that the bottom of the hole occasionally became contaminated by loose material from near the surface or still worse, the hole simply caved in. Nevertheless, samples were obtained.

B. Analysis:

All samples were analyzed for plutonium employing a modification of the procedure reported by Langham[1] for determining plutonium

in human urine. This involves extraction with cupferron (ammonium nitrosophenyl-hydroxylamine) and chloroform, precipitation with lanthanum fluoride, and counting the alpha emissions in a gas flow scaler. Results are expressed as disintegrations per minute per dry gram.

In addition to the above, four selected samples were sent to the Geochemistry and Petrology Branch, U.S.G.S., Washington, D. C. for mineralogical examination and determination of ion-exchange capacity proceeding as follows: The sample from the 12 foot slant depth of the DPW-3 hole was disaggregated and sieved to remove particles larger than 62 microns in median diameter. The remaining samples were crushed to pass this sieve size. The fine material was dispersed in distilled water with sodium metaphosphate added as a dispersing agent and fractionated at 2 u equivalent settling diameter by repeated centrifuging. The 2 μ (clay) fractions were saturated with Ca^{++} ions by passing the suspensions through a Ca ion exchange column. X-ray diffractometer patterns were made for each sample using both oriented aggregates and mounts in which preferred orientation is minimized. Patterns were also made after glycerol treatment and heat treatment to 400° C. The minerals indicated by the X-ray patterns are listed, the quantitative estimates as parts in ten being derived from the intensities of the diffraction lines. Since many factors, in addition to the quantity of a mineral,

affect diffraction intensity, those estimates are not intended to give more than a general indication of relative amounts of the various minerals present. The ion exchange capacity in milli-equivalents per kilogram (me/kg) was determined by the colorimetric manganese method (Bower and Truog). Sample descriptions and results are presented in Table VI.

C. Results:

Locations of holes with surface elevations are as shown in Figures 1 and 2.

TABLE I

Designation: DPL-0 Date: June 15, 1953

Description: Vertical hole dug by hand in Laundry Waste Seepage Bed. Bed is about six feet deep and consists of ungraded sand and gravel mixed with river rock ranging in size from one to fifteen inches. In addition to the earth samples, three different types of plant life growing in the vicinity were sampled.

Depth below Surface	Description	Plutonium dpm/dry gram
Surface	Loam (laundry waste residue)	26,000
3"	Sandy soil	360
5"	Sandy soil	74
16"	Sand, gravel, and river rocks	28
31"	" " " " "	20
40"	" " " " "	40
53"	" " " " "	24
55"	" " " " "	8
53"	" " " " "	120
Surface	Stagnant pond bacterial slimes	12,300
Surface	Weeds growing in pond	22
Surface	Algal slimes growing in clear waste	5,080

TABLE II

Designation: DPL-1 and DPL-2 Date: August 24, 1953

Description: Vertical machine bored trial holes outside of Laundry Waste Seepage Bed near rim of Los Alamos Canyon.

Depth below Surface	Description	Plutonium dpm/dry gram
	DPL-1	
Surface	Sandy soil	10
9' - 12'	Solid tuff core	6
	DPL-2	
Surface	Sandy soil	6
10'	Core of broken tuff	6
12'	Core of broken tuff	6
13'	" " " "	6
15'	" " " "	6

TABLE III

Designation: DPW-1 and DPW-2 Date: August 25-27, 1953

Description: Vertical holes drilled in earth fill berm between tile fields in back of Building 35, TA-21.

Depth below Surface	Description	Plutonium dpm/dry gram
	DPW-1	
Surface	Very fine sandy soil	70
1'	Very fine sandy soil	8
2'	Very sandy soil	4
3'	Sandy soil	8
4'	Coarse sand and clay	4
5'	Sand	4
6'	Sand	4
6' - 10'	Sand	2
10' - 14'	Sand	2
15'	Sand	4
	DPW-2	
Surface	Very fine sandy soil	9
1'	Very fine sandy soil	4
2'	Sandy soil	3
3'	Sandy soil	1
4'	Sand	2
5'	Sand	3
6'	Sand	2
7'	Sand and clay soil	4
8'	Sand	1
9'	Sand	3
10'	Very coarse sand	4
11'	Fine sand, some gravel	4
12'	Fine sand	3
13'	Fine sand	3
14'	Sand	3
15'	Sand	2
16'	Sand	4
17'	Sand	3
18'	Sand	2
19'	Sand	3
20'	Sand	3

TABLE IV

Designation: DPA-3 Date: August 28, 1953

Description: Hole drilled on a 45° slant starting on berm and extending under adjacent tile field near Building 35, TA-21.

Depth below Surface	Description	Plutonium dpm/dry gram
Surface	Very fine sandy soil	32
1'	Very fine sandy soil	5
2'	Very fine sandy soil	9
3'	Very fine sandy soil	7
4'	Sandy soil	8
5'	Sand	6
6'	Very sandy soil	4
7'	Very sandy soil	7
8'	Very sandy soil	3
9'	Sand and clay	3
10'	Fine sand	2
11'	Sand	2
12'	Loose tufaceous sand	450 *
12.5'	Loose tufaceous sand	1510
13'	Loose tufaceous sand	1330

*Point of intersection with tile field.

TABLE V

Designation: DPW-4 and DPW-5

Date: September 21, 24, 25, 30 & October 1, 1953

Description: Vertical holes drilled directly through the tile fields in back of Building 35, TA-21.

Depth below Surface	Description	Plutonium dpm/dry gram
	DPW-4	
Surface	Sandy soil	8
1'	Sand and gravel, some clay	400
2'	Sand and gravel, some clay	36,100
3'	Sand and gravel, some clay	45,600
12'	Very fine loose tuff	1,400
15'	Fine sand	5,000
16'	Fine sand	5,100
17'	Loose tuff	720
18'	Sandy loose tuff	24
19'	Loose tuff	12
20'	Broken tuff core	12
	DPW-5	
Surface	Sand and clay soil	410
1'	Sand and clay soil	600
2'	Sand and clay soil	10
3'	Sand and gravel	80
4'	Sand and gravel	3,400
5'	Solid tuff core from a boulder	530
6'	Solid tuff core from a boulder	80
7'	Friable tuff core	1,800
8'	Fine sand and clay	40
9'	Fine sand and clay	380
15'	Fine sand	2,400

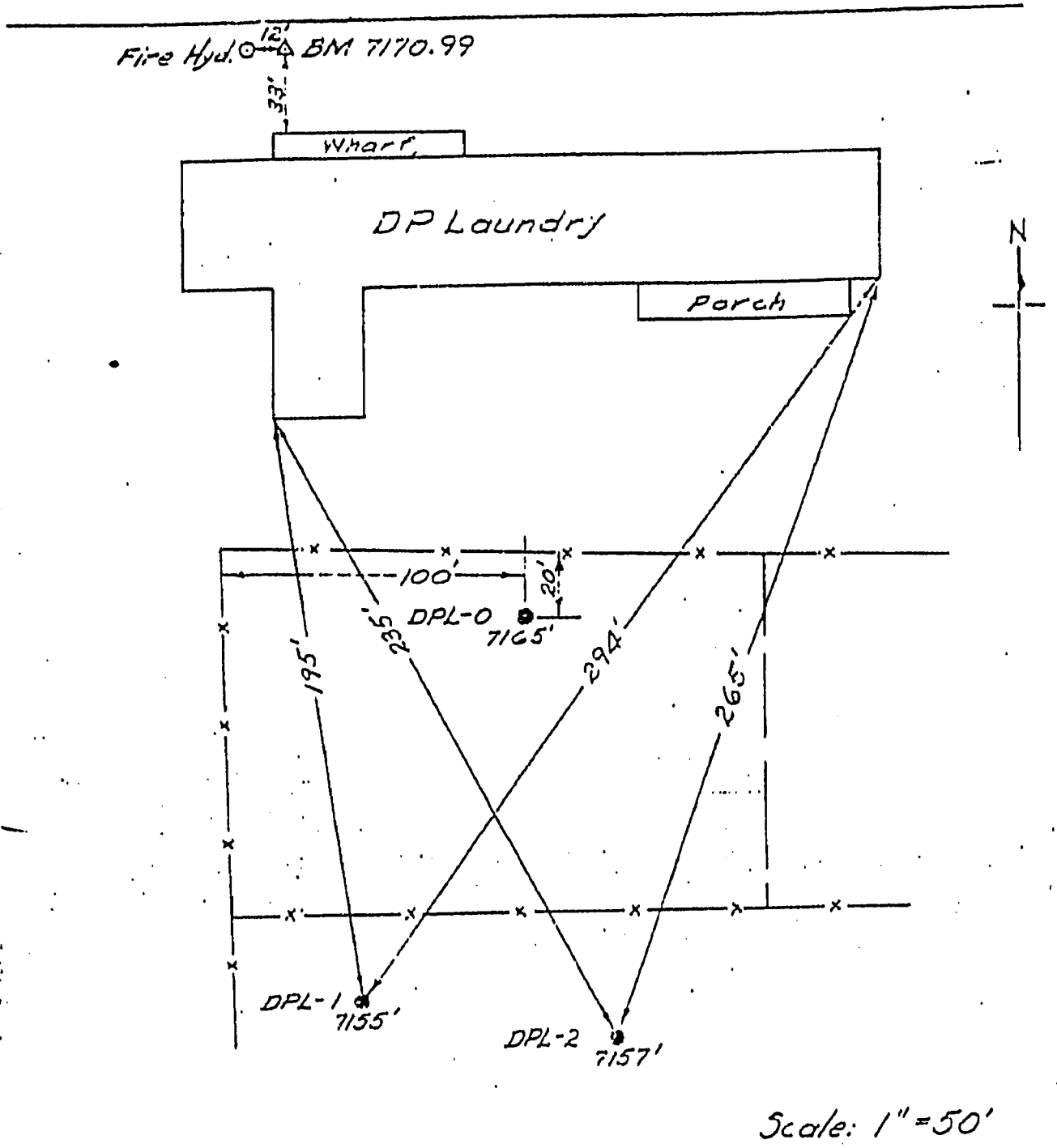
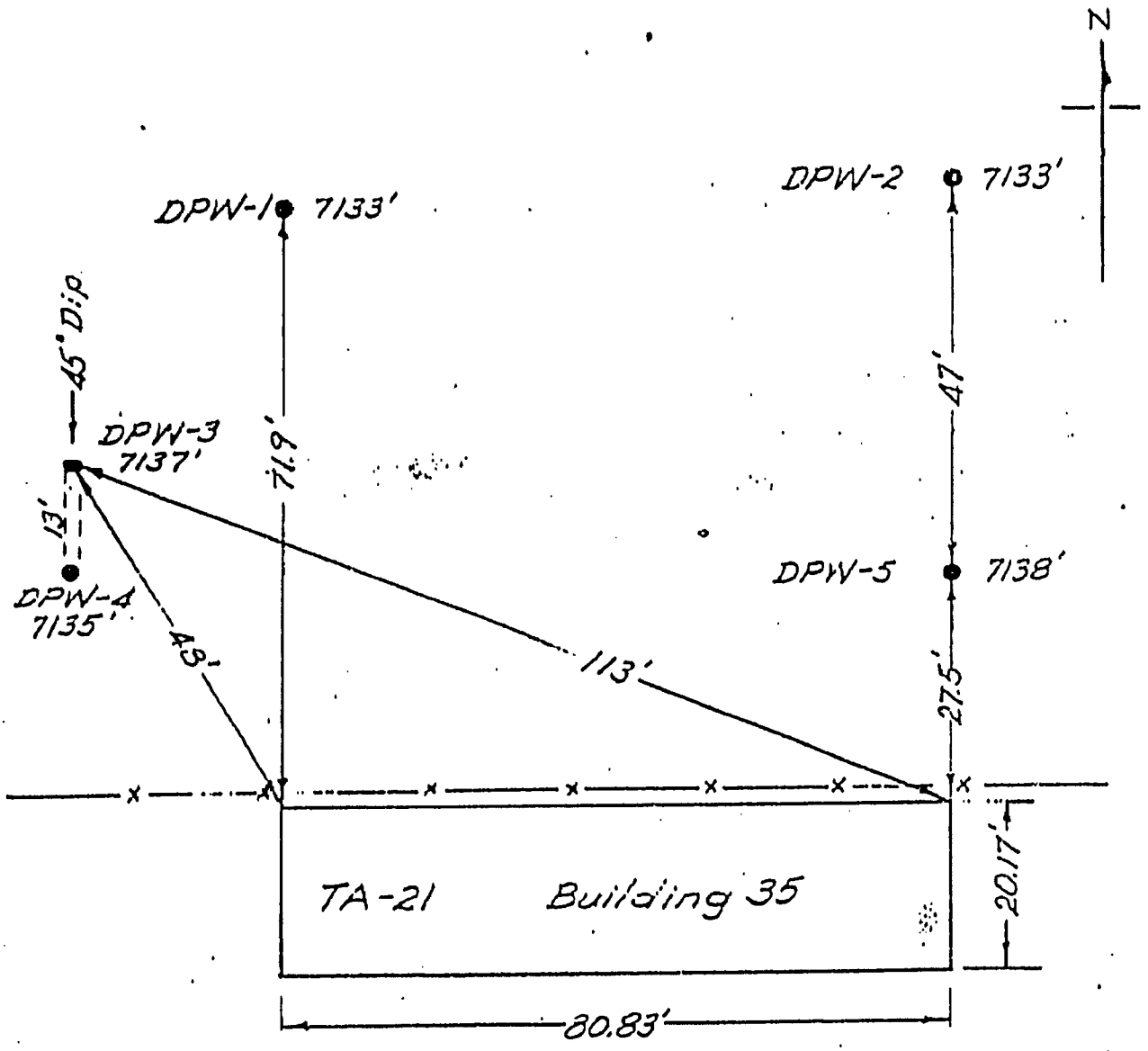


Figure 1. Location of Sampling Points DP Laundry



Scale: 1" = 20'

Figure 2. Location of Sampling Points DP West Tile Field.

TABLE VI

Sample Depth	Megascopic Description	Minerals Present (parts in ten)			Ion Exchange Capacity (me/kg)
DPL-1 from 9-12 ft.	Gray slightly pumiceous tuff typical of Tshirege member of Bandelier tuff.	Clay <2 μ	Feldspar	5	12
			Cristobalite	3	
			Tridymite	1	
			Quartz		
			Montmorillonite		
		Silt 2-62 μ	Feldspar	5	
			Cristobalite	3	
			Tridymite	2	
			Quartz		
DPW-3 at 12 ft. (slant)	Sandy to silty clay and gravel composed of gray pumiceous Bandelier tuff (probably Tshirege member). Due to 45° dip of hole, vertical depth of sample is 8.5 ft.	Clay <2 μ	Feldspar	4	17
			Montmorillonite	3	
			Hydrous mica	2	
			Kaolin		
			Cristobalite		
			Tridymite		
			Quartz		
		Silt 2-62 μ	Feldspar	4	
			Cristobalite	4	
			Tridymite	1	
			Quartz		

TABLE VI (CONT'D)

Sample Depth	Megascopic Description	Minerals Present (parts in ten)			Ion Exchange Capacity (me/kg)
DFW-4 at 19 ft.	Gray tuff of Tshirege member of Bandelier tuff. Contains some bit-broken fine particles.	Clay <2 μ	Feldspar	4	7
			Cristobalite	4	
			Tridymite	-	
			Quartz		
		Silt 2-62 μ	Feldspar	5	
			Cristobalite	4	
			Tridymite	1	
			Quartz		
DFW-5 at 5 ft.	Dark gray to white pumiceous tuff. Apparently cored from a boulder	Clay <2 μ	Feldspar	5	32
			Montmorillonite	2	
			Cristobalite		
		Silt 2-62 μ	Feldspar		
			Tridymite		
			Cristobalite		

III. Discussion:

Despite the shortcomings of the sampling techniques, the patterns of plutonium concentrations from the various borings were reasonably good and each pattern may be considered as representative of the particular sample hole. Furthermore, certain characteristics concerning the travel of plutonium through the earth, sand, gravel and rock media are indicated. These are:

1. No appreciable horizontal movement of the plutonium occurs in the first 20 feet of depth.
2. The plutonium is readily retained by the various earth media (sand, clay, gravel and rock).
3. Apparently retention of the plutonium is greater in the finer materials.
4. Penetration of the plutonium into the underlying strata is not to be expected.

The foregoing information, when coupled with Theis' estimate that the average ground water velocity in the formations underlying Los Alamos does not exceed 305 feet per year[2], greatly limits the possibility that significant contamination of the main aquifer with plutonium from liquid or solid industrial wastes will ever become a reality under present disposal practices.

In regard to the mechanism of retention, a crude projection of the ion exchange capacities (Table VI) to a full scale tile field containing

approximately 20,000 m^3 of soil, such as at TA-21, and a minimum estimate of the waste discharges to this tile field since 1943 indicates that even under ideal conditions the ion exchange capacity of the soil falls short of accounting for the retention phenomena. Instead it is probable that retention of plutonium by these particular local soils is essentially due to adsorption, while the effect of ion exchange is of secondary importance.

IV. Conclusions:

1. Monitoring of the Los Alamos and Guaje Canyon water supply wells and of the distribution system should be continued, since there are many other modes of contamination of a water supply system in addition to movement through the earth.
2. Routine determinations of plutonium and fluoride concentrations are essential.
3. The sampling frequency of each well and at selected distribution points should be about four times per year with confirmative testing of any source that shows an apparent rise in plutonium or fluoride concentration.
4. Extensive soil sampling of contaminated seepage beds, tile fields, and burial grounds with a view toward determining the burden of plutonium or other radioisotopes existing in the particular area is too expensive to be feasible.
5. Further laboratory bench scale work is desirable for determining the capacities of native rocks and soils for adsorption, ion exchange and filtration of plutonium 239 and other troublesome radioisotopes such as strontium 89 and strontium 90.

V. References:

1. Langham, Wright H., "Determination of Plutonium in Human Urine", MDDC-1555, U. S. Atomic Energy Commission.

2. Theis, C. V., "Geologic Background of Waste and Water-Supply Problems at Los Alamos". TID-460, Meeting of AEC Waste Processing Committee at Los Alamos, New Mexico -- U. S. Atomic Energy Commission -- October 1950.