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FIXATION OF FISSION PRODUCT ACTIVITY

BY SOILS OF HAMFORD AREA

by

R. Overstreet and L. Jacobson

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FIXATION OF FP ACTIVITY BY SOILS OF W ARLA

by

R. Overstreet and L. Jacobson

I. INTRODUCTION:

On August 9-12, 1944, a cursory inspection of the soils in the region of site W was carried out for the purpose of determining the behavior of this material towards water solutions containing radioactive elements.

A detailed soil survey of this region was carried out by the U. S. Department of Agriculture, Bureau of Soils in 1916*. The area is underlain by a succession of basaltic sheets of the Columbia River lava field. These basaltic sheets, known as Yakima basalt, have, in this area, an estimated thickness of more than 2500 feet. Overlying the basalt in various parts of this region are sedimentary beds which have been laid down in horizontal layers in former lakes. This material is known as the Ellensburg formation and consists for the most part of stratified, light-colored deposits of very fine sand, silt, and clay. The region is also characterized by areas of wind-blown and stream-laid materials. The soils in the site areas are generally loose and open, and of low moisture retaining capacities. They are uniformly of rather coarse texture. In some regions there are underlying layers of calcareous deposits, and frequently there are small fragments of calcareous material near the surface.

II. DETERMINATIONS:

Samples were taken from the tank-farm excavations 200 E, 200 W-T, and 200 W-U. Three samples were taken from each excavation at an approximate depth of 20 to 30 feet. Surface samples were taken in the neighborhood (a few hundred feet away) of 200 E and 200 W-T. Sand from the Columbia River and topsoil adjacent to the river were taken near area 100 F. The approximate locations of the various areas are given in the accompanying map.

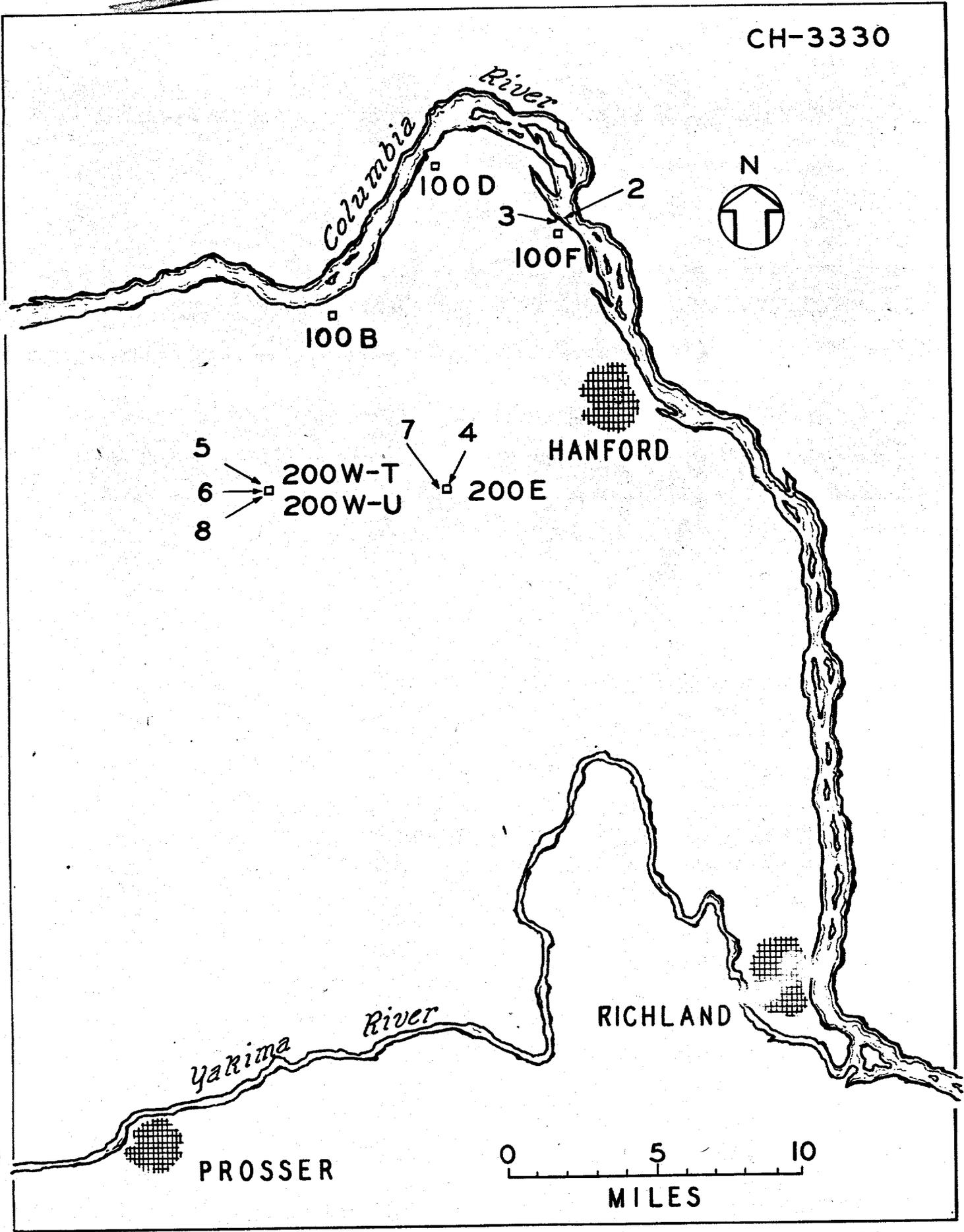
The samples were shipped to Berkeley where portions of them were passed through a 2 mm. screen. This procedure served to separate out the very coarse fraction which, in the experience of soil chemists, contributes very little to the chemical and water-holding properties of the soil. However, for certain extrapolations to field conditions, the fraction of particles larger than 2 mm. was determined for each sample. (See Table I)

* Field Operations of the Bureau of Soils, 1916, 13th report, page 2203.

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TABLE I

DESCRIPTION	PERCENT LESS THAN 2 MM. DIAMETER
Clinton top soil	100.0
Sand from Columbia river	100.0
Top soil near Columbia river	100.0
200 E tank-farm, south face	93.2
200 E tank-farm, east face	74.5
200 E tank-farm, north face	61.5
200 W-T tank-farm, west (1) face	39.0
200 W-T tank-farm, west (2) face	28.8
200 W-T tank-farm, south face	88.9
200 W-U tank-farm, west face	100.0
200 W-U tank-farm, south face	38.2
200 W-U tank-farm, east face	43.6
200 E top soil, 500 ft. east of tank-farm	87.5
200 W-T top soil, 100 ft. west of tank-farm	100.0

A study was then carried out with the samples, so prepared, to determine their ability for absorbing radioactive elements from W-6 waste liquor previously obtained from Clinton. An assay of the W-6 liquor made on June 30, 1944 gave the following composition of radioactive elements (values corrected to zero thickness):

Ba + Sr	.27%
Zr	58.4 %
Cb	25.6 %
Ce	4.75%
Y + Pr	.22%
Ru	1.69%
Te	4.29%
Cs	1.39%
UX ₁	3.27%

The solution also contains macro-amounts of Na⁺, K⁺, UO₂⁺⁺, NO₃⁻, SO₄⁻, PO₄⁻, F⁻, and OH⁻.

Samples of each soil were shaken with four dilutions of the W-6 solution. Composite samples for the tank-farm areas were prepared by combining equal weights of the <2 mm. fraction of the three samples taken in each area. In each case, 10 grams of soil was added to 100 ml. of appropriate dilution and allowed to stand for three days with frequent shaking. Following the absorption period, the soil was centrifuged out, and an aliquot of the supernatant liquid was measured for activity. The results are given in Table II. In addition to the Hanford soils, a sample of topsoil from Clinton** (taken near the site) is included as

** A detailed survey of the soils of the Clinton area has been carried out by U. S. Dept. Of Agric., Bureau of Plant Industry in 1936. See Series 1936, No. 15

well as a sample of quartz sand.

TABLE II

SAMPLE #	DESCRIPTION OF SAMPLE	% ACTIVITY ABSORBED FROM W-6 SOLUTION				BASE EXCHANGE CAPACITY
		UNDIL.	1:10	1:100	1:1000	ME/100 GMS.
1	Clinton topsoil	75	94	96	96	14.
2	Sand from Columbia river	37	85	75	76	2.8
3	Topsoil near Columbia river	53	59	66	76	4.2
4	Composite sample, 200 E tank-farm	25	77	93	79	3.6
5	Composite sample, 200 W-T tank-farm	40	87	93	81	4.9
6	Composite sample, 200 W-U tank-farm	54	90	94	75	6.3
7	Topsoil near 200 E tank-farm	59	91	94	85	5.3
8	Topsoil near 200 W-T tank-farm	49	73	80	74	5.6
9	Quartz sand	0	2	29	24	<1.
10	Blank	0	0	0	0	—

Included also in Table II are the base-exchange capacities of the samples studied. The base-exchange capacity is a measure of the amount of exchangeable adsorbed cations on the soil. The value is usually expressed as milli-equivalents per 100 grams of soil. Thus, a base-exchange capacity of 50 denotes that 100 grams of material contains 50 milli-equivalents of exchangeable adsorbed cations.

The contact of solutions of fission products with soil necessarily involves the water-relationships of the soil. For this reason, the moisture equivalent was determined for each sample. The moisture equivalent is the amount of water held by the soil under a centrifugal force of 1000 times gravity. It presumably corresponds to the water of hydration of the particles plus the water held in the smaller capillaries. It is expressed as percent water on the oven-dry basis. For example, a moisture equivalent of 10 means that 100 grams of the oven-dry soil after being saturated with water will retain 10 grams of water against a centrifugal force of 1000 times gravity. The values are presented in Table III.

TABLE III

SAMPLE #	DESCRIPTION	MOISTURE EQUIVALENT	
		< 2 MM. FRACTION	TOTAL SAMPLE
1	Clinton topsoil	31.7	31.7
2	Sand from Columbia river	3.27	3.27
3	Topsoil near Columbia river	7.97	7.97
4	Composite sample, 200 E tank-farm	2.36	1.75
5	Composite sample, 200 W-T tank-farm	5.72	2.39
6	Composite sample, 200 W-U tank-farm	5.16	3.08
7	Topsoil near 200 E tank-farm	7.54	6.59
8	Topsoil near 200 W-T tank-farm	5.36	5.36

The above table contains the results for the < 2 mm. fraction and the calculated results for the total sample. The calculations for the total sample are based on the general observation that the fraction > 2 mm. does not contribute significantly to the water-holding capacity of soils.

III. DISCUSSION:

Radioactive elements when applied to soil in aqueous solutions may be immobilized in two ways. First, the radioelements may become fixed on the soil particles. Second, the entire solution may be immobilized in the fine capillaries and as water of hydration of the soil particles.

The fixation of the elements on soil particles may occur by a process of exchange adsorption in which the radio-active ions exchange for an equivalent number of ions (i.e., 1 Sr^{++} exchanged for 2 Na^+) initially present in the exchange complex of the soil. Also, the addition of the aqueous solution to soil may result in the precipitation of the radioactive elements as the oxides or as inorganic salts which subsequently are fixed on the soil particles by a process of mutual flocculation. In the present study, no attempt has been made to determine the nature of the fixation.

In order to ascertain the effect of concentration of ions, such as Na, K, and CO_3^{--} , etc., which are present in W-6 solution in macro-amounts, on the fixation of the radioactive elements on soil, a given weight of soil was treated with various dilutions of the W-6 solution. Examination of Table II shows appreciable absorption of active elements at four concentrations of W-6 by all soils with the exception of the Ottawa sand. In all cases there is a marked effect of dilution on the absorption; the maximum absorption occurs in the neighborhood of 1:100 dilution. Although the results show decrease in absorption at the 1:1000 dilution, the results at this concentration are subject to considerable error. In order to obtain a 1:1000 dilution in an overall volume of 100 ml., 0.1 ml. of the W-6 stock solution was used. As a result, the total activity dealt with was quite low. Moreover, the low concentration of electrolytes at this dilution resulted in the dispersion of the soil colloids, rendering them difficult to remove by centrifugation.

All dilutions were made with distilled water. An examination of the blank in Table II reveals that the dilution of W-6 liquor with distilled water per se does not result in the precipitation of activity.

There is a rough correlation between the exchange capacity of the soils and their absorption of activity, particularly at the lower dilutions of W-6. The Clinton topsoil, with an exchange capacity of 14.0, is the best absorber of activity. On the other hand, the Quartz sand, with an exchange capacity of $<< 1$, is the poorest absorber. The Hanford soils, with exchange capacities of 2.8-6.3 are fair absorbers at low dilutions and quite good absorbers at 1:100 dilution. The base exchange capacity is often a measure of the clay content of the soil (particles < 1 micron in diameter) since, as a rule, the larger particles contribute very little to the exchange complex. This was quite evident in the

samples examined. The Clinton topsoil was quite clayey, the Hanford soils had a small amount of clay, and the Quartz sand had no noticeable clay content.

The fixation of aqueous solutions in fine capillaries and as water of hydration of the soil particles has been mentioned as another means of immobilization of radioactive solutions. This process would be particularly important in the fixation of those radioelements which behave as anions and hence do not undergo exchange adsorption on soils. When a given amount of water is added to a large amount of dry soil, the water will advance into the soil in a front. Beyond the front the soil will be obviously dry and behind the front the soil will be obviously wet. The advance of the water will continue until the wet soil behind the front has reached a uniform and characteristic water content. This characteristic water content is known as the field capacity of the soil. Water in soils at or below their field capacities moves extremely slowly, either under the influence of capillary forces or under gravitational pull. For example, if a soil that is wet to the field capacity is placed in contact with an air-dry soil, there will be over a period of many months but a very small change in the distribution of water in any direction. This is due largely to the fact that the capillary conductivity of dry soils is practically zero since there is no continuity of moisture films. Distillation from the wet to the dry soil is an extremely slow process and need not be considered. For medium-textured soils, the field capacity is approximately equal to the moisture equivalent. For coarse textured soils, such as those from Hanford, the field capacity tends to be higher than the moisture equivalent. For this reason, the moisture equivalents in Table III may be considered as minimum values for the water holding capacities of these soils. The amount of water so held may be quite appreciable, even for soils of low moisture equivalent. For example, the dry composite soil from the tank farm 200 A-T will immobilize 10 gallons of solution per cubic yard.

The rate of downward movement of water through soils is related to the amount and continuity of non-capillary pores, to the hydration of the pores, and to the resistance of entrapped air. In general, the rate of movement is greater in a wet soil than in a dry soil. Also, the rate is greater in sandy soils than in clayey soils. Thus, even though the Clinton topsoil has a high water holding capacity, the rate of water movement would be slow owing to its high clay content.

If a solution is added to a soil which is at or above its field capacity, the solution will move into the soil, displacing, but not mixing with the soil water. The diffusion of salts in soils occurs at a very slow rate.

In conclusion, our experiments show that the Hanford soils have a marked ability for the absorption of radioelements from W-6 solution. Moreover, it is probable that those elements which are not absorbed by the particles may nonetheless be fixed for considerable periods of time due to the property of the soils of immobilizing large amounts of aqueous solutions.