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Effects of Hanford High-Level Waste Components on Sorption of Cobalt, Strontium, Neptunium, Plutonium, and Americium on Hanford Sediments

C.H. Delegard
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EFFECTS OF HANFORD HIGH-LEVEL WASTE COMPONENTS ON SORPTION OF COBALT, STRONTIUM, NEPTUNIUM, PLUTONIUM, AND AMERICIUM ON HANFORD SEDIMENTS

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ABSTRACT

To judge the feasibility of continued storage of high-level waste solutions in existing tanks, effects of chemical-waste components on the sorption of hazardous radioelements were determined. Experiments identified the effects of 12 Hanford high-level waste-solution components on the sorption of cobalt, strontium, neptunium, plutonium, and americium on 3 Hanford 200 Area sediments. The degree of sorption of strontium, neptunium, plutonium, and americium on two Hanford sediments was then quantified in terms of the concentrations of the influential waste components. Preliminary information on the influence of the waste components on radioelement solubility was gathered.

Of the 12 Hanford waste-solution components studied, the most influential on radioelement sorption were $NaOH$, $NaAlO_2$, *HEDTA, and EDTA.* The chelating complexants, HEDTA and EDTA, generally decreased sorption by complexation of the radioelement metal ions. The components* $NaOH$ *and* $NaAlO_2$ *decreased neptunium and plutonium sorption and increased cobalt sorption. Americium sorption was increased by NaOH. The three Hanford sediments' radioelement sorption behaviors were similar, implying that their sorption reactions were also similar. Sorption prediction equations were generated for strontium, neptunium, plutonium, and americium sorption reactions on two Hanford sediments. The equations yielded values of the distribution coefficient,* K_d, *as quadratic functions of waste component concentrations and showed that postulated radioelement migration rates through Hanford sediment could change by factors of 13 to 40 by changes in Hanford waste composition.*

**N-hydroxyethylethylenediaminetriacetic acid and ethylenediaminetetraacetic acid, respectively.*

CONTENTS

Introduction 1

Experimental 3
    Materials 3
    Procedures 6
    Experimental Design and Data Analysis 10
    Radioelement Solubility Tests 11

Results and Discussion 13
    Preliminary Radioelement Solubility Tests 13
    Radioelement Solubility Screening Tests 15
    Radioelement Sorption Screening Tests 17
    Radioelement Sorption Differences for Different Sediments 22
    Radioelement Sorption Prediction Equations 24

Conclusions 31

Acknowledgments 33

References 35

Appendices:
    A. Statistical Methods A-1
    B. Sorption and Solubility Screening Test Data B-1
    C. Sorption Prediction Equation Data C-1

FIGURE:
    1. Sediment Strata Underlying 200 East and West Areas: Sources of Hanford Sediments Studied 5

TABLES:
    1. Component Concentration Values 4
    2. Sources and Properties of Hanford Sediment Samples . . 4
    3. Radioelement Concentrations 7
    4. Composition of Radioelement Solubility Test Solutions 12
    5. Preliminary Radioelement Solubility Results 14
    6. Factor Effects and Error Data for Radioelement Solubility 16
    7. Significant Components in Radioelement Sorption . . . 18
    8. Sediment Sorption Differences, Students' t Test . . . 23
    9. Radionuclide Sorption Prediction Equations 25
    10. Composition of Hanford Waste Solution Types 27
    11. Predicted Radionuclide Distribution Coefficients and Retardation Factors for the Hanford Waste Solution Types 28

INTRODUCTION

High-level radioactive defense waste solutions, resulting from plutonium recovery and waste processing activities at Hanford, currently are stored in underground mild steel-lined concrete tanks in the Hanford Site 200 Area plateau. Located in thick (∿45 m) sedimentary beds of sand and gravel well above the water table, the waste tanks provide a measure of isolation of the hazardous radionuclides from the biosphere.

One possible pathway for transport of radionuclides from Hanford waste tanks to the biosphere is migration of contaminated waste solutions from failed waste tanks through the surrounding sediment. Though waste tanks currently are being drained of mobile liquids, additional mobile liquids can accumulate by recrystallization and dewatering of amorphous hydrous oxides, crystal growth of waste salts resulting in drainage of pore liquids, transport of sediment pore water to the hygroscopic wastes, percolation of surface water to the tanks, or rise in groundwater level. Precipitation of radioelements from solution and sorption of radioelements on the sediments surrounding the tanks would maintain isolation if radioactive solutions were to leak from the waste tanks.

Chemical components in Hanford waste, however, could affect radioelement solubility and sorption reactions and thus enhance or reduce radionuclide transport from failed tanks to the biosphere. Radioelement sorption also can be affected by the sediment on which sorption occurs. To judge the feasibility of continued storage of Hanford high-level waste (HLW) solutions in existing tanks, effects of the chemical waste components on the solubility and sorption of hazardous radioelements must be determined.

In this study, a series of experiments were performed in which the effects of Hanford waste components on radioelement sorption reactions with Hanford sediments were determined and quantified. In performing the sorption experiments, tentative information concerning the influence of Hanford waste components on radioelement solubility also was obtained.

Twelve Hanford waste components, five radioelements, and three Hanford sediments were included for study. The 12 components were $NaNO_3$, $NaNO_2$, NaOH, $NaAlO_2$, Na_2CO_3, Na_2SO_4, Na_3PO_4, NaF, HEDTA,* EDTA,† hydroxyacetic acid, and citric acid. The five radioelements were cobalt, strontium, neptunium, plutonium, and americium. The three Hanford sediments were taken from wells located near and from strata underlying Hanford 200 Area high-level waste tank farms.

Two statistical approaches were used. The first approach identified waste components that significantly influenced radioelement sorption reactions. The second statistical approach quantified the degrees of sorption as functions of the concentrations of the significant waste components. This document describes and summarizes the results of these sorption studies. The effects of time, temperature, and radioelement concentration on the sorption reactions, the macrocomponent chemistry of the waste-sediment reactions, and the reversibility of the sorption reactions are not addressed here, but are worthy subjects for future studies.

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*N-hydroxyethylethylenediaminetriacetic acid.

†Ethylenediaminetetraacetic acid.

EXPERIMENTAL

MATERIALS

Reagents

Twelve chemical components of the Hanford high-level waste solutions were studied. The 12 waste components studied were $NaNO_3$, $NaNO_2$, NaOH, $NaAlO_2$, Na_2CO_3, Na_2SO_4, Na_3PO_4, NaF, HEDTA, EDTA, hydroxyacetic acid, and citric acid. Due to ease of dissolution, the complexants HEDTA and EDTA were introduced to the experimental solutions as their tri- and tetrasodium salts, respectively. Reagent-grade chemicals and distilled and deionized water were used to prepare all experimental solutions. Sodium aluminate, unavailable in reagent grade, was produced as required using equimolar quantities of reagent grade NaOH and $Al(OH)_3$.

The 12 waste components were selected for study based on their quantities in the Hanford HLW chemical inventory, their solubilities, and their abilities to complex or to influence the complexation of metallic radioelements. To enhance the effectiveness of the screening tests, the ranges of component concentrations selected were as broad as possible without causing precipitation, while also representing genuine Hanford HLW.(1) The component concentrations are given in Table 1. The reasons for the high, middle, and low concentration values are explained in Appendix A.

Sediments

Three Hanford sediments were selected for study. The three sediment samples were taken from wells located near and from strata underlying Hanford 200 Area tank farms. Selection of these particular strata was based on their varied radionuclide sorption and ion exchange properties.(2) The properties of the three sediments are given in Table 2 and the sources of the sediments are shown in Figure 1.

TABLE 1. Component Concentration Values.

Component	Concentration ($\underline{M}$) High (+)	Concentration ($\underline{M}$) Middle (0)	Concentration ($\underline{M}$) Low (-)
$NaNO_3$	2	1	0
$NaNO_2$	2	1	0
NaOH	4	2.5	1
$NaAlO_2$	0.5	0.25	0
Na_2CO_3	0.05	0.025	0
Na_2SO_4	0.01	0.005	0
Na_3PO_4	0.01	0.005	0
NaF	0.01	0.005	0
Na_3HEDTA	0.1	0.05	0
Na_4EDTA	0.05	0.025	0
Hydroxyacetic acid	0.1	0.05	0
Citric acid	0.03	0.015	0*

*The lower citric acid concentration was 0.005$\underline{M}$ for the americium sorption and solubility screening studies so that sufficient americium concentration would be present in the feed solution.

TABLE 2. Sources and Properties of Hanford Sediment Samples.(2)

Sediment	Source			K_d (mL/g) in 3$\underline{M}$ $NaNO_3$, 0.01$\underline{M}$ $Ca(NO_3)_2$			CEC* (meq/100 g) pH 7
	Well	Depth (m)	Approximate location	Sr	Cs	Co	
L	E17-4	10-78	241-A Tank Farm	0.30	23	1,500	3.64
P	E33-22	26-59	241-BY, -B Tank Farms	0.31	35	5,700	3.50
S	W10-9	14-24	241-T Tank Farm	0.32	54	4,100	6.79

*CEC = cation exchange capacity.

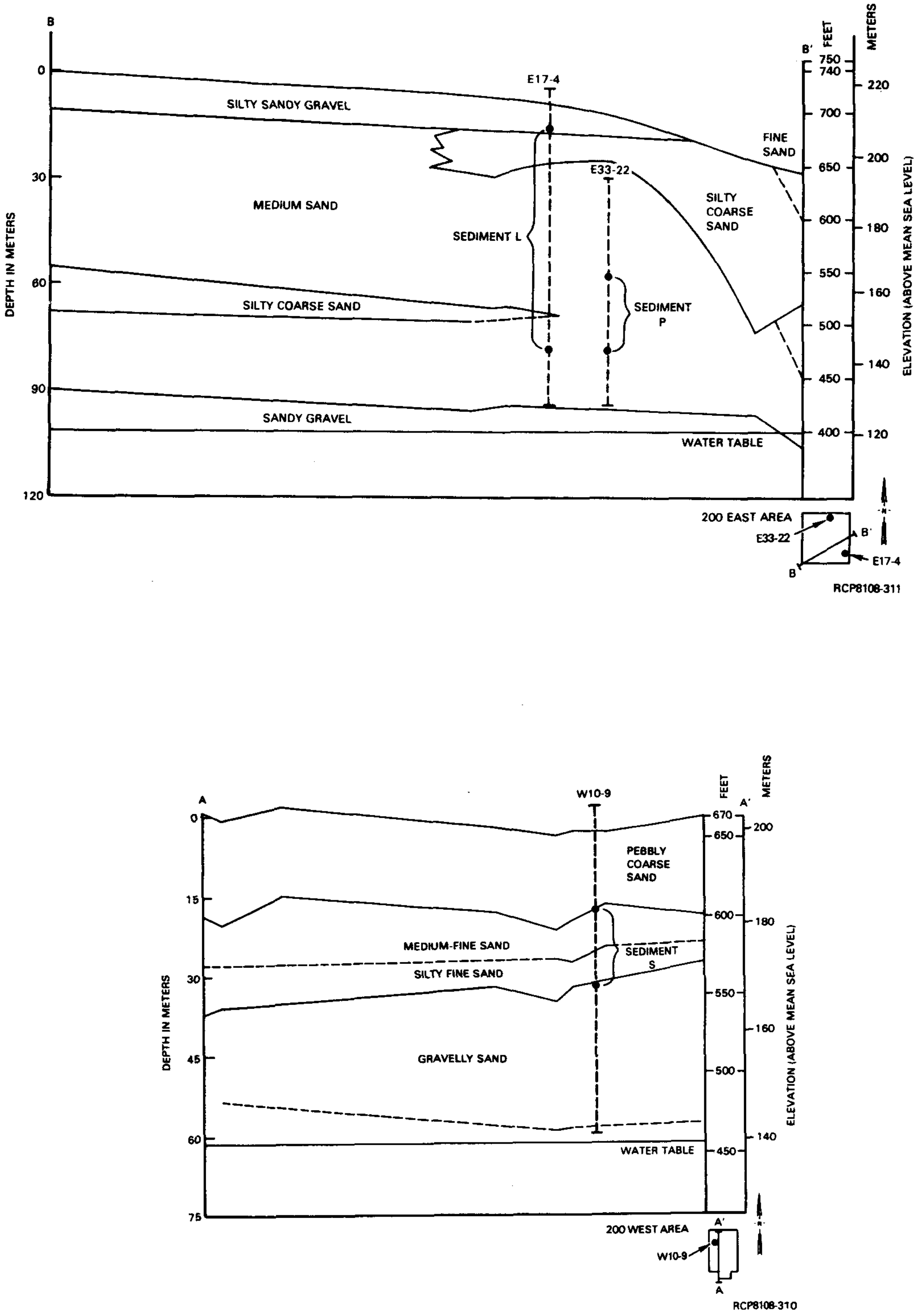

FIGURE 1. Sediment Strata Underlying 200 East and West Areas: Sources of Hanford Sediments Studied.

Radioelements

Five radioelements were selected for study: cobalt, strontium, neptunium, plutonium, and americium. These radioelements were selected because of their concentration in Hanford HLW, their potential ability to form complexed species, and their radiological hazard. Table 3 summarizes the activity concentrations of these radioelements in some filtered Hanford HLW solutions and the corresponding chemical concentrations of both radioactive and inactive forms of these radioelements.

PROCEDURES

This study was based on experiments in which distributions of radioelements between sediment and solution were determined. One measure of the radioelement sediment-solution distribution is the distribution coefficient, K_d. The K_d is defined as the equilibrium amount of radioelement reporting to the solid, per gram of solid, divided by the amount of radioelement dissolved in solution, per milliliter of solution. The K_d thus has units of milliliters per gram.

The fraction of radioelement sorbed is another measure of the radioelement sediment-solution distribution. Fraction sorbed has some arithmetic advantages over K_d for use in statistical evaluations. In cases of high radioelement sorption and low final solution concentration, the final solution concentration has high relative error. The low valued/high error final solution concentrations yield large K_d values with large error. If the final solution concentration is below the detection limit, the K_d assumes a value greater than some large number. Large K_d values can have errors larger than the lower K_d values themselves. In contrast, final solution concentrations near or below the detection limit yield fraction sorbed values that are bounded (cannot exceed 1.0) and have errors comparable to errors found at lower fraction sorbed values. The "well-behaved" nature of the fraction-sorbed function therefore makes it more useful than K_d in the statistical analyses.

TABLE 3. Radioelement Concentrations.

Radionuclide	Radionuclide concentration (μCi/L)	Total element concentration (μmoles/L)
Concentration in Hanford HLW[a]		
^{60}Co	153	0.002[b]
^{90}Sr	3,190 - 4,800	0.75 - 1.1[b]
^{237}Np	2.8	17[b]
^{239,240}Pu	0.46 - 6.8	0.023 - 0.38[b]
^{241}Am	0.12 - 21	0.00016 - 0.025[b]
Concentration in Experiments		
^{60}Co	-	0.08
^{85}Sr	-	0.8
^{237}Np	-	30
^{237}Pu[c]	-	0.00002
^{238}Pu[c]	-	0.002
^{241}Am	-	0.0006

[a] All HLW solutions were filtered through 0.45-μm pore filters before analyses.

[b] For Hanford HLW, assume:

total Co/^{60}Co = 1 (may not be valid, no figures available for chemical concentration of Co)

total Sr/^{90}Sr = 3

total Pu/^{239,240}Pu = 1

total Am/^{241}Am = 1

all Np in tank farms (106 Ci)(3) dissolved uniformly in waste liquor.

[c] In the experimental solutions, ^{237}Pu was used for the solubility scouting test; ^{238}Pu was used for the statistical solubility and sorption tests.

Each distribution coefficient experiment was conducted in the following manner. First, a representative subsample of sediment was taken using the cone and quarter method. In this method, the entire sediment sample is poured on a clean surface into a cone-shaped pile. The cone is flattened, then cut with a broad spatula into quarter segments. Each quarter may be split the same way until the desired sample size is obtained. The principal advantage of the cone and quarter method is that it generates subsamples without particle segregation according to size.

The sediment subsample was then added to a tare-weighed polyethylene vial. Sediment weights ranged from about 0.75 to 1.3 g. Next, 10 mL of the appropriate untraced solution were added. The vial, with sediment and solution, was gently shaken for at least 2 hr to pre-equilibrate the sediment with solution. Following shaking, the vial was centrifuged and the supernatant solution removed and discarded. The pre-equilibration was repeated.

The pre-equilibrium steps were included in the sorption experiments to simulate the migration of radioelements from the waste tanks through sediments wetted by waste solutions. Advancement of a plume of radioelements from a tank leak can occur only if the radioelements are not sorbed on sediment already wetted by waste solutions. Radioelements not sorbed on waste-wetted sediments will advance with the solution front to unwetted sediments.

Following decantation of the supernate from the second pre-equilibration step, the vial with sediment and residual solution was reweighed. The volume of residual solution was calculated using the solution's density and the residual solution's weight.

While the sediment was pre-equilibrating, radionuclide-traced solutions of the same chemical composition as the pre-equilibration solutions were prepared. Each solution was prepared by dissolving a portion of solid tracer (obtained by drying, under a heat lamp, precise volumes of stock tracer solution) in the appropriate volume of solution. Each vial of solution with dried tracer was shaken 3 days to aid tracer dissolution.

The traced solution was then filtered through a 0.2-μm pore filter to remove any undissolved tracer solids.

Five milliliters of traced solution were then added to the vial with the pre-equilibrated sediment. This mixture was gently shaken for 7 days. The vial was centrifuged and the supernatant solution filtered through a 0.003-μm pore ultrafilter to remove suspended solids.

Portions of the traced filtered solutions, from before and after contact with the sediment, were analyzed for radionuclide concentration. The tracer concentration before contact, C_1, and after contact, C_2 (both in units of activity per milliliter); together with the sediment weight, W (in grams); plus the residual solution volume, V (in milliliters); were then substituted into the following equation to give the distribution coefficient, K_d:

$$K_d = \frac{\text{activity/g soil}}{\text{activity/mL solution}} = \frac{(5C_1 - (5+V)C_2)/W}{C_2}$$

$$\text{or} \qquad (1)$$

$$K_d = \frac{5C_1 - (5+V)C_2}{C_2 W}$$

Fraction sorbed values were also calculated. To account for differences in sediment weights and solution volumes, each fraction sorbed value was corrected to a 1-g sediment weight and 5-mL solution volume using the following equations:

$$F_{Sor} = 1 - F_{Sol} = 1 - C_2/C_1 \qquad (2)$$

$$\text{Fraction sorbed (corrected)} = \frac{F_{Sor}(5+V)}{5W(1-F_{Sor}) + F_{Sor}(5+V)}$$

where

F_{Sor} = uncorrected fraction sorbed

and

F_{Sol} = uncorrected fraction in solution.

Concentrations of most of the radionuclides in the experiments were determined through gamma spectrometry of prominent gamma photon peaks. For the ^{237}Pu and ^{238}Pu tracers, concentrations were determined utilizing their low-energy X-ray peaks.

EXPERIMENTAL DESIGN AND DATA ANALYSIS

The objectives of these studies were to identify Hanford waste components that significantly affect radioelement sorption and then to quantify the effects of the component concentrations on radioelement sorption reactions on Hanford sediment. To ascertain efficiently the effects of the 12 waste components on the sorption of the 5 radioelements on the 3 Hanford sediments, a 20-run Plackett-Burman experimental design was employed.(4) The Plackett-Burman design identified waste components that significantly influence radioelement sorption. The components found to be significant in the Plackett-Burman screening tests then were used as variables in a 3-level experimental design, the Box-Behnken technique. The Box-Behnken technique was used to generate equations yielding sorption parameters as functions of the significant waste components' concentrations.(5) The sorption screening tests (Plackett-Burman) were run for cobalt, strontium, neptunium, plutonium, and americium. The sorption quantifying tests (Box-Behnken) were run for strontium, neptunium, plutonium, and americium.

The Plackett-Burman design is a 2-level fractional factorial design particularly effective for screening variables to determine their significance in affecting parameters of interest. In these tests, the 12 waste components were the variables; radioelement sorption was the parameter of interest. The 20-run design was selected as having sufficient capability to screen 12 variables while giving satisfactory error data for the significance tests required for the design.

The Box-Behnken design is a 3-level, multivariable design that yields, through regression analyses, values of the parameter of interest (radioelement sorption) as arithmetic functions of the significant variables (waste components' concentrations) identified from the Plackett-Burman tests. The 3- and 4-variable Box-Behnken designs used in these studies required 15 and 27 experiments, respectively. Details of the design and interpretation of the Plackett-Burman and Box-Behnken tests are given in Appendix A.

RADIOELEMENT SOLUBILITY TESTS

Before beginning the experiments, it was recognized that certain radioelements might not be measurably soluble in the presence of certain waste components. As a result of this low solubility, tracer concentration might have been insufficient to conduct the sorption experiments for certain solutions.

To guarantee that sufficient tracer concentration existed in solution to conduct the sorption experiments, the effects of the 12 waste components on radioelement tracer solubility were determined. The solubility of each of the 5 radioelements was determined for 13 different solutions. One solution was $4\underline{M}$ NaOH. Eleven solutions were $1\underline{M}$ NaOH solutions of the remaining non-NaOH components. The thirteenth solution contained all 12 waste components. The compositions of the 13 test solutions are given in Table 4.

To introduce the radioelement tracers, precisely measured, dried portions of each tracer were added to polyethylene vials that contained each of the 13 solutions, as well as to a reference solution of $6\underline{M}$ HNO_3. The solutions with tracers were shaken 3 days to aid tracer dissolution. The traced solutions then were filtered through 0.003-µm pore filters to remove any undissolved solids. The concentrations of dissolved radionuclides were determined in duplicate for the 13 test solutions and compared with the concentrations found, in duplicate, for the reference $6\underline{M}$ HNO_3 solution. Interpretation of these data allowed adjustment of the experiments' component concentrations to ensure that sufficient tracer concentration existed in solution to conduct the sorption experiments.

TABLE 4. Composition of Radioelement Solubility Test Solutions.

Solution	Concentration (M)
NaOH	4
$NaNO_3$	2[a]
$NaNO_2$	2[a]
$NaAlO_2$	0.5[a]
Na_2CO_3	0.05[a]
Na_2SO_4	0.01[a]
Na_3PO_4	0.01[a]
NaF	0.01[a]
Na_3HEDTA	0.1[a]
Na_4EDTA	0.05[a]
Hydroxyacetic acid	0.1[a]
Citric acid	0.03[a]
Hanford waste[b]	As above[c]

[a] 1M NaOH.

[b] Composed of all 12 components.

[c] 4M NaOH.

The effects of individual waste components on radioelement solubilities in Hanford HLW were determined more systematically by application of the Plackett-Burman technique. The input parameters were the initial solution concentrations. Since the feed solutions for the sorption screening experiments were produced by adding equal portions of precisely measured dried tracers to equal volumes of the 20 test solutions, the concentrations of tracer in the test solutions should have been equal. The actual concentrations of radionuclide in the feed solutions, analyzed by the Plackett-Burman technique, would then reveal which waste components significantly affected radioelement solubility at the levels of radioelements used in these experiments (see Table 3). Analysis of data was analogous to the technique described for the sorption tests; radionuclide concentration was substituted for fraction sorbed. Methods of data analysis are discussed in Appendix A.

RESULTS AND DISCUSSION

PRELIMINARY RADIOELEMENT SOLUBILITY TESTS

The preliminary radioelement solubility tests were conducted to determine the effects of single waste components, in 1M NaOH solution, on the solubilities of the five radioelements. The solubilities of the radioelements in the presence of 4M NaOH alone, as well as in the presence of all 12 waste components in a simulated Hanford waste, also were studied. The purpose of the preliminary tests was to determine if certain components cause low radioelement solubility and, if so, to adjust the experimental conditions appropriately to ensure that radionuclide concentration would be high enough to conduct the sorption experiments.

The concentrations of each radioelement in each waste component solution were determined and compared with the radioelement concentration found in identically prepared 6M HNO_3 control solutions. Ratios of the radioelement concentrations found in the test and 6M HNO_3 control solutions were calculated to determine fraction dissolved. The dissolved fraction values are presented in Table 5. Neptunium was soluble in the presence of most of the 12 components. Slightly reduced neptunium solubility was noted in the presence of NaOH, $NaAlO_2$, and NaF. Fractions of strontium dissolved were found to be near unity for all component solutions except Na_3PO_4. For cobalt, dissolved fraction values near unity were found only for EDTA, HEDTA, and citric acid. Of the remaining components, Na_3PO_4 gave the lowest ratio. For plutonium, Na_3PO_4 again gave the lowest dissolved fraction value. For the other components, dissolved fraction values were greater than unity. The high values found for plutonium might be explained by differences in container wall sorption in the alkaline test solutions as compared with the acidic control solution.

While cobalt, strontium, and plutonium were found to be less soluble in the presence of phosphate, their concentrations still allowed sorption experiments to be carried out. Likewise, no solubility problems were anticipated for neptunium. However, the preliminary solubility tests, summarized in Table 5, showed that only in the presence of EDTA, HEDTA, or citric acid was americium in sufficient concentration to conduct the sorption experiments. Examination of the Plackett-Burman factor matrix in Appendix A showed that for certain test solutions, low (-) concentrations were specified for each of these three components. To ensure that sufficient americium concentrations existed in solution to conduct the sorption tests, the low (-) concentration of citric acid was changed from zero to 0.005M for the americium sorption experiments.

TABLE 5. Preliminary Radioelement Solubility Results.

Solution component*	Fraction dissolved				
	Cobalt	Strontium	Neptunium	Plutonium	Americium
NaOH	0.67	1.01	0.67	1.12	0.098
$NaNO_3$	0.62	1.01	0.80	1.19	0.03
$NaNO_2$	0.74	0.99	0.95	1.24	0.047
$NaAlO_2$	0.62	0.98	0.74	1.07	<0.006
Na_2CO_3	0.74	0.99	1.01	1.11	0.049
Na_2SO_4	0.65	1.01	0.92	1.03	0.076
Na_3PO_4	0.53	0.47	0.97	0.31	<0.006
NaF	0.78	0.96	0.69	1.16	0.085
Na_3HEDTA	0.95	0.98	1.03	1.19	1.05
Na_4EDTA	0.95	0.99	1.03	1.12	0.94
Hydroxyacetic acid	0.79	1.01	1.00	1.09	0.11
Citric acid	0.94	0.97	1.03	1.26	0.98
Hanford waste	0.89	1.01	0.94	1.26	0.83

*Refer to Table 4 for solution compositions.

RADIOELEMENT SOLUBILITY SCREENING TESTS

The effects of individual waste components on radioelement solubilities in the presence of other waste components were determined systematically using the Plackett-Burman design. Parameters used in the Plackett-Burman analysis were the initial radionuclide concentrations for the 20 Plackett-Burman test solutions. The initial radionuclide test solution concentrations (defined as C_1 earlier) are given in Tables B-1 through B-5 of Appendix B for the five elements studied. The factor effects determined for each of the 12 components with respect to radioelement solubility are given in Table 6. Also given in Table 6 are the errors of these Plackett-Burman analyses calculated at the 80%, 90%, and 95% confidence level (CL). The factor effects given in Table 6 are the average effects the waste components have on the radionuclide concentrations. For example, $NaNO_3$ decreased cobalt solubility by 8.46 μCi/L while Na_3HEDTA increased cobalt solubility by 21.25 μCi/L, on the average. Components having absolute values of factor effects greater than the error of the analysis are judged to be significant in affecting solubility at the CL selected. Thus, the effect of $NaNO_3$ on cobalt solubility was found to be not significant at the 80% CL while Na_3HEDTA was significant at the 95% CL.

Only two components, HEDTA and EDTA, were found to be significant in influencing cobalt solubility. At the 95% CL, both increased cobalt solubility. Inspection of formation constant data revealed that Co^{2+} forms strong complexes with HEDTA and EDTA.(6) The chelates EDTA and HEDTA were also found, at the 95% CL, to increase Sr^{2+} solubility. Formation constant data showed that strontium forms strong EDTA and HEDTA complexes.(6)

Neptunium (V) is the probable oxidation state for neptunium in the oxygenated solution matrix of these experiments. As such, the hydrolyzed neptunium (V) species expected in these high pH solutions is the neptunyl ion, $NpO_2(OH)_2^-$.(7) Hydroxyacetic acid was found to be significant at the 95% CL in increasing neptunium solubility. The NpO_2^+ species does form stable hydroxyacetate complexes.(8) Next in importance in influencing neptunium solubility was Na_2SO_4. At the 90% CL, Na_2SO_4 increased neptunium solubility, presumably through complexation. Sodium hydroxide decreased neptunium solubility at the 90% CL, presumably due to precipitation as $NpO_2OH(s)$. The organic chelating agent HEDTA increased neptunium solubility at the 80% CL. The increased solubility was probably due to complexation.(8)

TABLE 6. Factor Effects and Error Data for Radioelement Solubility.

Factors	Factor effects (μCi/L)				
	Cobalt	Strontium	Neptunium	Plutonium	Americium
Component					
$NaNO_3$	-8.46	-31.63	-0.089	-0.089	-0.041
$NaNO_2$	-5.78	-37.79	-0.040	0.067	-0.044
NaOH	6.34	45.53	-0.296◐	-0.181	0.003
$NaAlO_2$	-5.62	0.41	-0.152	-0.286	-0.013
Na_2CO_3	7.31	9.99	-0.125	0.523	0.071
Na_2SO_4	1.06	-15.05	0.356◐	0.402	0.018
Na_3PO_4	1.23	11.63	-0.147	0.327	0.069
NaF	-0.52	12.57	0.100	0.251	0.029
Na_3HEDTA	21.25●	107.99●	0.252^0	0.649^0	0.165●
Na_4EDTA	21.27●	95.43●	0.103	0.129	0.163●
Hydroxyacetic acid	8.29	25.21	0.536●	0.651^0	0.090^0
Citric acid	-0.61	-11.39	-0.089	0.453	0.034
Error					
95% CL	17.11	82.58	0.367	0.994	0.127
90% CL	13.71	66.17	0.294	0.796	0.102
80% CL	10.24	49.41	0.220	0.595	0.076

NOTE: Significant at: ● - 95% CL
◐ - 90% CL
0 - 80% CL.

Some uncertainty exists over the dissolved plutonium species in high pH, high Eh (i.e., oxygenated) systems such as the Hanford waste/ Hanford sediment system considered here. Singly charged, anionic, hydroxide-coordinated plutonium species are postulated for all oxidation states, (III), (IV), (V), and (VI).(7)

At the 80% CL, only HEDTA and hydroxyacetic acid affected plutonium solubility significantly. Presumably, both components increased plutonium solubility, whether in the (III), (IV), (V), or (VI) state, through complexation. High formation constants for Pu (IV) acetate, Pu (IV) oxalate, Pu (V) HEDTA and Pu (VI) hydroxyacetate are known.(9)

At the 95% CL, both EDTA and HEDTA were found to increase americium solubility. At the 80% CL, hydroxyacetic acid was also found to increase americium solubility. As shown in the preliminary solubility tests, citric acid also could enhance americium solubility. Solubility of americium (III) was increased by these organic ligands through formation of stable complexes. High formation constants for americium complexes of EDTA, HEDTA, hydroxyacetate, and citrate are known.(10)

RADIOELEMENT SORPTION SCREENING TESTS

Initial and final radionuclide concentrations and the resultant K_d values for the Plackett-Burman screening sorption tests are summarized in Appendix B in Tables B-1 through B-5 for cobalt, strontium, neptunium, plutonium, and americium, respectively. The fractions sorbed, shown in Tables B-6 through B-10, were used in the Plackett-Burman analysis. The resulting component factor effects and error data, which highlight components most important in influencing fraction sorbed, are summarized in Tables B-11 through B-15.

In Table 7, the factor effect data and error analyses for the Plackett-Burman sorption screening tests are summarized by ranking, in order of significance, the effects of the waste components on radionuclide sorption on the three different sediments. Inspection of Table 7 showed that the three sediments agreed well in their significance rankings. This similarity in rankings favored the conclusion that sorption mechanisms for the three sediments also were similar.

TABLE 7. Significant Components in Radioelement Sorption.*

Radioelement	Sediment	Component											
		$NaNO_3$	$NaNO_2$	NaOH	$NaAlO_2$	Na_2CO_3	Na_2SO_4	Na_3PO_4	NaF	HEDTA	EDTA	Hydroxy-acetic acid	Citric acid
Cobalt	L	-6	-4	+2	+3		+8	-7		-1	+5		
	P	-6	-4	+2	+3			-7		-1	+5		
	S	-5	-4	+3	+2		+7	-8	-9	-1	+6		
Strontium	L									-2	-1		
	P									-2	-1		
	S									-1	-2		
Neptunium	L			-2	-3		-5			-1	-4	-6	
	P			-2	-3		-5			-1	-4	-6	
	S			-2	-3					-1	-4	-5	
Plutonium	L	-5		-1	-2					-3	-4		
	P	-5		-1	-2					-3	-4		
	S			-2	-1	+5		+6		-3	-4		
Americium	L			+3						-1	-2		-4
	P			+2						-1	-3		
	S			+2				+4		-1	-3		

*Numbers indicate rank in importance; + indicates component increases sorption, - indicates component decreases sorption.

Cobalt

As shown in Table 7, almost all waste components affected cobalt sorption significantly. Restricting discussion to only those components significant at the 80% CL for all three sediments, it was shown that, in decreasing order of importance, HEDTA, NaOH, $NaAlO_2$, $NaNO_2$, EDTA, $NaNO_3$, and Na_3PO_4 were judged significant in these experiments.

The components HEDTA, $NaNO_2$, $NaNO_3$, and Na_3PO_4 decreased cobalt sorption. Most likely, HEDTA decreased cobalt sorption through complexation of Co^{2+}, forming a poorly sorbed anionic species such as $CoHEDTA^-$.(6) Similarly, $NaNO_2$ decreased cobalt sorption through formation of an anionic cobalt-nitrite complex. Nitrite decreased cobalt sorption on Hanford sediment at sediment pH (approximately pH 8).(11) Sodium nitrate also decreased cobalt sorption. In the case of $NaNO_3$, decreased cobalt sorption probably was caused by competition of the Na^+ with the cobalt ions for sorption sites, not by nitrate complexation of cobalt. While the neutral $Co(NO_3)_2^0$ complex is known, the formation constant is too low to affect cobalt sorption seriously.(12) Sodium ion competition also may have contributed to the negative effect of $NaNO_2$ on cobalt sorption. Phosphate and cobalt form the weak complex $CoHPO_4^o$; such a complex would decrease cobalt sorption.(6) Apparently, the low solubility compound $Co_3(PO_4)_2$ ($\log K_{sp} = -34.7$) was unimportant in determining cobalt solubility or sorption in the Hanford waste/sediment system.(6)

The components NaOH, $NaAlO_2$, and EDTA all increased cobalt sorption and/or precipitation. Sodium hydroxide may have precipitated Co^{2+} as $Co(OH)_2$.(6) Precise data on the solubility of Co^{2+} in the presence of aluminate were not found. However, the CRC Handbook of Chemistry and Physics describes cobalt aluminate as insoluble.(13) Alternatively, both NaOH and $NaAlO_2$ could react with the sediment minerals, removing Co^{2+} from solution by inclusion in or sorption on new mineral phases. Further studies are required to deduce the mechanism of NaOH and $NaAlO_2$ in increasing cobalt sorption.

The increase in cobalt sorption caused by EDTA was surprising in the light of the opposite observation made for HEDTA's effect on cobalt fraction sorbed. The compounds EDTA and HEDTA are structurally similar multidentate organic complexing agents. Both components were found to be statistically significant in increasing cobalt solubility as shown in Table 6. Both form anionic complexes with Co^{2+}.(6) Further studies are required to explain the unexpected effect of EDTA on cobalt sorption.

Strontium

Table 7 shows that only EDTA and HEDTA significantly affected strontium sorption. Both components reduced strontium sorption, probably through formation of poorly sorbed anionic complexes such as $SrEDTA^{2-}$ and $SrHEDTA^-$.(6)

Inspection of the strontium sorption data in Table B-7 in conjunction with the Plackett-Burman factor matrix in Table A-1 showed that only when both EDTA and HEDTA were absent (solutions number 5, 6, 7, 14, and 20) were the strontium fractions sorbed appreciably greater than 0.1. Within these five experiments for each sediment, the solutions having greater sodium ion concentrations (5, 6, and 7) had lower strontium sorption. It therefore appeared that sodium ion was competing with uncomplexed strontium for sorption sites. The competition of strontium with sodium for sorption sites has been observed previously for Hanford sediments at about pH 8.(11)

Neptunium

The neptunium sorption screening results summarized in Table 7 show that, in order of decreasing importance, HEDTA, NaOH, $NaAlO_2$, EDTA, and hydroxyacetic acid were judged significant at the 80% CL for all three sediments. All five components decreased neptunium sorption.

As discussed earlier, the likely solution species in high pH oxygenated solutions is $NpO_2(OH)_2^-$.(7) Sorption of the $NpO_2(OH)_2^-$ species on the sediment particles was probably through chemisorption. In chemisorption, the sorbed metal, such as neptunium, is bonded directly to the surface oxygen atoms of the mineral crystal matrix. To make the chemisorption bond, ligands coordinated on the metal, such as the hydroxide, must be displaced. Due to the anionic nature of the neptunium solution species and the negative surface charge of the sediment minerals at high pH, the mechanism of neptunium sorption is more likely chemisorption than simple electrostatic attraction as in ion exchange.

The components HEDTA, EDTA, and hydroxyacetic acid decreased neptunium sorption by forming stable complexes with the neptunium, thus tying up neptunium chemisorption bonding sites. As shown earlier, HEDTA and hydroxyacetic acid significantly increased neptunium solubility, probably through complex formation.

Several explanations can be offered to rationalize the decreased neptunium sorption observed with increased NaOH or $NaAlO_2$ concentration. Sodium hydroxide and $NaAlO_2$ may have decreased neptunium sorption through reaction with the sediment minerals. Many minerals undergo hydrolysis reactions or mineral transformations in the presence of high NaOH concentrations. The NaOH/sediment mineral reaction products may have had lower chemisorption capacity for neptunium. Sodium aluminate also could have reacted with the sediment, yielding products having lower neptunium sorption capacity. Alternatively, $NaAlO_2$ could have supplied hydrolyzed anionic aluminate species, such as $Al(OH)_4^-$, to the solution, which may have competed with $NpO_2(OH)_2^-$ for chemisorption sites and thus reduced neptunium sorption. Finally, both NaOH and $NaAlO_2$, by supplying hydroxide ion, may have stabilized the species $NpO_2(OH)_2^-$ and thus decreased the availability of neptunium for chemisorption. The mechanism of decreased neptunium sorption with increased NaOH or $NaAlO_2$ concentration clearly requires further study.

Plutonium

Plutonium sorption, as shown in Table 7, was reduced by NaOH, $NaAlO_2$, HEDTA, and EDTA. These four waste components were significant in reducing plutonium sorption at the 80% CL for all three sediments.

In the earlier discussion on plutonium solubility, it was shown that the likely uncomplexed plutonium solution species were hydrolyzed, singly charged anions. As was the case for neptunium, therefore, plutonium sorption probably occurred through chemisorption. Significantly, neptunium and plutonium shared the same first four waste components influential in their sediment sorption reactions. Neptunium's and plutonium's similarity in sorption behavior suggested that they had similar sorption reactions as well as solution species.

The components HEDTA and EDTA were found to decrease plutonium sorption through formation of stable complexes. The chelate complexes were unsusceptible to chemisorption reactions since the ligands occupied chemisorption bonding sites on the metal.

Sodium hydroxide and $NaAlO_2$ probably decreased plutonium sorption through mechanisms similar to those postulated for neptunium. Further work must be done to deduce the effects of NaOH and $NaAlO_2$ on plutonium sorption.

Americium

Components found significant at the 80% CL in influencing americium sorption for all three sediments were, in order, HEDTA, NaOH, and EDTA. The complexants HEDTA and EDTA decreased americium sorption through formation of the poorly sorbed complexes such as $AmHEDTA^0$ and $AmEDTA^-$.(6)

Sodium hydroxide increased americium sorption. Since NaOH did not decrease americium solubility significantly in the solubility screening tests, americium sorption probably was not increased due to precipitation. The cause of increased americium sorption with increased NaOH concentration will require further study.

RADIOELEMENT SORPTION DIFFERENCES FOR DIFFERENT SEDIMENTS

Data in Table 7 show that for the three sediments, components significant in affecting cobalt, strontium, neptunium, plutonium, and americium sorption were ranked similarly. Analysis of the sorption data revealed, however, that some sediments differed in their degree of radioelement sorption. The fraction sorbed values found in the Plackett-Burman screening tests were analyzed statistically to compare the sediments' radioelement sorption capacity. The fraction sorbed values are given in Appendix B in Tables B-6 through B-10. Statistical comparisons of the sorption data yielded the Students' t values given in Table 8.

As shown in Table 8, absolute values of t greater than 2.093 indicate differences in radioelement sorption among the sediments significant at the 95% CL. No significant differences in cobalt sorption were found for the three sediments. In addition, sediments L and P were found to be statistically indistinguishable in strontium, plutonium, and americium sorption. Sediments L and S were statistically different in degree of strontium, neptunium, plutonium, and americium sorption; so were sediments P and S for the same four elements. Sediments L and P were statistically different in neptunium sorption.

Using the findings given in Table 8, it was determined that the behavior of the three sediments could be represented by two sediments for strontium, plutonium, and americium sorption: either L and S or P and S, since L and P were equivalent. Therefore, only P and S were studied for determination of strontium, plutonium, and americium sorption prediction equations. It was decided, for the purposes of this study, that sorption prediction equations for neptunium would be determined for sediments P and S only. Due to limited resources, it also was decided that cobalt sorption prediction equations would not be determined at this time.

TABLE 8. Sediment Sorption Differences, Students' t Test.

Radioelement	Sorption differences for sediment								
	L-P			L-S			P-S		
	$\bar{d}$[a]	$S_d(\bar{d})$[b]	t[c]	$\bar{d}$	$S_d(\bar{d})$	t	$\bar{d}$	$S_d(\bar{d})$	t
Cobalt	0.00120	0.04107	0.131	0.01025	0.06161	0.744	0.00905	0.06164	0.657
Strontium	-0.00380	0.04661	-0.365	-0.09230	0.08349	-4.944[d]	-0.08850	0.05660	-6.992
Neptunium	0.02410	0.03560	3.027	-0.18915	0.08399	-10.072	-0.21275	0.08431	-11.285
Plutonium	-0.02375	0.05343	-1.988	-0.12105	0.12656	-4.277	-0.09780	0.11632	-3.760
Americium	0.01495	0.03664	1.825	-0.13820	0.11979	-5.159	-0.15315	0.11958	-5.728

[a] $\bar{d}$ = average difference between fraction sorbed for the 20 Plackett-Burman experiments.

[b] $S_d(\bar{d})$ = standard deviation of $\bar{d}$.

[c] $t = \dfrac{\bar{d}\sqrt{20}}{S_d(\bar{d})}$; at 95% CL for 19 degrees of freedom, t = 2.093.

[d] Underlined t values, greater than 2.093 in absolute values, represent sediments significantly different in their sorption behaviors.

RADIOELEMENT SORPTION PREDICTION EQUATIONS

Initial and final radionuclide concentrations and the resultant K_d and fraction sorbed values for the Box-Behnken sorption prediction equation experiments are given in Tables C-1 through C-4 for strontium, americium, neptunium, and plutonium, respectively. The K_d and fraction sorbed values, as well as log (K_d) values, were analyzed by the Box-Behnken technique to yield sorption prediction equations (see Appendix A). The sorption equations are summations of linear (x,y,z...) as well as interaction (xy,xz,yz...) and curvature (x^2, y^2, z^2...) terms in which the x,y,z... values represent concentrations of components judged significant in affecting sorption in the screening tests.

Thus, sorption prediction equations were generated for strontium in terms of sodium ion, HEDTA, and EDTA concentrations. Prediction equations for neptunium and plutonium sorption were derived based on NaOH, $NaAlO_2$, HEDTA, and EDTA concentrations. Americium sorption equations were based on NaOH, HEDTA, and EDTA concentrations. Prediction equations were generated for sorption of strontium, neptunium, plutonium, and americium on sediments P and S.

For each radioelement/sediment combination, three prediction equations were generated: K_d, fraction sorbed, and log (K_d). The goodness of fit for each equation was calculated by statistically comparing its predicted log (K_d) values with the experimental log (K_d) values. The equation yielding the lowest variance of error was judged to have the best fit of the data and was selected as the sorption prediction equation for each radioelement/sediment interaction.

The sorption prediction equations and their relative errors in predicting K_d values are given in Table 9. Judging from the relative errors presented in Table 9, the neptunium and plutonium sorption phenomena were quantified adequately by the prediction equations.

The derived equations were not as successful in predicting strontium and americium sorption. The poorer fit was due to the range of sorption values observed in the Box-Behnken experiments for these elements. Strontium K_d values ranged from 0.2 to 53 mL/g for sediment P and 0.8 to 95 for sediment S; roughly a 200-fold spread of

TABLE 9. Radionuclide Sorption Prediction Equations.

Radioelement/ sediment	Sorption prediction equation[a]	K_d relative error estimate, 1σ[b]
Sr/P	$\log(K_d) = -2.5498 + 89.84\,[\text{EDTA}] + 0.62\,[\text{Na}^+] - 24.8\,[\text{Na}^+]\,[\text{EDTA}]$	5.3
Sr/S	$\log(K_d) = 1.3266 - 16.66\,[\text{HEDTA}] - 34.28\,[\text{EDTA}] + 444\,[\text{HEDTA}]\,[\text{EDTA}]$	2.2
Am/P	$\log(K_d) = 1.8837 - 28.88\,[\text{HEDTA}] + 175.2\,[\text{HEDTA}]^2$	2.6
Am/S	$\log(K_d) = 2.0006 + 0.118\,[\text{NaOH}] - 26.82\,[\text{HEDTA}] + 158.4\,[\text{HEDTA}]^2$	1.9
Np/P	$\log(K_d) = 1.3047 - 0.115\,[\text{NaOH}] - 0.368\,[\text{NaAlO}_2] - 2.22\,[\text{HEDTA}] - 1.76\,[\text{EDTA}] + 0.6\,[\text{NaOH}]\,[\text{HEDTA}]$	1.14
Np/S	$\text{Fraction sorbed} = 0.9729 - 0.024\,[\text{NaOH}] - 0.147\,[\text{NaAlO}_2] - 2.86\,[\text{HEDTA}] - 3.36\,[\text{EDTA}] - 0.077\,[\text{NaOH}]\,[\text{NaAlO}_2] + 3.28\,[\text{NaAlO}_2]\,[\text{HEDTA}] + 43.2\,[\text{HEDTA}]\,[\text{EDTA}]$	1.13
Pu/P	$\text{Fraction sorbed} = 1.1437 - 0.143\,[\text{NaOH}] - 0.896\,[\text{NaAlO}_2] - 0.900\,[\text{HEDTA}] - 2.04\,[\text{EDTA}] + 0.720\,[\text{NaAlO}_2]^2$	1.26
Pu/S	$\log(K_d) = 2.0839 - 0.235\,[\text{NaOH}] - 0.768\,[\text{NaAlO}_2] - 4.98\,[\text{HEDTA}] - 8.40\,[\text{EDTA}] + 1.52\,[\text{NaAlO}_2]\,[\text{HEDTA}] + 98.4\,[\text{HEDTA}]\,[\text{EDTA}]$	1.25

[a] $$K_d = \left(\frac{\text{Fraction sorbed}}{1\text{g}}\right) \div \left(\frac{\text{Fraction in solution}}{5\text{ mL}}\right) = 5\left(\frac{\text{Fraction sorbed}}{1-\text{Fraction sorbed}}\right)\left(\frac{\text{mL}}{\text{g}}\right)$$

[b] To obtain ±1σ error bounds of K_d values, multiply and divide K_d values by the figure in this column. For example, the ±1σ error bounds for a K_d of 50 for americium on sediment P would be 50 x 2.6 = 130 and 50/2.6 = 19.

values. Similarly, americium K_d values had a greater than 200-fold range for sediment P; and a greater than 70-fold range for sediment S. Neptunium and plutonium K_d values on the other hand, had a 2.5- to 30-fold range.

It was evident from these data that the concentrations of the significant components affected strontium and americium sorption more severely than neptunium and plutonium sorption. Examination of the strontium sorption data in Table C-1 showed that if either HEDTA or EDTA were present, the K_d values remained approximately 1 to 2; with both complexants absent, the K_d became 50 or more. Similarly, the highest americium K_d values were observed when the complexants HEDTA and EDTA were both absent. It was not surprising, then, that the prediction equations had the greatest divergence from the data at the high K_d data points. The large differences between the predicted and actual values contributed strongly to the K_d relative error estimates given in Table 9. The fit of the prediction equations to the strontium and americium K_d data was substantially better at lower K_d values than the figures in Table 9 indicate.

Using the derived sorption prediction equations, radioelement distribution coefficients were estimated for several representative Hanford waste solution types. Four solution types were selected: dilute noncomplexed, dilute complexed, concentrated noncomplexed, and concentrated complexed. The dilute waste was defined as having low (-) concentrations of Na^+ (3.8$\underline{M}$), NaOH (1.0$\underline{M}$), and $NaAlO_2$ (0.0$\underline{M}$). The concentrated waste was defined as having high (+) concentrations of Na^+ (5.8$\underline{M}$), NaOH (4.0$\underline{M}$), and $NaAlO_2$ (0.5$\underline{M}$). The noncomplexed waste had low concentrations of both HEDTA and EDTA (0.0$\underline{M}$ each) while the complexed waste had high concentrations of HEDTA (0.1$\underline{M}$) and EDTA (0.05$\underline{M}$). The waste compositions are presented in Table 10.

The predicted radioelement distribution coefficients for the four waste solution types are presented in Table 11. As expected, the data show that sediment S sorbed the radioelements more strongly than sediment P. Also evident from Table 11 is the strong dependence of K_d on waste solution composition. For strontium and americium, K_d is more dependent on complexant (HEDTA/EDTA) concentration than on whether the waste is dilute or concentrated (i.e., whether the waste has low or high concentrations of sodium ion, NaOH, and $NaAlO_2$). On the other hand, neptunium and plutonium K_d values are more dependent on the waste concentration than on whether the waste is complexed.

TABLE 10. Composition of Hanford Waste Solution Types.*

Component	Concentration (M)			
	Dilute noncomplexed	Dilute complexed	Concentrated noncomplexed	Concentrated complexed
Na^+	3.8	3.8	5.8	5.8
NaOH	1.0	1.0	4.0	4.0
$NaAlO_2$	0	0	0.5	0.5
HEDTA	0	0.1	0	0.1
EDTA	0	0.05	0	0.05

*All other components at middle (o) concentrations as shown in Table 1.

It is important to note the magnitude of the changes in sorption as waste solution composition is varied. These changes can be seen easily in comparing the retardation factors, R, given in Table 11 for the radioelements with the four waste types. The retardation factor is the ratio of solution velocity in a flowing solution environment to the radioelement velocity (see Table 11). A retardation factor of 10, for example, means the radioelement moves one-tenth as fast as the solution carrying it. Comparison of retardation factors for strontium shows that changes in waste composition can change strontium migration rates by up to a factor of 40. Americium migration rates can change by a factor of 30, while neptunium migration rates can change by a factor of 13 with changes in solution composition. The plutonium migration rates can change by at least a factor of 25.

TABLE 11. Predicted Radionuclide Distribution Coefficients and Retardation Factors for the Hanford Waste Solution Types.

Radioelement/ sediment	Dilute noncomplexed		Dilute complexed		Concentrated noncomplexed		Concentrated complexed	
	K_d (mL/g)	R[a]	K_d (mL/g)	R	K_d (mL/g)	R	K_d (mL/g)	R
Sr/P	0.64	3.8	0.39	2.7	11	49	0.022	1.1
Sr/S	21	92	1.5	7.3	21	92	1.5	7.3
Am/P	76	330	5.6	25	76	330	5.6	25
Am/S	130	560	10	46	300	1300	24	100
Np/P	16	68	8.7	38	4.6	21	3.9	18
Np/S	93	400	12	54	9.3	41	6.8	30
Pu/P	∞[b]	∞	21	92	2.2	10	0.63	3.7
Pu/S	71	300	26	120	5.8	26	2.6	12

[a] $R \equiv$ retardation factor. The retardation factor is the ratio of the solution velocity to the radioelement velocity in a system of solution flow through a porous medium. The retardation factor $R = 1 + K_d (\rho/\phi)$ where ρ is bulk density of Hanford sediment ($\approx$1.65 g/cm^3) and ϕ is the fraction of void volume in the sediment ($\approx$0.38).

[b] The fraction sorbed value predicted by the Pu/P sorption equation was 1.0007.

These results indicate the severe effects waste composition can have on radionuclide sorption. These numerical results are presented, however, with the following limitations. First, these studies were done at only one radioelement concentration. Other radioelement concentrations no doubt would change the derived K_d values. Also, these results are only valid in the range of chemical component concentrations studied. Due to solubility limitations, investigations at higher concentrations were not undertaken. All experiments were conducted at room temperature; individual tank waste temperatures can be higher or lower. Sorption equilibria can change with reaction temperature. Sorption kinetics, which can affect migration rates severely, were not studied. Finally, no studies were done to determine the products of the reaction between Hanford waste and Hanford sediment. Such products may influence sorption reactions profoundly.

CONCLUSIONS

In this study, the effects of 12 Hanford waste solution components on the sorption of cobalt, strontium, neptunium, plutonium, and americium on three Hanford sediments were determined. The degree of sorption of strontium, neptunium, plutonium, and americium on two sediments was then quantified in terms of the concentrations of the significant components. Tentative information concerning the influence of the waste components on the radioelements' solubilities also was obtained during the sorption experiments. A number of general observations can be made based on the findings of this study.

First, for the three Hanford sediments studied, components significant in affecting radioelement sorption were ranked similarly. The similarity favors the conclusion that sorption mechanisms for the three sediments were also similar. In addition, one of the sediments had significantly higher strontium, neptunium, plutonium, and americium sorption than the other two.

The same four waste components influenced sorption reactions of neptunium and plutonium. The similarity in neptunium and plutonium sorption behavior suggests that these elements may be sorbed by similar mechanisms and may exist as similar species in solution.

As was suspected, the multidentate ligands, HEDTA and EDTA, significantly decreased sorption for strontium, neptunium, plutonium, and americium. For cobalt, HEDTA also decreased sorption while EDTA surprisingly increased sorption.

Sodium hydroxide was found to influence cobalt, neptunium, plutonium, and americium sorption significantly; it increased cobalt and americium sorption and decreased neptunium and plutonium sorption. Sodium aluminate increased cobalt sorption but decreased neptunium and plutonium sorption. The effects of NaOH and $NaAlO_2$ on sorption will require further study. Further studies to determine the products of reaction of Hanford waste with Hanford sediment minerals, as well as sorption studies for the waste/sediment reaction products, also are suggested.

Tentative solubility screening data indicated that HEDTA significantly increased the solubility of all radioelements studied while the related compound, EDTA, increased cobalt, strontium, and americium solubility. The bidentate ligand, hydroxyacetic acid, significantly increased neptunium, plutonium, and americium solubility. Citric acid also may have increased americium solubility significantly. The solubility screening data were not optimal for solubility studies because radioelement contents of most of the test solutions were not above saturation.

Finally, equations predicting log (K_d) or fraction sorbed values as arithmetic functions of waste chemical component concentrations were derived for strontium, neptunium, plutonium, and americium sorption on two Hanford sediments. The prediction equations offered satisfactory descriptions of the neptunium and plutonium sorption data. The prediction equations were less successful in predicting strontium and americium sorption parameters, especially at high sorption levels, due to the large range of sorption values observed for these elements.

Using the sorption prediction equations, K_d values were estimated for sorption of strontium, neptunium, plutonium, and americium on two sediments from four representative Hanford waste solution types. Retardation factor values derived from the resultant K_d values showed that radioelement migration rates could be altered by factors of 13 to 40 by changes in waste composition.

It must be noted in closing that the predictor equations are only valid within the range of test conditions studied in this report. Variations in radioelement and chemical component concentrations and reaction temperatures beyond the conditions specified in this report no doubt would change the derived K_d values seriously. Sorption kinetics and waste/sediment reaction products were not investigated in this study but also could influence K_d values in field conditions.

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APPENDICES

APPENDIX A

STATISTICAL METHODS

Statistical design of experiments obtains fast, accurate results with a minimum of time and effort, particularly when a large number of variables are of potential importance. Interpretation of the results is simplified, interactions among the variables can be seen, and reliability of the data can be assessed. Factorial designs allow estimation of the effects of several factors simultaneously. In a full factorial design, experiments are run for all combinations of ρ factors with ℓ levels per factor. The number of experimental runs (n) is

$$n = \ell^{\rho}$$

Two-level factorial designs ($\ell = 2$) are useful for a wide variety of problems. They are easy to plan and analyze; both continuous and discrete factors can be used; and they yield reliable models for response variables that have no strong curvature in the experimental region. However, when the number of factors is greater than ≈6, the number of runs required ($2^6 = 64$) may not be practical. In this case, an appropriate first step would be a screening design that identifies the significant variables.

Most screening designs are obtained by using a fraction of the 2^{ρ} factorial design. The Plackett-Burman design is a specific fraction that allows efficient estimation of the effects of the variables under study.* The most useful Plackett-Burman designs are for 12, 20, and 28 runs, which can reliably handle up to 7, 15, and 23 variables, respectively. There is obviously a tremendous reduction in the number of experiments compared with the 2^{ρ} factorial design. This great advantage does have drawbacks. Plackett-Burman designs do not provide estimates of the interactions between variables, nor do they estimate response-

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*R. L. Plackett and J. P. Burman, "The Design of Optimum Multi-Factorial Experiments," Biometrika 33, 305 (1946).

variable curvature. However, it should be remembered that the purpose of a screening design is to select a few variables from many so that the few can be investigated in detail. More detailed investigation can then be done by using a more complete factorial or a response surface design.

Response surface experimental designs allow simultaneous estimation of linear (x,y) as well as interaction (xy) and curvature (x^2, y^2) effects of several factors. Mathematical solutions to such designs are in the form of full quadratic polynomial equations. To provide data for estimating curvature, at least three factor levels, low (-), middle (o), and high (+), are required. Using the equation $n = \ell^\rho$, full 3-level designs require 27 and 81 experiments, respectively, for 3 and 4 factors. Performing the large number of experiments in a full 3-level design is often not practical. Instead, a subset of the full factorial design is used.

One type of subset of the full 3-level factorial design is the Box-Behnken design.* In each experiment of the Box-Behnken design, all but two of the factors are at the middle (o) level. The remaining two factors are varied, over four experiments, through the high/low permutations ++, +-, -+, and --; and the response measured. This process, repeated for all two factor combinations, results in $2(\rho^2-\rho)$ experiments. In addition, multiple determinations of the response at the experimental center point (the point of which all factors are at the middle level) are made. The multiple determinations are used to evaluate experimental error. Three determinations of the center point are made for both the 3 and 4 factor designs. The total number of experiments are then 15 and 27, respectively, for 3 and 4 factors, a substantial decrease from 27 and 81 experiments in the full 3-level designs.

The 20-run Plackett-Burman screening design was used in the sorption and associated solubility screening experiments reported in this document. The screening tests were designed to identify the waste components that affect solubility and sorption significantly. The design called for a set of 20 different test solutions. To make the

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*Box, G. E. and D. W. Behnken, "Some New Three Level Designs for the Study of Quantitative Variables," <u>Technometrics</u> <u>2</u>, 455 (1960).

20 test solutions, high (+) and low (-) concentration combinations of the 12 components, values of which are shown in Table A-1, were mixed according to the first 12 columns of the Plackett-Burman Factor matrix shown in Table A-2. For example, solution number 1 had high (+) concentrations of $NaNO_3$, $NaNO_2$, Na_2CO_3, Na_2SO_4, Na_3PO_4, NaF, Na_4EDTA, and citric acid and low (-) concentrations of NaOH, $NaAlO_2$, Na_3HEDTA, and hydroxyacetic acid. The remaining seven unassigned columns in Table A-2 provided the estimate of error, shown below.

TABLE A-1. Component Concentration Values.

Component	Concentration (M)		
	High (+)	Middle (0)	Low (-)
$NaNO_3$	2	1	0
$NaNO_2$	2	1	0
NaOH	4	2.5	1
$NaAlO_2$	0.5	0.25	0
Na_2CO_3	0.05	0.025	0
Na_2SO_4	0.01	0.005	0
Na_3PO_4	0.01	0.005	0
NaF	0.01	0.005	0
Na_3HEDTA	0.1	0.05	0
Na_4EDTA	0.05	0.025	0
Hydroxyacetic acid	0.1	0.05	0
Citric acid	0.03	0.015	0*

*The lower citric acid concentration was 0.005M for the americium sorption and solubility screening studies so that sufficient americium concentration would be present in the feed solution.

The sorption experiments were then conducted, the radionuclide concentrations determined, and the fractions sorbed evaluated. The factor effect for each column was evaluated; factor effects found for individual waste components in the assigned columns were compared statistically with factor effects in the unassigned columns to determine the significance of the waste component in affecting solubility or sorption.

Factor effects for sorption were calculated for each column in the Plackett-Burman matrix according to the formula

$$\text{Factor effect} = \frac{\Sigma \text{ fraction sorbed } (+) - \Sigma \text{ fraction sorbed } (-)}{10}$$

where Σ fraction sorbed (+) and Σ fraction sorbed (-) are the sums of the fraction sorbed values corresponding to the + and - symbols, respectively, for the column being considered. It is noted that for the assigned columns, the factor effect is merely the difference between the average fraction sorbed found when the relevent component is in high concentration and the average fraction sorbed found when that component is in low concentration. A similar equation was used to evaluate factor effects for solubility. For solubility factor effects, radionuclide concentrations were used in place of fractions sorbed.

The experimental error was calculated from the factor effects found for the unassigned columns 13 through 19. The Pooled Standard Deviation (PSD) was calculated according to the formula

$$\text{PSD} = \left(\frac{\sum_{i=13}^{19} [\text{Factor effect } (i)]^2}{7} \right)^{1/2}$$

where i represents the unassigned column numbers. Multiplying PSD by the appropriate 2-sided Students' t value for, in this case, seven degrees of freedom gave the error of the analysis. Components having absolute values of factor effects greater than the error were then judged to be significant, at the confidence level (CL) selected, in affecting solubility or sorption.

TABLE A-2. Twenty-Run Plackett-Burman Factor Matrix.

Solution number	Solution density (g/mL)	Variables												Unassigned factor						
		$NaNO_3$	$NaNO_2$	NaOH	$NaAlO_2$	Na_2CO_3	Na_2SO_4	Na_3PO_4	NaF	Na_3HEDTA	Na_4EDTA	Hydroxy-acetic acid	Citric acid	13	14	15	16	17	18	19
		1	2	3	4	5	6	7	8	9	10	11	12							
1	1.21	+	+	-	-	+	+	+	+	-	+	-	+	-	-	-	-	+	+	-
2	1.19	+	-	-	+	+	+	+	-	+	-	+	-	-	-	-	+	+	-	+
3	1.17	-	-	+	+	+	+	-	+	-	+	-	-	-	-	+	+	-	+	+
4	1.27	-	+	+	+	+	-	+	-	+	-	-	-	-	+	+	-	+	+	-
5	1.35	+	+	+	+	-	+	-	+	-	-	-	-	+	+	-	+	+	-	-
6	1.30	+	+	+	-	+	-	+	-	-	-	-	+	+	-	+	+	-	-	+
7	1.25	+	+	-	+	-	+	-	-	-	-	+	+	-	+	+	-	-	+	+
8	1.25	+	-	+	-	+	-	-	-	-	+	+	-	+	+	-	-	+	+	+
9	1.17	-	+	-	+	-	-	-	-	+	+	-	+	+	-	-	+	+	+	+
10	1.24	+	-	+	-	-	-	-	+	+	-	+	+	-	-	+	+	+	+	-
11	1.13	-	+	-	-	-	-	+	+	-	+	+	-	-	+	+	+	+	-	+
12	1.15	+	-	-	-	-	+	+	-	+	+	-	-	+	+	+	+	-	+	-
13	1.05	-	-	-	-	+	+	-	+	+	-	-	+	+	+	+	-	+	-	+
14	1.08	-	-	-	+	+	-	+	+	-	-	+	+	+	+	-	+	-	+	-
15	1.18	-	-	+	+	-	+	+	-	-	+	+	+	+	-	+	-	+	-	-
16	1.24	-	+	+	-	+	+	-	-	+	+	+	+	-	+	-	+	-	-	-
17	1.27	+	+	-	+	+	-	-	+	+	+	+	-	+	-	+	-	-	-	-
18	1.29	+	-	+	+	-	-	+	+	+	+	-	+	-	+	-	-	-	-	+
19	1.22	-	+	+	-	-	+	+	+	+	-	+	-	+	-	-	-	-	+	+
20	1.03	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-

The screening experiments showed that strontium sorption was significantly dependent on the concentrations of HEDTA and EDTA and possibly dependent on sodium ion concentration. Both neptunium and plutonium sorption were found to be significantly dependent on NaOH, $NaAlO_2$, HEDTA, and EDTA concentrations. Americium sorption was found to depend significantly on NaOH, HEDTA, and EDTA concentrations. Hence, the 3-variable (factor) Box-Behnken design was selected to quantify strontium and americium sorption; the 4-variable design was selected for neptunium and plutonium. The Box-Behnken designs shown in Tables A-3 and A-4 were used in conjunction with Table A-1 to generate test solutions for the 3- and 4-variable experiments, respectively.

Sorption experiments using the Box-Behnken test solutions then were conducted, the radionuclide concentrations determined, and the sorption parameters evaluated. The sorption parameters evaluated were fraction sorbed, K_d, and log (K_d). Values of the sorption parameters were input to regression analyses, producing coefficients for a quadratic best-fit polynomial. The polynomial evaluated the sorption parameter in terms of concentrations of significant components, the concentrations' cross-products, and the concentrations squared. In the case of americium, whose sorption was found to be significantly dependent on NaOH, HEDTA, and EDTA concentrations, the K_d sorption polynomial has the form

$$\begin{aligned}K_d = {} & C_0 + C_1\,[\mathrm{NaOH}] + C_2\,[\mathrm{HEDTA}] + C_3\,[\mathrm{EDTA}] + C_4\,[\mathrm{NaOH}] \\ & [\mathrm{HEDTA}] + C_5\,[\mathrm{NaOH}]\,[\mathrm{EDTA}] + C_6\,[\mathrm{HEDTA}]\,[\mathrm{EDTA}] + \\ & C_7\,[\mathrm{NaOH}]^2 + C_8\,[\mathrm{HEDTA}]^2 + C_9\,[\mathrm{EDTA}]^2\end{aligned}$$

Analogous sorption polynomials can be written for other sorption parameters in terms of the significant components' concentrations. In the regression analyses, the C_x coefficients were each evaluated for significance by comparison of their respective errors with a Students' t statistic. Only those coefficients found to be statistically significant were retained for the final sorption prediction equations.

TABLE A-3. Three-Variable Box-Behnken Design for Strontium and Americium.

Solution number	Strontium solution density (g/mL)	Americium solution density (g/mL)	Variable					
			Strontium*			Americium		
			Na^+	HEDTA	EDTA	NaOH	HEDTA	EDTA
1	1.28	1.28	+	+	o	+	+	o
2	1.26	1.26	+	-	o	+	-	o
3	1.18	1.18	-	+	o	-	+	o
4	1.17	1.17	-	-	o	-	-	o
5	1.28	1.29	+	o	+	+	o	+
6	1.26	1.28	+	o	-	+	o	-
7	1.18	1.18	-	o	+	-	o	+
8	1.18	1.18	-	o	-	-	o	-
9	1.24	1.24	o	+	+	o	+	+
10	1.22	1.22	o	+	-	o	+	-
11	1.22	1.25	o	-	+	o	-	+
12	1.21	1.22	o	-	-	o	-	-
13	1.22	1.24	o	o	o	o	o	o
14	1.22	1.24	o	o	o	o	o	o
15	1.22	1.23	o	o	o	o	o	o

NOTE: All other components at middle (o) concentrations as shown in Table A-1.

*For strontium, Na^+ concentration was varied by varying $NaNO_3$ concentration from 0M (-) to 1M (o) to 2M (+), thus making [Na^+] 3.8, 4.8, and 5.8M, respectively.

Using the significant C_x coefficients, sorption prediction equations were written for each respective sorption parameter: fraction sorbed, K_d, and log (K_d).

TABLE A-4. Four-Variable Box-Behnken Design for Neptunium and Plutonium.

Solution number	Np/Pu solution density (g/mL)	Variable			
		NaOH	$NaAlO_2$	HEDTA	EDTA
1	1.29	+	+	o	o
2	1.26	+	-	o	o
3	1.19	-	+	o	o
4	1.16	-	-	o	o
5	1.24	o	o	+	+
6	1.22	o	o	+	-
7	1.23	o	o	-	+
8	1.21	o	o	-	-
9	1.22	o	o	o	o
10	1.29	+	o	o	+
11	1.27	+	o	o	-
12	1.18	-	o	o	+
13	1.17	-	o	o	-
14	1.26	o	+	+	o
15	1.24	o	+	-	o
16	1.22	o	-	+	o
17	1.21	o	-	-	o
18	1.24	o	o	o	o
19	1.28	+	o	+	o
20	1.27	+	o	-	o
21	1.19	-	o	+	o
22	1.15	-	o	-	o
23	1.26	o	+	o	+
24	1.24	o	+	o	-
25	1.21	o	-	o	+
26	1.20	o	-	o	-
27	1.22	o	o	o	o

NOTE: All other components at middle (o) concentrations as shown in Table 1.

The experimental sorption log (K_d) values then were compared with the respective predicted log (K_d) values for each sorption equation and a variance of error value was calculated using the following equation:

$$\text{Variance of error} = \frac{\Sigma\,(\text{Experimental } K_d - \text{Predicted } K_d)^2}{\begin{matrix}\text{Number of experiments;} \\ \text{15 or 27}\end{matrix} - \begin{matrix}\text{Number of terms in} \\ \text{the sorption equation}\end{matrix}}$$

The sorption equation with the lowest variance of error then was selected as giving the best description of the experimental data.

APPENDIX B

SORPTION AND SOLUBILITY SCREENING TEST DATA

TABLE B-1. Sorption Screening Data for Cobalt.

Solution number	Initial ^{60}Co concentration (µCi/L)	Sediment L		Sediment P		Sediment S	
		Final ^{60}Co concentration (µCi/L)	K_d (mL/g)	Final ^{60}Co concentration (µCi/L)	K_d (mL/g)	Final ^{60}Co concentration (µCi/L)	K_d (mL/g)
1	126.4	126.3	-0.350	127.9	-0.391	117.7	-0.057
2	134.4	125.3	-0.020	138.0	-0.409	122.8	0.095
3	146.0	25.91	20.39	29.67	16.73	37.03	14.81
4	147.2	129.5	0.404	120.4	0.782	115.3	1.21
5	86.24	46.48	3.13	46.67	3.71	55.83	2.41
6	109.8	89.52	0.927	81.27	1.44	86.37	0.786
7	112.6	79.18	1.96	79.77	2.02	75.11	1.95
8	156.0	67.38	5.79	74.02	4.54	85.95	3.77
9	144.2	128.9	0.257	131.2	0.122	129.8	0.121
10	141.2	123.3	0.248	125.2	0.298	131.0	-0.036
11	145.4	138.0	-0.078	133.9	0.139	127.3	0.366
12	142.4	137.7	-0.169	140.7	-0.210	133.5	-0.053
13	154.8	150.8	-0.168	146.4	-0.049	146.1	-0.025
14	105.7	61.96	2.97	65.28	2.58	66.32	2.84
15	146.9	23.14	23.36	27.44	21.93	36.66	13.25
16	150.4	131.7	0.382	129.0	0.574	123.9	0.572
17	143.7	133.1	0.116	130.8	0.183	131.1	0.102
18	142.8	104.8	1.45	102.8	1.68	118.7	0.723
19	143.0	127.2	0.251	133.9	-0.033	132.5	-0.079
20	96.56	74.37	1.10	79.78	0.915	71.04	1.22

TABLE B-2. Sorption Screening Data for Strontium.

Solution number	Initial ^{85}Sr concentration (μCi/L)	Sediment L		Sediment P		Sediment S	
		Final ^{85}Sr concentration (μCi/L)	K_d (mL/g)	Final ^{85}Sr concentration (μCi/L)	K_d (mL/g)	Final ^{85}Sr concentration (μCi/L)	K_d (mL/g)
1	663.2	708.3	-0.885	662.1	-0.432	575.8	0.500
2	708.5	678.4	-0.145	676.8	-0.223	635.3	0.140
3	821.9	721.9	0.462	627.5	0.921	509.1	2.29
4	821.8	701.3	0.585	671.0	0.603	595.9	1.48
5	518.0	52.36	51.35	57.94	45.22	7.914	312.5
6	586.1	139.8	21.13	83.97	28.74	50.07	70.96
7	628.9	104.6	19.72	159.9	16.84	43.61	60.59
8	803.8	681.2	0.507	665.3	0.785	611.4	1.47
9	787.8	680.9	0.234	686.7	0.338	673.0	0.274
10	806.2	653.0	0.669	654.0	0.824	582.7	1.61
11	761.6	720.8	-0.076	691.1	0.208	615.3	0.871
12	785.4	705.3	0.272	709.0	0.242	637.3	0.765
13	757.8	647.2	0.599	648.7	0.496	614.6	0.945
14	617.8	6.449	493.3	7.725	487.1	7.358	402.4
15	755.6	668.2	0.286	660.3	0.117	559.5	1.24
16	756.1	689.0	0.133	693.2	0.064	652.2	0.328
17	758.2	706.1	0.057	759.1	-0.320	698.4	0.057
18	828.8	697.0	0.564	732.0	0.331	744.0	0.263
19	774.6	646.1	0.630	702.8	0.254	592.8	1.05
20	548.4	54.99	48.90	49.04	50.53	26.18	127.5

TABLE B-3. Sorption Screening Data for Neptunium.

Solution number	Initial ^{237}Np concentration (μCi/L)	Sediment L		Sediment P		Sediment S	
		Final ^{237}Np concentration (μCi/L)	K_d (mL/g)	Final ^{237}Np concentration (μCi/L)	K_d (mL/g)	Final ^{237}Np concentration (μCi/L)	K_d (mL/g)
1	3.021	0.9730	9.40	1.282	7.06	0.3548	32.86
2	3.506	2.430	1.62	2.702	0.947	1.805	3.60
3	2.976	2.204	1.75	2.274	1.12	1.853	3.06
4	2.736	2.124	1.09	2.059	1.13	1.777	2.40
5	2.954	1.741	2.95	1.906	2.48	0.8356	12.80
6	2.189	0.5896	12.43	0.6423	10.47	0.0865	117.5
7	3.657	2.266	2.43	2.316	2.06	1.623	6.17
8	3.506	2.304	1.89	2.419	2.12	1.774	4.66
9	3.308	2.381	1.76	2.358	1.83	1.648	4.59
10	3.514	2.327	1.80	2.314	2.03	1.412	5.42
11	3.720	1.393	6.93	1.863	5.01	0.9668	13.88
12	3.665	2.031	4.14	1.710	4.34	0.9186	13.11
13	3.836	2.079	3.62	2.315	3.75	1.003	11.90
14	3.162	0.1892	92.07	0.1982	75.23	0.0670	221.9
15	3.398	2.659	0.786	2.845	0.635	1.912	2.93
16	3.380	2.373	1.61	2.683	1.11	2.127	2.88
17	3.450	2.843	0.751	2.882	0.660	2.096	2.91
18	2.479	1.868	1.22	1.916	1.00	1.689	2.05
19	3.775	2.553	1.71	2.691	1.83	1.991	3.80
20	2.544	0.0445	295.50	0.0762	141.70	<0.0056	>2090.0

TABLE B-4. Sorption Screening Data for Plutonium.

Solution number	Initial ^{238}Pu concentration (μCi/L)	Sediment L		Sediment P		Sediment S	
		Final ^{238}Pu concentration (μCi/L)	K_d (mL/g)	Final ^{238}Pu concentration (μCi/L)	K_d (mL/g)	Final ^{238}Pu concentration (μCi/L)	K_d (mL/g)
1	6.695	0.8464	29.98	0.8570	34.50	0.9591	39.01
2	6.910	3.148	5.24	3.356	4.82	2.740	7.60
3	5.788	3.516	2.83	3.704	2.57	2.896	4.46
4	6.682	4.941	1.27	5.151	1.50	2.845	5.96
5	5.796	3.632	2.13	4.062	1.98	2.552	7.43
6	6.288	1.854	10.42	2.462	7.80	0.5059	49.28
7	6.427	1.875	10.15	2.295	12.07	2.505	7.15
8	6.825	4.166	3.16	3.067	5.68	1.853	11.16
9	6.594	2.912	6.52	2.937	5.17	2.960	5.26
10	7.104	3.818	3.61	3.430	4.99	2.843	7.92
11	7.108	0.7172	42.52	0.9221	33.95	0.6108	44.76
12	7.268	1.417	20.14	1.132	26.00	1.327	19.33
13	8.228	1.386	23.54	1.240	28.47	1.059	28.66
14	7.306	0.3610	94.93	0.3079	124.57	0.2105	144.90
15	6.827	4.578	1.79	4.346	2.84	3.028	5.16
16	7.229	3.805	4.03	3.319	4.86	2.636	8.49
17	6.678	3.630	4.03	3.438	4.06	2.612	6.62
18	5.647	4.271	1.12	4.192	1.43	3.460	2.60
19	6.922	3.595	4.21	2.753	6.53	1.843	9.64
20	3.776	<0.0814	>207.9	<0.0808	>193.5	<0.0816	>207.3

TABLE B-5. Sorption Screening Data for Americium.

Solution number	Initial ^{241}Am concentration (μCi/L)	Sediment L		Sediment P		Sediment S	
		Final ^{241}Am concentration (μCi/L)	K_d (mL/g)	Final ^{241}Am concentration (μCi/L)	K_d (mL/g)	Final ^{241}Am concentration (μCi/L)	K_d (mL/g)
1	0.5277	0.2875	4.02	0.3018	4.40	0.07353	28.63
2	0.5506	0.3329	2.85	0.3694	2.20	0.2738	5.91
3	0.5466	0.01552	148.4	0.01820	118.7	<0.000830	>3160
4	0.5301	0.1991	7.79	0.2127	6.68	0.07620	28.54
5	0.02968	<0.000862	>153	<0.000933	>169	<0.000961	>133
6	0.2578	<0.000955	>1080	<0.000886	>1670	<0.000849	>1300
7	0.3324	0.005157	337.1	0.005203	288.2	0.005098	439.1
8	0.5340	0.02473	99.62	0.03505	66.24	0.006657	380.4
9	0.5408	0.4216	1.11	0.4339	0.97	0.3392	2.47
10	0.5470	0.3031	3.25	0.2779	4.25	0.1870	10.84
11	0.5480	0.1224	15.93	0.1752	10.11	0.02983	72.02
12	0.5349	0.2014	6.87	0.2236	5.99	0.1459	11.98
13	0.5526	0.3122	3.22	0.3782	2.43	0.2176	5.70
14	0.3759	<0.000949	>1730	0.006412	306.1	<0.000868	>2350
15	0.5462	0.04348	48.62	0.05455	41.02	0.01493	186.9
16	0.5518	0.3077	3.43	0.2862	4.13	0.1637	10.36
17	0.5432	0.3485	2.03	0.3682	2.01	0.2601	4.68
18	0.5540	0.3367	2.95	0.3318	2.76	0.2044	9.45
19	0.5329	0.1773	9.52	0.1967	7.50	0.08603	24.61
20	0.09141	<0.000933	>470	<0.000955	>520	<0.000988	>481

TABLE B-6. Fraction Cobalt Sorbed in Screening Tests.

Solution number	Fraction sorbed for sediment		
	L	P	S
1	-0.075	-0.085	-0.011
2	-0.004	-0.089	0.019
3	0.803	0.770	0.748
4	0.075	0.135	0.194
5	0.385	0.426	0.325
6	0.156	0.224	0.136
7	0.281	0.288	0.281
8	0.537	0.476	0.430
9	0.049	0.024	0.024
10	0.047	0.056	-0.007
11	-0.016	0.027	0.068
12	-0.035	-0.044	-0.011
13	-0.035	-0.010	-0.005
14	0.373	0.340	0.362
15	0.824	0.814	0.726
16	0.071	0.103	0.103
17	0.023	0.035	0.020
18	0.225	0.251	0.126
19	0.048	-0.007	-0.016
20	0.181	0.155	0.196

TABLE B-7. Fraction Strontium Sorbed in Screening Tests.

Solution number	Fraction sorbed for sediment		
	L	P	S
1	-0.215	-0.095	0.091
2	-0.030	-0.047	0.027
3	0.085	0.156	0.314
4	0.105	0.108	0.229
5	0.911	0.900	0.984
6	0.809	0.852	0.934
7	0.798	0.771	0.924
8	0.092	0.136	0.227
9	0.045	0.063	0.052
10	0.118	0.141	0.244
11	-0.015	0.040	0.148
12	0.052	0.046	0.133
13	0.107	0.090	0.159
14	0.990	0.990	0.988
15	0.054	0.023	0.198
16	0.026	0.013	0.061
17	0.011	-0.068	0.011
18	0.101	0.062	0.050
19	0.112	0.048	0.173
20	0.907	0.910	0.962

TABLE B-8. Fraction Neptunium Sorbed in Screening Tests.

Solution number	Fraction sorbed for sediment		
	L	P	S
1	0.653	0.585	0.868
2	0.244	0.159	0.419
3	0.259	0.183	0.380
4	0.179	0.185	0.324
5	0.371	0.332	0.719
6	0.713	0.677	0.959
7	0.327	0.292	0.552
8	0.275	0.298	0.482
9	0.261	0.268	0.479
10	0.264	0.289	0.520
11	0.581	0.500	0.735
12	0.453	0.464	0.724
13	0.420	0.429	0.704
14	0.948	0.938	0.978
15	0.136	0.113	0.369
16	0.243	0.181	0.366
17	0.131	0.117	0.368
18	0.197	0.167	0.290
19	0.255	0.268	0.432
20	0.983	0.966	>0.998

TABLE B-9. Fraction Plutonium Sorbed in Screening Tests.

Solution number	Fraction sorbed for sediment		
	L	P	S
1	0.857	0.873	0.886
2	0.512	0.491	0.603
3	0.361	0.340	0.472
4	0.203	0.230	0.544
5	0.299	0.283	0.598
6	0.671	0.609	0.908
7	0.670	0.700	0.588
8	0.388	0.532	0.691
9	0.566	0.508	0.513
10	0.419	0.500	0.613
11	0.895	0.872	0.900
12	0.801	0.839	0.794
13	0.825	0.851	0.851
14	0.950	0.961	0.967
15	0.264	0.362	0.508
16	0.446	0.493	0.629
17	0.446	0.448	0.570
18	0.183	0.222	0.342
19	0.457	0.566	0.658
20	>0.977	>0.975	>0.976

TABLE B-10. Fraction Americium Sorbed in Screening Tests.

Solution number	Fraction sorbed for sediment		
	L	P	S
1	0.446	0.468	0.851
2	0.363	0.305	0.542
3	0.967	0.960	>0.998
4	0.609	0.572	0.851
5	>0.968	>0.971	>0.964
6	>0.995	>0.997	>0.996
7	0.985	0.983	0.989
8	0.952	0.930	0.987
9	0.181	0.163	0.331
10	0.394	0.459	0.684
11	0.761	0.669	0.935
12	0.579	0.545	0.706
13	0.392	0.327	0.533
14	>0.997	0.984	>0.998
15	0.907	0.891	0.974
16	0.407	0.452	0.674
17	0.288	0.287	0.484
18	0.371	0.356	0.654
19	0.656	0.600	0.831
20	>0.990	>0.990	>0.990

TABLE B-11. Factor Effects for Cobalt Sorption.

Factor	Factor effect values for sediments (fraction sorbed)		
	L	P	S
	Component		
$NaNO_3$	-0.083◒	-0.081◒	-0.109●
$NaNO_2$	-0.192●	-0.155●	-0.146●
NaOH	0.243●	0.261●	0.182●
$NaAlO_2$	0.216●	0.209●	0.194●
Na_2CO_3	-0.007	-0.009	0.028
Na_2SO_4	0.061^{0}	0.044	0.061◒
Na_3PO_4	-0.077◒	-0.076◒	-0.052^{0}
NaF	-0.036	-0.028	-0.049^{0}
Na_3HEDTA	-0.299●	-0.298●	-0.281●
Na_4EDTA	0.090●	0.085◒	0.074●
Hydroxyacetic acid	0.045	0.020	0.026
Citric acid	-0.008	0.012	-0.024
	Error		
95% CL	0.089	0.094	0.068
90% CL	0.071	0.075	0.054
80% CL	0.053	0.056	0.041

NOTE: Significant at: ● - 95% CL

◒ - 90% CL

0 - 80% CL

TABLE B-12. Factor Effects for Strontium Sorption.

Factor	Factor effect values for sediments (fraction sorbed)		
	L	P	S
	Component		
$NaNO_3$	0.023	0.026	0.034
$NaNO_2$	0.011	0.012	0.030
NaOH	-0.024	-0.026	-0.008
$NaAlO_2$	0.108	0.078	0.064
Na_2CO_3	-0.110	-0.087	-0.083
Na_2SO_4	-0.126	-0.133	-0.078
Na_3PO_4	-0.114	-0.109	-0.097
NaF	-0.065	-0.061	-0.059
Na_3HEDTA	-0.377●	-0.423●	-0.463●
Na_4EDTA	-0.459●	-0.439●	-0.434●
Hydroxyacetic acid	-0.075	-0.105	-0.091
Citric acid	0.060	0.068	0.049
	Error		
95% CL	0.328	0.298	0.252
90% CL	0.263	0.239	0.202
80% CL	0.196	0.178	0.151

NOTE: Significant at: ● - 95% CL

◐ - 90% CL

○ - 80% CL

TABLE B-13. Factor Effects for Neptunium Sorption.

Factor	Factor effect values for sediments (fraction sorbed)		
	L	P	S
Component			
$NaNO_3$	-0.064	-0.065	0.014
$NaNO_2$	-0.047	-0.060	-0.006
NaOH	-0.211●	-0.203●	-0.198●
$NaAlO_2$	-0.179●	-0.190●	-0.191●
Na_2CO_3	0.024	0.009	0.003
Na_2SO_4	-0.117^{0}	-0.140◐	-0.060
Na_3PO_4	0.082	0.070	0.053
NaF	0.026	0.020	0.032
Na_3HEDTA	-0.260●	-0.236●	-0.241●
Na_4EDTA	-0.152◐	-0.166●	-0.154●
Hydroxyacetic acid	-0.108^{0}	-0.110^{0}	-0.122◐
Citric acid	0.043	0.047	0.050
Error			
95% CL	0.168	0.165	0.139
90% CL	0.135	0.133	0.111
80% CL	0.101	0.099	0.083

NOTE: Significant at: ● - 95% CL
◐ - 90% CL
0 - 80% CL

TABLE B-14. Factor Effects for Plutonium Sorption.

Factor	Factor effect values for sediments (fraction sorbed)		
	L	P	S
Component			
$NaNO_3$	-0.070^{0}	-0.066^{0}	-0.043
$NaNO_2$	-0.017	-0.049	-0.002
NaOH	-0.381●	-0.338●	-0.169●
$NaAlO_2$	-0.228●	-0.257●	-0.220●
Na_2CO_3	0.013	0.000	0.063^{0}
Na_2SO_4	-0.021	-0.006	-0.044
Na_3PO_4	0.040	0.039	0.061^{0}
NaF	0.019	0.018	0.010
Na_3HEDTA	-0.147●	-0.136●	-0.138●
Na_4EDTA	-0.078◐	-0.068^{0}	-0.100●
Hydroxyacetic acid	0.030	0.019	-0.016
Citric acid	0.051	0.050	0.000
Error			
95% CL	0.091	0.085	0.081
90% CL	0.073	0.068	0.065
80% CL	0.055	0.051	0.049

NOTE: Significant at: ● - 95% CL
◐ - 90% CL
0 - 80% CL

TABLE B-15. Factor Effects for Americium Sorption.

Factor	Factor effect values for sediments (fraction sorbed)		
	L	P	S
	Component		
$NaNO_3$	-0.053	-0.031	-0.026
$NaNO_2$	-0.062	-0.059	-0.016
NaOH	0.124^{0}	0.147◐	0.125●
$NaAlO_2$	0.006	0.003	-0.040
Na_2CO_3	-0.038	-0.035	-0.014
Na_2SO_4	0.013	0.009	0.015
Na_3PO_4	0.016	-0.014	0.070^{0}
NaF	-0.073	-0.075	-0.011
Na_3HEDTA	-0.473●	-0.478●	-0.339●
Na_4EDTA	-0.149◐	-0.147◐	-0.078^{0}
Hydroxyacetic acid	0.021	0.021	0.022
Citric acid	-0.106^{0}	-0.075	-0.060
	Error		
95% CL	0.157	0.159	0.111
90% CL	0.126	0.127	0.089
80% CL	0.094	0.095	0.066

NOTE: Significant at: ● - 95% CL
◐ - 90% CL
0 - 80% CL

APPENDIX C

SORPTION PREDICTION EQUATION DATA

TABLE C-1. Sorption Prediction Equation Data for Strontium.

Solution number	Initial ^{85}Sr concentration (μCi/L)	Sediment P			Sediment S		
		Final ^{85}Sr concentration (μCi/L)	K_d (mL/g)	Fraction sorbed	Final ^{85}Sr concentration (μCi/L)	K_d (mL/g)	Fraction sorbed
1	22.05	19.11	0.261	0.043	15.17	1.274	0.189
2	22.69	18.34	0.944	0.159	16.02	1.489	0.229
3	21.44	17.53	0.676	0.118	15.10	1.191	0.178
4	22.08	18.63	0.706	0.122	11.46	2.778	0.335
5	20.79	19.50	-0.019	-0.003	16.55	0.662	0.114
6	21.26	16.31	0.895	0.130	14.96	1.213	0.194
7	21.11	17.62	0.458	0.078	14.78	1.517	0.231
8	21.40	15.89	1.354	0.213	12.67	2.511	0.331
9	22.86	19.92	0.605	0.104	16.27	1.319	0.204
10	21.04	16.69	0.918	0.155	14.82	0.947	0.137
11	21.11	18.72	0.308	0.058	16.86	0.803	0.138
12	21.82	1.858	52.844	0.914	0.9229	95.510	0.950
13	21.22	16.32	0.846	0.139	15.98	0.894	0.144
14	20.99	19.06	0.271	0.050	16.47	0.808	0.138
15	21.80	19.65	0.205	0.039	17.43	0.564	0.095

TABLE C-2. Sorption Prediction Equation Data for Americium.

Solution number	Initial ^{241}Am concentration (μCi/L)	Sediment P			Sediment S		
		Final ^{241}Am concentration (μCi/L)	K_d (mL/g)	Fraction sorbed	Final ^{241}Am concentration (μCi/L)	K_d (mL/g)	Fraction sorbed
1	0.5172	0.2109	7.747	0.607	0.08876	18.758	0.787
2	0.5111	0.03073	76.946	0.939	0.005700	316.304	0.984
3	0.4346	0.2380	4.167	0.454	0.09282	13.620	0.726
4	0.5153	0.1697	11.746	0.701	0.03783	48.582	0.905
5	0.5257	0.1869	7.506	0.598	0.07153	22.488	0.814
6	0.5378	0.1318	12.975	0.720	0.06765	30.544	0.859
7	0.5378	0.3038	3.873	0.436	0.09644	14.150	0.726
8	0.5166	0.2140	5.616	0.525	0.09792	16.737	0.767
9	0.5077	0.2250	5.555	0.526	0.1034	16.636	0.768
10	0.5198	0.2395	5.469	0.523	0.1113	14.580	0.742
11	0.5204	0.06695	40.986	0.892	0.02164	113.743	0.958
12	0.1494	<0.000944	>920	>0.995	<0.000982	>870	>0.994
13	0.5109	0.1744	7.724	0.604	0.07068	31.074	0.862
14	0.5094	0.1783	10.487	0.676	0.06576	27.734	0.846
15	0.5200	0.1965	8.159	0.621	0.07944	20.221	0.798

TABLE C-3. Sorption Prediction Equation Data for Neptunium.

Solution number	Initial ^{237}Np concentration (μCi/L)	Sediment P			Sediment S		
		Final ^{237}Np concentration (μCi/L)	K_d (mL/g)	Fraction sorbed	Final ^{237}Np concentration (μCi/L)	K_d (mL/g)	Fraction sorbed
1	5.580	2.619	4.140	0.446	2.035	6.055	0.538
2	5.506	2.320	6.445	0.564	1.556	11.741	0.702
3	5.464	1.995	8.686	0.635	1.317	15.056	0.751
4	5.299	1.688	10.025	0.668	0.9330	19.866	0.798
5	5.504	2.240	5.438	0.514	1.682	9.676	0.658
6	5.552	2.245	7.078	0.586	1.609	8.855	0.632
7	5.384	2.289	7.048	0.585	1.779	10.182	0.671
8	4.756	1.641	10.325	0.673	0.6458	31.376	0.863
9	5.523	2.047	9.294	0.650	1.512	11.149	0.689
10	5.475	2.556	4.437	0.466	1.955	7.261	0.590
11	5.444	2.299	5.633	0.528	1.670	9.162	0.645
12	5.318	1.772	9.427	0.654	1.353	14.113	0.739
13	5.448	2.049	8.974	0.642	1.159	14.816	0.745
14	4.971	2.091	5.608	0.527	1.628	9.385	0.653
15	5.630	2.143	6.182	0.547	1.706	9.532	0.654
16	5.556	2.155	9.567	0.653	1.535	10.393	0.672
17	5.597	1.827	7.926	0.608	0.7519	25.879	0.836
18	5.181	2.011	5.986	0.539	1.425	10.156	0.666
19	5.708	2.701	5.179	0.509	1.961	7.577	0.600
20	5.642	2.586	5.235	0.511	1.665	11.915	0.705
21	5.290	2.008	7.339	0.595	1.326	13.673	0.733
22	5.404	1.756	11.222	0.692	0.9925	19.098	0.792
23	5.084	2.342	4.523	0.471	1.975	8.673	0.634
24	5.565	2.193	5.998	0.541	1.728	10.715	0.683
25	5.518	1.917	8.941	0.642	1.489	10.584	0.675
26	5.680	1.779	9.837	0.662	1.568	15.456	0.755
27	5.556	2.116	6.493	0.562	1.682	12.232	0.711

TABLE C-4. Sorption Prediction Equation Data for Plutonium.

Solution number	Initial ^{238}Pu concentration ($\mu Ci/L$)	Sediment P			Sediment S		
		Final ^{238}Pu concentration ($\mu Ci/L$)	K_d (mL/g)	Fraction sorbed	Final ^{238}Pu concentration ($\mu Ci/L$)	K_d (mL/g)	Fraction sorbed
1	9.542	6.426	1.700	0.251	5.689	3.068	0.380
2	10.18	4.539	5.763	0.536	4.121	7.319	0.595
3	10.20	3.570	8.705	0.636	2.446	16.451	0.768
4	9.862	0.9631	41.772	0.893	1.161	29.920	0.855
5	10.04	5.229	3.843	0.434	3.059	8.191	0.613
6	9.520	4.416	4.948	0.496	3.382	8.958	0.643
7	10.12	4.874	4.457	0.469	3.124	8.965	0.639
8	9.888	2.379	14.639	0.745	1.199	30.496	0.858
9	10.18	4.291	5.296	0.509	3.353	8.156	0.617
10	9.376	6.571	1.401	0.214	5.788	2.912	0.368
11	9.770	6.619	2.251	0.309	5.197	3.981	0.444
12	10.51	3.504	10.516	0.678	2.141	20.199	0.802
13	10.55	2.384	18.965	0.792	1.977	23.196	0.824
14	9.896	5.650	2.947	0.368	3.633	7.214	0.590
15	9.869	5.584	3.339	0.401	3.204	7.777	0.603
16	10.24	2.961	10.536	0.677	2.712	11.640	0.698
17	10.12	3.025	10.583	0.679	2.936	15.014	0.749
18	10.32	5.718	4.316	0.461	3.840	7.652	0.605
19	9.900	6.398	1.857	0.263	5.779	3.160	0.388
20	9.914	6.584	2.274	0.313	3.496	6.321	0.547
21	10.44	3.913	9.143	0.646	2.436	14.256	0.740
22	9.609	1.641	23.292	0.823	1.350	26.398	0.840
23	9.952	5.461	3.686	0.425	3.545	5.827	0.522
24	9.936	5.073	4.117	0.451	3.475	5.884	0.523
25	10.56	3.436	9.566	0.657	2.529	12.710	0.715
26	10.40	3.282	11.252	0.693	1.771	23.525	0.825
27	9.838	4.911	4.970	0.499	3.295	8.492	0.629

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