Characterization of the Hanford 300 Area Burial Grounds

Task II - Geochemical Analysis

April 1979

**Prepared for the U.S. Department of Energy
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**Pacific Northwest Laboratory
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CHARACTERIZATION OF THE HANFORD 300 AREA
BURIAL GROUNDS

TASK II - GEOCHEMICAL ANALYSIS

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April 1979

Prepared for the U.S. Department of Energy
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PREFACE

The United States Department of Energy (DOE) is considering decontamination and decommissioning alternatives for its retired nuclear facilities in order to provide adequate protection from radioactive and other hazardous materials potentially originating from such sources. Decontamination and decommissioning of these facilities may involve the selective removal of radioactive or other materials from retired nuclear facilities and selection of the ultimate disposition of the facilities. Among the facilities being evaluated are retired radioactive waste burial grounds at DOE's Hanford Site, Richland, Washington.

The "Characterization of the Hanford 300 Area Burial Grounds" project was initiated to provide an assessment of retired radioactive waste burial grounds prior to selection of decontamination and decommissioning alternatives at the Hanford Site. The project was sponsored by the U.S. Department of Energy, Division of Environmental Control Technology and was conducted by Pacific Northwest Laboratory from 1975 through 1978.

The original intent of this project was to develop technology and methods for characterization of waste burial sites and to apply them to the 300 Area Burial Grounds. During the course of the study, the scope was broadened to include development of recommendations pertaining to buried waste disposition alternatives.

Project tasks were initiated to provide the necessary technology and to characterize burial ground sites. These tasks included:

Task I - Geophysical Evaluation

Task II - Geochemical Analysis

Task III - Fluid Transport Characterization and Modeling

Task IV - Biological Transport

Topical reports from each of these tasks have been issued under separate cover.

Two other reports have been issued: "Decontamination and Decommissioning" provides an overview of the 300 Area Burial Grounds, project results and recommendations. "Decontamination and Decommissioning Regulatory Issues" details regulatory issues to be considered when evaluating the alternatives.

SUMMARY

The geochemical interaction of radionuclides with geologic materials must be understood in order to assess radionuclide migration from waste burial structures. In Task II, Geochemical Analysis, soil samples were taken from a representative disposal site to provide information that could be applied to the retired 300 Area Burial Grounds on the Department of Energy's Hanford Site, Washington. The site chosen for evaluation was the 316-4 Crib, which received liquid waste containing hexone and a total of 939 kg of uranium between 1948 and 1956.

The methodology developed to analyze the soil samples combined neutron-enhanced autoradiography, X-ray fluorescence, and electron microprobe and radiochemical data. Results showed that the uranium content of the soil immediately underlying the crib is small. Furthermore, the uranium in the 316-4 Crib generally formed an insoluble phosphorus compound that was not readily leached under ambient conditions. However, uranium was easily leached in dilute acid solutions. Consequently, acidic solutions should not be added to waste burial structures that contain uranium compounds.

The buffering and cation exchange capacity of the soil clay fraction tends to retard migration of the water-soluble forms, and uranium from the 316-4 Crib has not been detected in the Columbia River. No significant health hazard is apparent at present as a result of uranium disposal at the 316-4 Crib; however, efforts should continue to restrict access to this site.

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INTRODUCTION

Decontamination and decommissioning plans and activities concerning retired nuclear facilities are being established on the Hanford Site. These activities will assign nuclear waste disposal facilities to specific use categories. These categories include: 1) restricted use, i.e., restricted for nuclear activities only; 2) conditional use, i.e., public use above ground, waste stabilization below ground; and 3) unconditional use, i.e., public use of the site.

In order to select a specific use category or any combinations of use categories, the relative safety of a waste burial disposal site must be assessed. To assess the safety of a site, evaluations of many factors must be considered, including analyses of the nuclear waste and the matrix within which it is contained. This report summarizes the application of a systematic methodology developed to evaluate uranium-contaminated radioactive waste and its interactions with sedimentary materials. The methodology combines neutron-enhanced autoradiography, X-ray fluorescence, and electron microprobe and radiochemical analyses. This methodology was applied to a specific radioactive waste disposal structure on the Hanford Site. The utility of this report will be to provide geochemists and other nuclear waste management experts an applied methodology to investigate phenomena relative to migration of uranium from structures. The information derived from this investigation will aid in development of decontamination and decommissioning recommendations for a specific disposal facility and those similar facilities containing the same or closely related materials.

As part of this study, cores of soils underlying the 316-4 Crib, located approximately 8 km north-northwest of the 300 Area (Figure 1), were collected for analytical evaluation. The 316-4 Crib (Figure 2) consists of a buried, bottomless, stainless steel tank with discharge and ventilation pipes leading to the surface. Liquid waste containing hexone (isopropylacetone) and a total of 939 kg of uranium were discharged to the crib from the 326 Building between 1948 and 1956 (Heid, 1956). Other solution constituents and their concentrations in the wastes are largely unknown.

FIGURE 1. Location of the 316-4 Crib Relative to Regional and Local Boundaries

Ground Surface and Physical Structures at the 316-4 Crib Site. The 300 North Burial Ground is in the background. The foreground contains 4 monuments marking the approximate outer dimensions of the disposal facility and internal to these monuments are discharge and ventilation pipe risers.

Generalized Illustration of a Bottomless Disposal Structure Located at the 316-4 Crib Site (Cross-Sectional View)

FIGURE 2.

The 316-4 disposal site was evaluated because of the reported occurrence of uranium in groundwater wells that monitor the disposal site. In addition, uranium in the form of aqueous uranyl is considered to be the predominant (volume basis) radioactive waste disposed to all 300 Area disposal facilities (Table 1). Thus, analysis of waste media interactions at this site should be generally applicable to those at other 300 Area waste disposal facilities. The activity (concentrations) for alpha emitters (primarily uranium) in the water from several wells sampled during January, February and March 1955 is given in Table 2a. Although the alpha concentrations are very low, it is apparent that some alpha, most likely uranium, migrated to the ground water during disposal operations. According to Heid (1956), the southeastward movement of the alpha emitters (uranium) in the ground water from the site of Well 321-1 toward Wells 5, 6, 7, 8 and 9 averaged about 8 m/month. Wells 2, 4 and 9 show activity concentrations comparable to those reported as background concentrations.

The flow of ground water from this waste disposal structure within the saturated groundwater system is to the southeast where the system terminates at the Columbia River. The travel time for water between the 316-4 Crib and the Columbia River is approximately 42 years at a rate of about 100 m/yr.

Uranium concentration in the ground water determined by the Pacific Northwest Laboratory Routine Monitoring Program has been periodically monitored between 1969 and the present. The occurrence of uranium in Wells 2, 3, and 5 is reported in Table 2b. The concentration of uranium in the ground water at these wells has significantly decreased since initial monitoring in 1955; however, one result, i.e., Well 321-5 where uranium concentration exceeds 3.0×10^{-9} Ci/cm³, suggests residual uranium concentration in ground water at this waste disposal site. The concentration of uranium, if present, in other groundwater monitoring wells down gradient from the 316-4 Crib near the Columbia River is below detection limits, suggesting that no migration of uranium from the above site to the biosphere has occurred.

TABLE 1. Solid, Liquid, and Mixed Waste From Contaminated Radioactive Waste Burial Facilities, Hanford 300 Area

	<u>Predominant Radionuclides and Waste Form</u>		
	<u>Solid</u>	<u>Liquid</u>	<u>Mixed Solid and Liquid</u>
300 No. 1	U, Pu and Fission Products		
300 No. 2			U, Pu and Fission Products
300 No. 3			U
300 No. 4			U
300 No. 5			U
300 No. 7		U,Th	
300 West		U	
300 North			U, Pu and Fission Products
300 WYE			U, Pu and Fission Products

TABLE 2a. Alpha Emitter (Uranium) Activities and Nonradioactive Ion Concentrations in Water from Wells Around the 316-4 Crib Site in 1955 (Brown et al., 1955)

<u>Well No.</u>	<u>Alpha Activity, x 10⁻⁹</u>		<u>Ci/cm³</u>	<u>Na⁺, ppm</u>	<u>NO₃, ppm</u>
	<u>January</u>	<u>February</u>	<u>March</u>	<u>March</u>	<u>March</u>
321-2	2.7	3.2	2.7	20	2.8
321-4	2.9	4.7	2.0	22	1.4
321-5	24.3	30.4	29.7	27	39.4
321-6	37.1	36.2	34.2	25.5	20.7
321-7	3.4	3.8	2.5	25	30.3
321-8	6.1	9.0	6.3	26.5	32.9
321-9	1.8	1.0	1.1	26	3.4

TABLE 2b. Alpha Emitter (Uranium) Activities in Water from Wells Around the 316-4 Crib Site

<u>Well No.</u>	<u>Date Sampled</u>	<u>Alpha Activity</u> <u>10^{-12} Ci/cm³</u>
321-2	6/30/75	9.6
"	10/27/75	11.0
"	12/29/75	<3.4
"	4/26/76	<3.4
"	6/28/76	<3.4
"	9/27/76	19.0
"	1/4/76	<3.4
"	3/28/77	<3.4
321-4	9/27/76	16.0
"	1/4/77	<3.4
"	3/2/77	<3.4
"	3/28/77	<3.4
321-5	6/15/71	3425.4

CONCLUSIONS

A methodology has been developed to qualitatively and quantitatively assess the geochemical interactions of uranium and natural geologic materials at buried waste disposal sites. This methodology has been applied to an existing waste disposal site for demonstration purposes. As a result of the application, the following conclusions have been derived.

- No significant nuclear health hazard has been perpetuated by disposal of uranium at the 316-4 Crib. No significant nuclear health hazard is expected from buried waste containing uranium in the Hanford 300 Area Burial Grounds if it were to enter the saturated groundwater system under similar conditions as those existing at the 316-4 Crib.
- Uranium from the 316-4 Crib has not reached the Columbia River in detectable quantities.
- Uranium disposed to Hanford 300 Area Burial Grounds is somewhat leachable by water and organic solvents; however, this material is readily leachable in dilute acid solutions. Acidic solutions, therefore, should not be introduced into retired waste burial structures containing uranium compounds.
- Uranium in the 316-4 Crib typically forms a rather insoluble compound of the form $(\text{UO}_2)_2 [(\text{H}_2\text{PO}_4)_2(\text{HPO}_4)]$ minimally leachable under ambient conditions.
- Radiological exposure based on preliminary field radiological surveys within the 316-4 Crib may exceed permissible values and, as such, an effort should be expended to reduce the access to this structure by inadvertent intruders.

METHODS OF INVESTIGATION

Soil cores were recovered from below the 316-4 Crib with a hand-operated auger apparatus. Access into the crib was gained through a 10-cm stainless steel riser through which approximately 5 m of auger drill string was lowered. Approximately 1.5 m of soil were collected to a depth of 75 cm from the bottom of the crib. The three cores had dimensions 25 cm in length by 5 cm in diameter. Core 1 was from the surface of the crib floor to 25 cm beneath it; Core 2 was from 25 to 50 cm; and Core 3 from 50 to 75 cm below the crib floor. The cores retrieved from beneath the crib were placed in polyethylene containers for transport to analytical laboratories. Several surface counts were taken in the field with survey instruments. The average count rate was 800 with a maximum of greater than 1000 counts per minute. The material collected typically consisted of unconsolidated pebbles (16 mm mean grain size) bimodally distributed with very coarse sand (1.5 mm mean grain size).

Soil core samples were dried at 105°C and vacuum impregnated with an epoxy resin. Following resin impregnation and heat curing, the mounted samples were cut across the sediment grains to give a cross-sectional surface area that contained uranium contamination. The surface was ground, polished and cleaned of any uranium that may have become smeared during these operations. Optical photomicrographs were taken at several magnifications of the surface features with ordinary and plane polarized, reflected light.

Following the optical photography, a Lexan^(a) sheet was attached to the surface of the mount and the mount sealed into a canister. The canister was then subjected to a high neutron flux ($\sim 1 \times 10^{15}$ n/cm²/sec) from an operating nuclear reactor. After 15 min of neutron irradiation, the canister was removed, and the mount was recovered after another 15 min to allow neutron-induced radioactivity in the sample to decay to a reasonable level.

(a) Trade names are included as common usage for the convenience of the reader and do not imply endorsement by DOE or PNL.

The Lexan was removed and developed with dilute sodium hydroxide etching to give a uranium autoradiograph. Optical photographs of the developed Lexan show the location of uranium particles that fissioned in the high neutron flux in proportion to their concentration in the sample. Matching optical photographs of the same areas of the original surface of the mount and the autoradiograph allow a qualitative estimate of uranium concentrations and their locations. The technique is referred to as neutron-enhanced autoradiography, and allows relatively rapid autoradiographs of low uranium concentrations that would require many months by the firm contact technique.

Following the autoradiography and location of high uranium concentrations in the mounts, certain high concentration uranium particles in one of the mounts were extracted under a binocular microscope and the particles mounted on a glass capillary for X-ray diffraction analysis. A small particle technique allowed X-ray identification of particles down to 2 μm in diameter.

Other sample mounts were vacuum-coated with gold and used in electron microprobe and scanning electron microscope examination. The microprobe was used to obtain chemical analyses of uranium and other constituents down to 1 μm in size. Another use of the microprobe was to qualitatively determine the spatial distribution of elements present in a specific area. This in turn permitted identification of rock types and mineral phases. An element such as silicon is excited by electron bombardment and the characteristic X-rays emitted are superimposed at the same scale as the original photograph of the area. As a result, the silicon locations and its qualitative concentrations become known. Other elements spatially determined included aluminum, calcium, potassium, iron, phosphorus and uranium. The above investigative techniques are similar to those used in a study of plutonium in the soil of 216-Z-9 covered trench (Ames, 1974; Price and Ames, 1975; Ames, 1977).

RESULTS

Samples of the recovered soil were leached of their uranium content with a boiling nitric acid solution. Fluorometric analysis of the acid solution showed that the soil samples typically contained 100 μg uranium per g of soil, i.e., ~ 0.01 wt% uranium bearing soil. Thus, the overall uranium content of the soils immediately underlying the 316-4 Crib is small. The following text gives the results of the autoradiography for each soil mount, along with the electron microprobe data and X-ray identification.

SOIL MOUNT 1A

A low magnification view of a portion of Soil Mount 1A, with a matching autoradiograph, is given in Figure 3. Magnified areas are outlined in black squares and labeled A, A', C and D. Two large pebbles are shown in Figure 3 surrounded by smaller fragments cemented on their exterior surfaces. The small amount of radioactivity on the interior of the pebbles was probably translocated there during cutting, grinding and polishing of the epoxy-impregnated mount. A bubble in the epoxy also can be seen in the optical micrograph but is missing in the autoradiograph. Reflected light outlines the bubble in the optical photo, but the lack of uranium content causes it to disappear in the autoradiograph. All four areas examined on the electron microprobe are located in the smaller fragments cemented on the pebble exteriors.

In area A (Figure 4), the uranium particle looks dark in the optical photo square and light, or white, in the magnified electron scattering photo. The same small uranium particle is also dark and left of center in the autoradiograph. The image appears much larger here because the Lexan on which the image is developed is about 1 mm in thickness. Under neutron bombardment, the ^{235}U fissions in three dimensions around the particle in the shape of a sphere. The autoradiograph is the Lexan surface, or a section through the sphere. The X-ray emission photo for the element silicon (Si) showed that both of the large fragments in the electron scattering photo were quartz (SiO_2).

Optical Micrograph 8 X Neutron Autoradiograph 8 X

FIGURE 3. Optical and Neutron-enhanced Autoradiography
Photomicrographs of Contaminated Soil Mount 1A

A

FIGURE 4. Optical, Electron Scattering and X-ray Emission Photomicrographs of Contaminated Soil Mount 1A

The remaining X-ray emission photos of area A for the elements, potassium, iron, aluminum, calcium, uranium, and phosphorus, are given in Figure 5. The orientation and magnifications are the same as those shown in Figure 4. The high concentration uranium particle can be seen in the uranium X-ray emission photo. The only other element coincidental with the uranium as a major constituent was phosphorus. This was confirmed by electron microprobe chemical analyses to be presented later in this section.

Area A' includes a long finger of contaminated silicon as well as several uranium particles, as seen in the autoradiograph and silicon X-ray emission photos of Figure 6. The contaminated silica is not a natural material but was part of the waste constituents. Note that at a magnification of 200X, the individual alpha tracks can be distinguished in the Lexan autoradiograph.

The remainder of the macro element distributions can be seen in Figure 7. The uranium is distributed in discrete particles rather than homogeneously throughout the soil. The uranium particles shown in Figure 8 are difficult to locate in the electron scattering or optical photos. The main uranium particles in the C area can be seen in the 200 and 500 magnifications of Figure 9.

The uranium in area D, however, is easily seen in both the optical and electron scattering photos in Figure 10. The uranium is dark in appearance within the circle on the optical photo, and light-colored to white due to its higher density in the electron scattering photo. Phosphorus is the only element associated with the uranium in an appreciable amount as seen in Figure 11.

SOIL MOUNT 1B

Optical photographs and matching autoradiographs of Mount 1B are shown in Figures 12 and 13. Note that the uranium is located on the exterior surfaces of the pebble or near the exterior surface in most cases. There has been some minor penetration into cracks, voids, or conduits by the uranium-bearing solutions. The fine material around the outside of the pebble contains the bulk of the uranium, and is apparently cemented to the pebble surface.

FIGURE 5. X-ray Emission Photomicrographs of Contaminated Soil Mount 1A

A'

(A')

200 X

(A')

200 X

Electron Scattering

200X

Si

200X

FIGURE 6. Optical Neutron-enhanced Autoradiography, Electron Scattering, and X-ray Emission Photomicrographs of Contaminated Soil Mount 1A

FIGURE 7. X-ray Emission Photomicrographs of Contaminated Soil Mount 1A

C

FIGURE 8. Optical, Neutron-enhanced Autoradiography, Electron Scattering, and X-ray Emission Photomicrographs of Contaminated Soil Mount 1A

FIGURE 9. X-ray Emission Photomicrographs of Contaminated Soil Mount 1A

(D) 50 X

Electron Scattering 200X

Si 200X

FIGURE 10. Optical, Neutron-enhanced Autoradiography, Electron Scattering, and X-ray Emission Photomicrographs of Contaminated Soil Mount 1A

FIGURE 11. X-ray Emission Photomicrographs of Contaminated Soil Mount 1A

8X

8X

50X

50X

U SOIL 1B

FIGURE 12. Optical and Neutron-enhanced Autoradiography
Photomicrographs of Contaminated Soil Mount 1B

50X

50X

200X

200X

U SOIL 1B

FIGURE 13. Optical and Neutron-enhanced Autoradiography
Photomicrographs of Contaminated Soil Mount 1B

A scanning electron microphotograph (SEM) in Figure 14 shows the location of two high concentration uranium particles. The elemental X-ray emission photographs in Figures 14 and 15 show the mineralogy of the associated rock fragments. The two largest fragments, for example, are a plagioclase feldspar, $(\text{Ca,Na})\text{AlSi}_3\text{O}_8$, and orthoclase or microcline, KAlSi_3O_8 . The smaller fragment above the plagioclase is quartz, SiO_2 . Note that once again phosphorus is associated with highest uranium concentrations.

SOIL MOUNT 2A

Mount 2A contains specific metal concentrations as shown in Figures 16 and 17. Chemical analyses of the metal are given in Table 3. The analyses show that the metal is probably stainless steel. It has apparently been melted since it is spongy in appearance and has partially recrystallized. X-ray emission photographs of part of area B shown in Figure 16 are given in Figure 18. The lightest colored areas are the stainless steel analyzed in Table 3. The origin of the stainless steel and the circumstances surrounding its disposal into the 316-4 Crib are unknown.

SOIL MOUNT 2B

Optical photographs and matching autoradiographs are shown of Mount 2B in Figures 19 and 20. Note that the pebble with no surrounding fines in the low magnification of Figure 19 (8X) shows very little exterior surface radioactivity. The bulk of the uranium is associated with the finer rock fragments surrounding the surfaces of the larger pebbles. The area in the lower right-hand corner of Figure 20 is explored further in Figure 21. The metallic phases are ilmenite (FeTiO_3) and magnetite (Fe_3O_4). The small high concentration uranium particle is circled in the 500X SEM photograph.

FIGURE 14. Scanning Electron Microscopy, Electron Scattering, and X-ray Emission Photomicrographs of Contaminated Soil Mount 1B

K

P

Si

U

URANIUM SOIL-1B

FIGURE 15. X-ray Emission Photomicrographs of Contaminated Soil Mount 1B

FIGURE 16. Optical Photomicrographs of Contaminated Soil Mount 2A

2- A

FIGURE 17. Optical and Neutron-enhanced Autoradiography Photomicrographs of Contaminated Soil Mount 2A

FIGURE 18. Electron Scattering and X-ray Emission Photomicrographs of Contaminated Soil Mount 2A

U SOIL 2B

FIGURE 19. Optical and Neutron-enhanced Autoradiography
Photomicrographs of Contaminated Soil Mount 2B

U SOIL 2B

FIGURE 20. Optical and Neutron-enhanced Autoradiography
Photomicrographs of Contaminated Soil Mount 2B

FIGURE 21. Scanning Electron Microscopy, Electron Scattering and X-ray Emission Photomicrographs of Contaminated Soil Mount 2B

TABLE 3. Electron Microprobe Chemical Analyses
of Metal in Mount 2A

<u>Constituents</u>	<u>wt%</u>			
SiO ₂	0.6	0.4	0.5	0.5
Al ₂ O ₃	0.3	0.4	1.0	0.4
MgO	0.7	0.7	0.7	0.4
P ₂ O ₅	-	-	-	-
Cr	18.4	17.6	20.7	19.4
Mn	1.7	1.4	1.9	1.7
Fe	68.5	70.7	65.7	68.3
Ni	9.9	10.1	11.0	11.6
K ₂ O	-	-	-	-
CaO	-	-	-	-
UO ₂	-	-	-	-
Total	100.1	102.0	101.5	102.3

SOIL MOUNT 3B

Mount 3B also illustrates the many finer fragments adhering to the pebble surfaces and containing the bulk of the uranium in Figure 22. Several larger magnifications of matching autoradiographs and optical photographs of selected areas containing uranium are given in Figures 22 and 23. At 200X magnification, the Lexan can be seen to resolve individual tracks caused by fissioning around a uranium particle. The 200X magnified view in the lower right-hand corner of Figure 23 is explored further in Figures 24 and 25. The uranium particle is circled in the 500X SEM photograph. Based on the X-ray emission photographs, the centralized fragment is a pyroxene with a probable composition of Ca(Mg, Fe, Al) (Al, Si)₂O₆, containing a bladed crystal of ilmenite. Most of the larger fragments are quartz with a microcline fragment in the lower left-hand corner. There are several small particles of uranium in the area.

8X

8X

50X

50X

U SOIL 3B

FIGURE 22. Optical and Neutron-enhanced Autoradiography
Photomicrographs of Contaminated Soil Mount 3B

FIGURE 23. Optical and Neutron-enhanced Autoradiography
Photomicrographs of Contaminated Soil Mount 3B

FIGURE 24. Scanning Electron Microscopy, Electron Scattering, and X-ray Emission Photomicrographs of Contaminated Soil Mount 3B

Mg

Si

Ti

U

URANIUM SOIL- 3B

FIGURE 25. X-ray Emission Photomicrographs of Contaminated Soil Mount 3B

ELECTRON MICROPROBE CHEMICAL ANALYSES AND THE URANIUM COMPOUND

Selected high concentration uranium areas in the soil mounts were chemically analyzed on the electron microprobe. These data are given in Table 4. Note that they do not total 100 wt%. Any element lighter in atomic weight than sodium generates an X-ray that is of insufficient energy for detection.

TABLE 4. Electron Microprobe Chemical Analyses of High Uranium Areas

	wt%									
	1A					1B				
SiO ₂	2.8	16.2	8.9	7.1	2.6	1.2	4.4	2.5	6.9	5.8
Al ₂ O ₃	<0.1	9.9	5.0	7.4	0.3	9.2	8.8	0.8	1.7	11.5
MgO	<0.3	-	-	-	-	1.1	0.5	1.1	0.7	0.8
CaO	1.3	8.7	1.2	0.9	1.5	2.0	0.7	1.3	0.8	0.7
FeO	1.4	3.5	2.1	1.5	2.3	2.3	2.4	5.2	9.4	2.1
TiO ₂	<0.1	-	-	-	-	0.4	0.4	0.4	4.0	0.3
K ₂ O	2.9	1.4	2.8	1.7	1.4	1.1	0.6	0.6	0.6	0.7
P ₂ O ₅	22.2	21.7	25.5	27.1	27.1	16.3	14.1	18.7	13.2	14.8
UO ₂	69.1	38.7	53.8	53.4	63.1	37.6	25.7	47.8	30.5	24.6

	2A		2B		3B		
SiO ₂	4.2	1.2	7.1	3.6	13.3	14.3	2.0
Al ₂ O ₃	10.8	15.1	5.1	3.6	5.3	5.8	2.3
MgO	0.2	0.9	0.4	-	0.4	0.2	-
CaO	0.7	0.5	0.3	0.3	1.3	0.3	0.3
FeO	1.1	18.5	1.3	1.6	0.7	1.7	1.5
TiO ₂	0.2	1.2	0.3	0.6	0.3	0.5	1.1
K ₂ O	0.6	0.8	0.6	0.2	1.5	0.4	0.6
P ₂ O ₅	11.8	4.4	15.5	3.0	14.4	8.4	8.4
UO ₂	22.2	19.0	34.4	12.7	34.5	29.8	23.7

A visual inspection of the analytical data of Table 4 leads to a suspected relationship between phosphorus and uranium concentrations. To obtain the identity of the uranium compound, it is necessary to statistically show that such a uranium-phosphorus correlation exists. The correlation coefficient (r_{12}) between two data sets (X_1 and X_2) is defined as:

$$r_{12} = \Sigma(X_1 - \bar{X}_1)(X_2 - \bar{X}_2) / \sqrt{[\Sigma(X_1 - \bar{X}_1)^2][\Sigma(X_2 - \bar{X}_2)^2]} \quad (\text{Snedecor, 1946})$$

where $\bar{X}_1$ and $\bar{X}_2$ are the mean values of X_1 and X_2 data sets, respectively, shown in Table 5, and

$$\Sigma(X - \bar{X}_1)^2 = 4077.48, \Sigma(X_2 - \bar{X}_2)^2 = 858.82, \Sigma(X_1 - \bar{X}_1)(X_2 - \bar{X}_2) = 1667.93$$

$$\begin{aligned} \text{so } r_{12} &= 1667.93 / \sqrt{(4077.48)(858.82)} \\ &= 0.89 \end{aligned}$$

TABLE 5. Associated Uranium and Phosphorus Analytical Values Listed in wt%

X_1 (UO_2)	X_2 (P_2O_5)	X_1 (UO_2)	X_2 (P_2O_5)
22.2	11.8	34.5	14.4
19.0	4.4	29.8	8.4
37.6	16.3	23.7	8.4
25.7	14.1	69.1	22.2
47.8	18.7	38.7	21.1
30.5	13.2	53.8	25.5
24.6	14.8	53.4	27.1
34.4	15.5	63.1	27.1
12.7	3.0		

According to Snedecor (1946), "student's" $t =$

$$r \sqrt{(n-2)/(1-r^2)} = 0.89 \sqrt{15/1-(0.89)^2} = 7.56$$

at 15 degrees of freedom. At the 1% level, $t = 2.898$, or stated in another way, if there were no correlation between UO_2 and P_2O_5 , a t as large as +7.56 would be expected less than 1 in 100 times from random sampling. Chances are excellent, therefore, that there is a real and positive correlation between uranium and phosphorus concentrations in the chemical analyses.

In view of the suspected UO_2 - P_2O_5 correlation, the possibility that the cause was due to the existence of a definite uranium phosphate compound was investigated through the use of a regression relationship expressed as:

$$x_2 = \bar{x}_1 + b_{x_2 x_1} (x_1 - \bar{x}_1) \quad (\text{Dixon and Massey, 1969})$$

where

$$b_{x_2 x_1} = \Sigma(x_1 - \bar{x}_1)(x_2 - \bar{x}_2) / \Sigma(x_1 - \bar{x}_1)^2$$

$$x_2 = 15.6 + 0.409 (x_1 - 36.5)$$

$$= 0.67 + 0.409 x_1$$

From the above equation, the regression line shown in the data plot of Figure 26 was constructed. Any point on this regression line will yield corresponding wt% values for P_2O_5 and UO_2 . For example, at 10.0 wt% P_2O_5 , 24.0 wt% is the corresponding UO_2 concentration. The 10.0 wt% P_2O_5 is equal to 4.36 wt% P and 24.0 wt% UO_2 is 21.16 wt% U. Dividing the 4.36 wt% P by the molecular weight of P (30.974 g/mole) and the 21.16 wt% U by the molecular weight of U (238.04 g/mole) yields a molar ratio of 0.14 P/0.09 U or approximately 3 P to 2 U. The compound, then, is very likely $(UO_2)_2^{4+} [(H_2PO_4)_2^{2-} (HPO_4)^{2-}]^{4-}$. It is useful to determine the pH that would yield the above $H_2PO_4^-/HPO_4^{2-}$ ratio = 2.

FIGURE 26. The Regression of X_2 (P_2O_5) on X_1 (UO_2)

Using the three phosphate dissociation constants and substituting these into the expression for the average number of protons (H^+) bound to a phosphate ion, $\bar{n}$, so as to eliminate everything but constants and hydrogen ion concentration, yields:

$$\bar{n} = \frac{3[H_3PO_4] + 2[H_2PO_4^-] + [HPO_4^{2-}]}{[H_3PO_4] + [H_2PO_4^-] + [HPO_4^{2-}] + [PO_4^{3-}]}$$

$$\text{With } \frac{[H^+][H_2PO_4^-]}{[H_3PO_4]} = K_1 = 10^{-2.23}, \quad \frac{[H^+][HPO_4^{2-}]}{[H_2PO_4^-]} = K_2 = 10^{-7.21}$$

$$\text{and } \frac{[H^+][PO_4^{3-}]}{[HPO_4^{2-}]} = K_3 = 10^{-12.32}, \quad \bar{n} \text{ then becomes:}$$

$$\frac{3 + \frac{2K_1}{[H^+]} + \frac{K_1K_2}{[H^+]^2}}{1 + \frac{K_1}{[H^+]} + \frac{K_1K_2}{[H^+]^2} + \frac{K_1K_2K_3}{[H^+]^3}}$$

The above expression gives the average number of protons, $\bar{n}$, bound to a phosphate ion in terms of the phosphate stepwise dissociation constants, K_1 , K_2 and K_3 , and the hydrogen ion concentration, $[H^+]$. The latter is easily manipulated to pH as follows:

$$\log \bar{n} = \frac{\frac{10^{\log 3}}{10^{\log 1 + 10^{\log K_1 + pH}}} + \frac{10^{(\log 2 + \log K_1 + pH)}}{10^{\log K_1 + 10^{\log K_2 + 2pH}}} + \frac{10^{(\log K_1 + \log K_2 + 2pH)}}{10^{\log K_1 + \log K_2 + \log K_3 + 3pH}}}{1 + \frac{10^{(-1.929 + pH)}}{10^{(-2.23 + pH)}} + \frac{10^{(-9.440 + 2pH)}}{10^{(-9.440 + 2pH)}} + \frac{10^{(-21.760 + 3pH)}}{10^{(-21.760 + 3pH)}}}$$

$$\bar{n} = \frac{3 + 10^{(-1.929 + pH)} + 10^{(-9.440 + 2pH)}}{1 + 10^{(-2.23 + pH)} + 10^{(-9.440 + 2pH)} + 10^{(-21.760 + 3pH)}}$$

Since the usual number of protons associated with each phosphate is three, n minus three is the average negative charge on the phosphate. At pH 6.9, n is 1.67 and the average negative charge, -1.33. If the compound contains three phosphate ions, as $(\text{UO}_2)_2[(\text{H}_2\text{PO}_4)_2(\text{HPO}_4)]$, then the negative charge on phosphate ions is $3(-1.33)$ or -3.99 at pH 6.9. Thus, a $(\text{H}_2\text{PO}_4)^-/(\text{HPO}_4)^{2-}$ ratio equal to 2 in the derived uranyl phosphate compound would appear to be reasonable because a near neutral pH is required to produce this ratio.

Assuming the following conditions:

1. uranium is present in the U(VI) state as UO_2^{+2}
2. the compound contains $\text{UO}_2\text{-H}_x\text{PO}_4$ only
3. the compound has neutral charge
4. the pH environment present during precipitation and currently is neutral, a probable formula for the observed compound can be estimated.

The observed P/U ratio of 3/2 and charge balance relationships, assuming U(VI), suggest that the compound is very likely $(\text{UO}_2)_2[(\text{H}_2\text{PO}_4)_2(\text{HPO}_4)]$. With the added constraints of a measured 6.9 pH and a net negative charge per phosphorus ion of -1.33, this compound appears likely (2P with -1 charge and 1P with -2 charge averages -1.33/P). Although the original uranyl nitrate and phosphorus compound may have been more acidic, the buffering capacity of the soil would tend to return it to a near-neutral pH condition. Thus, soluble uranyl nitrate solution injected into the groundwater system during past discharge operations possibly migrated from the 316-4 Crib under slightly acidic conditions. As the pH of the groundwater system was effectively buffered the solubility decreased and migration subsequently was retarded. Uranyl phosphate, $\text{UO}_2\text{HPO}_4 \cdot 4\text{H}_2\text{O}$, is reported to be insoluble in water at pH 7.0 and in acetic acid but soluble in nitric acid and sodium carbonate solutions (Weast, 1977).

X-RAY IDENTIFICATION OF OTHER MINERAL PHASES ASSOCIATED WITH URANIUM

In addition to the high concentration uranium particles, other uranium occurrences were located that were much lower in uranium concentration, but the concentrations were homogeneous over a relatively large area. Chemical analyses of the solid phase on which the uranium was apparently adsorbed are given in Table 6. After extraction and mounting on a capillary, the sample was X-rayed using a small particle X-ray diffraction technique. The resulting pattern was that of nontronite, $[\text{Fe}^{3+}]_{2.00}[\text{Al}_{0.33}\text{Si}_{3.67}\text{O}_{10}(\text{OH})_2\text{X}_{0.33}]$, in which $\text{X}_{0.33}$ represents the exchangeable cations. Nontronite may also contain magnesium in the octahedral (iron) position. The analyses also show that the formulation contains excess hydrated iron oxides mixed with the nontronitic clay and that the clay is predominantly potassium based. Nontronite is a member of the smectite group of clay minerals. Smectite group clay minerals are a natural soil constituent present in most Hanford soils. Hydrous oxides also are naturally present in Hanford soils.

TABLE 6. Electron Microprobe Chemical Analyses of Low Concentration Uranium Particles in Mount 3B

<u>Constituents</u>	<u>A, wt%</u>	<u>B, wt%</u>
SiO_2	38.5	38.2
Al_2O_3	13.1	12.5
TiO_2	4.0	3.8
Fe_2O_3	21.6	21.8
CaO	0.5	0.4
MgO	7.7	8.1
K_2O	7.9	8.4
Na_2O	-	-
P_2O_5	1.2	1.0
UO_2	1.2	1.2
Total	95.7	95.4

Particle A was examined by X-ray emission in Figures 27 and 28, and particle B in Figures 29 and 30. The uranium content of the smectite clays is relatively low compared to the uranium found in the phosphorus compound, and it is evenly distributed throughout the particle. An adsorption or ion exchange mechanism of uranyl ion uptake by the clay particles is suggested by the above uranium distribution pattern.

URANIUM LEACHING RESULTS

Three uranium-contaminated soil samples were collected from directly beneath the disposal site at 316-4 Crib. The uranium concentration of the three samples is given in Table 7.

TABLE 7. Uranium Concentration in Soil Samples
Collected from Beneath 316-4 Crib

<u>Soil Sample</u>	<u>Depth, cm</u>	<u>U, $\mu\text{g/g}$ Soil</u>	<u>Total</u>
1	0-25	444 $\pm$ 155	Background
2	25-50	623 $\pm$ 220	"
3	50-75	912 $\pm$ 320	"
Uncontaminated	-	9.24 $\pm$ 3.2	-

The uncontaminated soil sample was collected from several meters below the surface at a nearby construction site for the purpose of obtaining a uranium background value. One reason that the uranium concentration in the crib soils increases with depth was the custom of rinsing the transport vessel which delivered contaminated liquid waste to the site with water. The soluble uranyl tended to be flushed down the soil column by the disposal of the dilute wash water.

Small columns of uranium-contaminated Soils 1, 2 and 3 were used to determine the leachability of the uranium. Distilled water, 0.1 N HNO_3 and methyl isobutyl ketone, respectively, were used as leaching agents.

Electron Scattering 300X

Al

Ca

Fe

K

SMECTITE 3B-A

FIGURE 27. Electron Scattering and X-ray Emission Photomicrographs of Soil Mount 3B (Particle A)

P

Si

Ti

U

SMECTITE 3B-A

FIGURE 28. X-ray Emission Photomicrographs of Contaminated Soil Mount 3B (Particle A)

Electron Scattering 300X

Al

Ca

Fe

K

SMECTITE 3B-B

FIGURE 29. Electron Scattering and X-ray Emission Photomicrographs of Soil Mount 3B (Particle B)

P

Si

Ti

U

SMECTITE 3B- B

FIGURE 30. X-ray Emission Photomicrographs of Contaminated Soil Mount 3B (Particle B)

The leaching agents were added slowly to the top of the soil columns, with flow rates determined by soil column packing. The column flow rates averaged about $0.08 \text{ cm}^3/\text{min}/\text{cm}^2$.

The total amounts of uranium eluted by each agent from Soils 1, 2 and 3 are given in Table 8. In general, the 0.1 N HNO_3 solution ($\text{pH} = 1.0$) eluted the most uranium from all three soils, while the organic solvent, methyl isobutyl ketone, eluted the least amount of uranium. The leaching results are consistent with uranium present as a sparingly water soluble uranyl phosphate. Curves for cumulative uranium leaching vs. column volumes of throughput are given in Figures 31, 32 and 33 for distilled water, methyl

TABLE 8. Total Uranium Eluted from Soils 1, 2 and 3 by Approximately Three Column Volumes of Distilled Water, Methyl Isobutyl Ketone and $0.1 \text{ Normal Nitric Acid}$ (Total U on the columns is given in Figures 24, 25 and 26.)

<u>Soil</u>	<u>Column Volumes</u>		<u>Total U Eluted, pCi</u>
		<u>Distilled Water</u>	
1	3.0		1.98
2	3.3		36.22
3	3.4		16.05
		<u>Methyl Isobutyl Ketone</u>	
1	3.1		1.59
2	3.2		1.17
3	3.7		1.45
		<u>0.1 N HNO_3</u>	
1	2.8		224.3
2	2.7		3123.7
3	3.0		589.3

FIGURE 31. Cumulative Leaching of Uranium from Soils 1, 2 and 3 by Distilled Water. The uranium contents per g of each soil was given in Table 7. Total U on columns of soils 1, 2 and 3 were 18,041 pCi, 28,522 pCi and 48,772 pCi, respectively, with column volumes of 9.45 cm^3 , 11.34 cm^3 and 11.34 cm^3 .

FIGURE 32. Cumulative Leaching of Uranium from Soils 1, 2 and 3 by Methyl Isobutyl Ketone. The uranium contents per g of each soil was given in Table 7. Total uranium on columns of Soils 1, 2 and 3 were 21,429 pCi, 27,265 pCi and 46,888 pCi, respectively, with column volumes of 11.34 cm³, 11.40 cm³ and 11.34 cm³.

FIGURE 33. Cumulative Leaching of Uranium from Soils 1, 2 and 3 with 0.1 N HNO_3 . The uranium contents per g of each soil was given in Table 7. Total uranium on columns of Soils 1, 2 and 3 were 7733 pCi, 14,529 pCi and 17,805 pCi, respectively, with column volumes of 5.42 cm^3 , 4.51 cm^3 and 4.96 cm^3 .

isobutyl ketone and dilute nitric acid, respectively. Note that in each case, the uranium elution after about one column volume remained nearly constant. The final uranium-elution concentrations given in pCi U/ℓ of leaching agent passed through columns of Soils 1, 2 and 3 are shown in Table 9. The maximum permissible concentration of soluble ^{238}U is 40,000 pCi/ℓ in water for unrestricted areas and 13,333 pCi/ℓ as the population guide, while the comparable values for soluble natural uranium are 30,000 pCi/ℓ and 10,000 pCi/ℓ above the natural background (10 CFR-Part 20--Standards for Protection Against Radiation--U.S. Nuclear Regulatory Commission, April 30, 1975). Hence, only the acid leachates exceed the above water standards for dissolved uranium. It would seem prudent to avoid adding acidic, aqueous solutions to the 316-4 Crib. Neither rainwater nor certain organic solvents appear to constitute effective uranium leaching agents for the uranium forms found in Soils 1, 2 and 3 from 316-4 Crib.

TABLE 9. Final Uranium Concentrations in Eluting Solutions (3 Column Volumes) from Soils 1, 2 and 3

<u>Soil</u>	Uranium Eluate Concentration, pCi/ℓ
	<hr/>
	<u>Distilled Water</u>
1	88.4
2	664.0
3	366.0
	<u>Methyl Isobutyl Ketone</u>
1	41.9
2	24.7
3	64.1
	<u>0.1 N HNO₃</u>
1	21,116
2	325,372
3	55,641

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