

NUREG/CR-3712
PNL-5040

Radionuclide Migration in Groundwater

Annual Report for FY 1983

Prepared by J. S. Fruchter, C. E. Cowan, D. E. Robertson, D. C. Girvin,
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Pacific Northwest Laboratory
Operated by
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Prepared for
U.S. Nuclear Regulatory
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Manuscript Completed: April 1984
Date Published: December 1984

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NRC FIN B2380

EXECUTIVE SUMMARY

Staff at Pacific Northwest Laboratory for the past several years have been collecting the available data concerning radionuclide migration in the groundwater at a low-level disposal site and studying the physico-chemical processes that control the mobility of radionuclides in the groundwater. This report summarizes the results obtained and progress made during FY83.

Samples of the influent and groundwater were collected for analysis of the chemical composition and were pumped through the Battelle large volume water sampler (BLVWS) to identify the charge-forms of the migrating radionuclides. Most of the radionuclides in the influent water are removed during the residence time in the disposal basin and the trench probably by sorption onto the soil. Only trace amounts of several radionuclides have been observed to migrate to the seepage springs. The group of mobile radionuclides includes the isotopes of Co, Tc, I, Ru, Sb, Fe, and Ni, and the mobile forms are anionic and nonionic charge-forms. Typically, radionuclides that enter the disposal facility predominantly in particulate forms were observed to have higher total concentrations at the beginning of the trench than at the end, whereas radionuclides that enter the trench predominantly in soluble forms tended to be more homogeneously distributed. Data on the concentration of radionuclides in the soils from Well #1 and Well #3 and the concentrations of the radionuclides in the ambient groundwater were used to estimate in situ K_d values and the velocity of the advancing radionuclide front. The estimated in situ K_d values and the velocity of the radionuclide front were in accordance with the observed behavior of ^{60}Co , ^{90}Sr , ^{106}Ru , ^{125}Sb , and ^{137}Cs in the groundwater. Although the chemistry of the influent water was somewhat variable during the sampling period, the chemistry of the groundwater was quite consistent indicating an effective buffering system in the groundwater.

Samples of the influent and groundwater were analyzed by gas chromatography (GC) and gas chromatography-mass spectroscopy (GC-MS) for the presence of and the composition of the low molecular weight (MW) hydrophilic organic compounds. The low MW hydrophilic organic compounds identified in the influent and groundwaters consisted of carboxylic acids generally at part per billion (ppb) levels. Four classes of carboxylic acids, monocarboxylic acids, dicarboxylic acids, oxygenated acids, and aromatic acids, were identified in the water samples. These carboxylic acids represent bioorganic and microbial diagenesis products. The organic acids in the trench waters appear to arise from the rich biological activity of the trench muds. Although most of the carboxylic acids identified in the groundwater are similar to those present in the trench waters, a number of acids identified in the trench waters were not

found in the groundwater. These absent organic compounds include a number of small dicarboxylic acids and all of the oxygenated acids. The low MW organic compounds constitute only a small percentage of the total organic carbon in the water (0.05 to 2.6%) and no specific chelating compounds were identified in the water. These results suggest that if organic species contribute to radionuclide transport then the high MW organic compounds will be most important.

A potentially important mechanism for retardation or removal of radionuclides in groundwater is adsorption onto amorphous manganese and iron oxides and organic compounds that have a high affinity for cationic and anionic forms of radionuclides. A task was initiated in FY83 to experimentally determine the adsorption behavior, of ^{235}Np on the sediments at the site, to test the hypothesis that iron oxides dominate the adsorption behavior, and to test the alternate hypothesis that adsorption can be described as a linear combination of the adsorption on the individual sinks. The hydroxylamine hydrochloride extraction technique together with curve stripping and sequential regression of the quantities of manganese, iron, aluminum, and silica extracted versus time were used to estimate the composition and quantity of amorphous oxide phases in the soils. Three amorphous oxide phases with differing solubility and Fe:Al:Si composition were identified. Using the estimated quantity of amorphous iron oxides in the soil, it was determined that the Np adsorption data obtained to date on these low organic carbon soils are consistent with the hypothesis that amorphous iron oxide fractions of the amorphous oxides in the soil determine the adsorption behavior of the neptunyl oxy cation NpO_2^+ . Np[V] is strongly bound to amorphous iron and approaches complete reversibility within 24 hours. Oxidation state analysis of Np in soil suspensions is consistent with, but does not prove, the hypothesis that Np[V] adsorbed from solution exists as Np[V] while bound to the soils.

Geochemical modeling of the influent and groundwaters was conducted to identify the specific aqueous species of the radionuclides that are mobile in the groundwater and to identify solid phases that may be controlling the radionuclide concentrations in the groundwaters. The mobile species of the radionuclides are the anionic and nonionic oxy- and hydroxy complexes, although organic complexes may be important mobile species for iron, zinc, and cobalt. Those radionuclides that occur in the groundwater predominantly as the uncomplexed cation, such as cesium, cerium, and manganese, appear to be the most greatly retarded. The groundwaters are calculated to be in equilibrium with several solid phases that could be important for controlling the concentrations of trace elements and radionuclides. These solid phases include calcite, several aluminosilicates and ferrihydrite. Calcite and the feldspar appear to be dissolving in this system; however, the status of the aluminosilicates, and ferrihydrite need to be evaluated as soon as more accurate and more complete analyses of the groundwater composition are obtained.

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1. INTRODUCTION

Safe disposal of radioactive wastes is one of the challenges faced today by the nuclear industry. Information presented in this report will lead to better predictions of the behavior of long-lived radionuclides in buried nuclear waste.

Predicting the environmental behavior of long-lived radionuclides will be enhanced by a better understanding of the solute forms in which radionuclides are transported in groundwater. Radionuclide mobility in groundwater sometimes has been underestimated when based on laboratory K_d (distribution coefficient) values (Means et al., 1978; Coles and Ramspott, 1982). This is because laboratory-derived K_d values have been obtained using ideal soluble tracers and a physicochemical form of the mobile radionuclide in the laboratory that is probably different from that found in the field. Other parameters not considered in the K_d experiments are the effects of long-term leaching of soil-adsorbed radionuclides by ligands and complexing agents in the groundwaters.

We used the infiltrating water immediately below and surrounding a low-level disposal site in the identification and evaluation of the processes controlling radionuclide mobility for the purpose of developing the capability to predict radionuclide migration at potential commercial disposal sites. The specific objectives of this research project were 1) to identify the major inorganic and organic mobile species, 2) to identify and evaluate the factors that contribute to the mobility, and 3) to identify and evaluate the mechanisms that attenuate radionuclide concentrations. A deterministic geochemical model was used to interpret the data that could provide a basis for eventual prediction of radionuclide migration at other disposal sites. This data development and the understanding of the processes are necessary for identifying the limitations and validity of the available data and the research outline at potential sites that would permit valid application of our obtained results. The resulting information can be used in formulating guidelines for operating existing commercial low-level waste disposal sites, in determining essential requirements for remedial action and decommissioning, in determining long-term surveillance and maintenance requirements, and in developing criteria for selection of future burial sites.

Staff at Pacific Northwest Laboratory for the past several years have been collecting the available data concerning radionuclide migration in the groundwater at a low-level disposal site and studying the physicochemical processes that control the mobility of radionuclides in the groundwater. This report summarizes the results obtained and progress made during FY83. Previous activities and results are summarized in the annual reports (Robertson et al., 1981, 1983).

1.1 References

D. G. Coles, and L. D. Ramspott, "Migration of Ruthenium-106 in a Nevada Test Site Aquifer: Discrepancy between Field and Laboratory Results," Science 215, 1235-1236 (1982).

J. L. Means, D. A. Crerar, and J. O. Duguid, "Migration of Radioactive Wastes: Radionuclide Mobilization by Complexing Agents," Science 200, 1477-1481 (1978).

D. E. Robertson, A. P. Toste, K. H. Abel, and R. L. Brodzinski, Pacific Northwest Laboratory, "Annual Progress Report for 1981. Influence of the Physicochemical Forms of Radionuclides During Migration in Groundwaters," USNRC Report, 1981.

D. E. Robertson, A. P. Toste, K. H. Abel, and R. L. Brodzinski, Pacific Northwest Laboratory, "Radionuclide Migration in Groundwater. Annual Progress Report for 1982," USNRC Report NUREG/CR-3554, August 1983.

2. SITE

The disposal facility consists of an unlined basin and connecting trench that receives influent water containing low levels of fission and activation products and trace amounts of transuranic radionuclides (Figure 1). The influent water percolates through the soil and a small fraction of it emerges at seepage springs located some 260 meters from the trench. The disposal basin and trench are very efficient in retaining most of the radionuclides, but trace amounts of a number of radionuclides existing in mobile forms migrate in the groundwater from the trench to the springs. Before January 1983, the influent water samples were collected from a sampling line which tapped the influent water flow just before entering the disposal facility. In 1983, influent water was sampled at the inlet (manway #1 or #2) and the far end of the concrete covered trench (manway #10). Spring water was sampled from a well emplaced above the spring because the springs were often inaccessible. Three wells were installed in FY82 at distances of 30, 46, and 73 m from the trench to permit sampling of the groundwater between the trench and seepage springs (Robertson et al., 1983). Well #1 at 30 m and Well #3 at 73 m were used for sampling; the well at 46 m had a casing that would not accommodate the sampling pump.

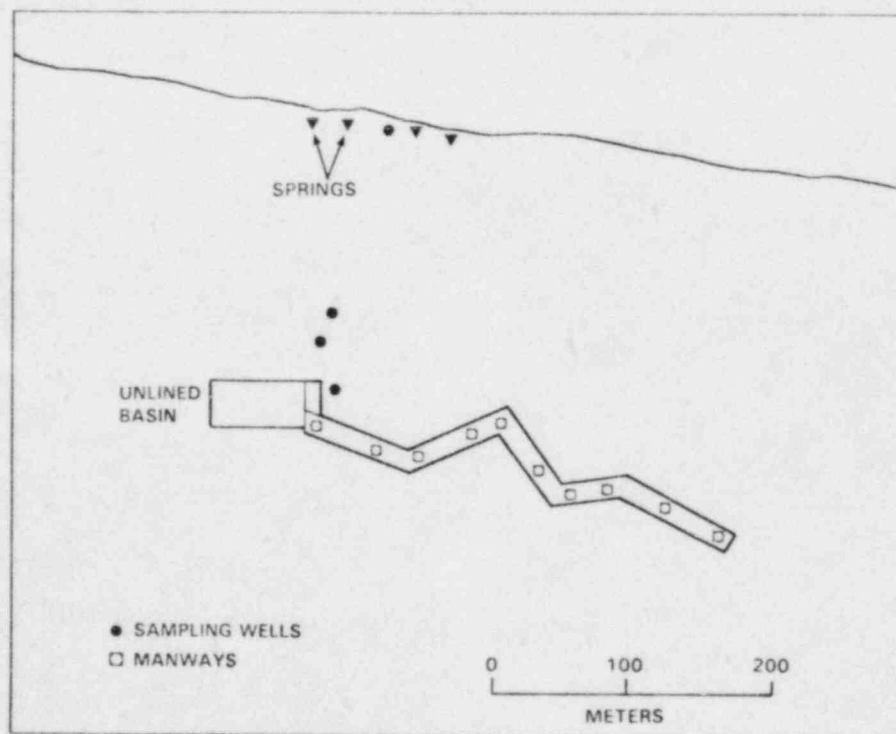


Figure 1. The low-level radioactive waste disposal facility showing locations of sampling points.

3. CHARGE-FORMS OF RADIONUCLIDES

Filtered groundwater from sampling points in and around the disposal site was pumped through the Battelle large volume water sampler (BLVWS) to identify the charge-forms of the migrating radionuclides. The cationic, anionic and nonionic charge-forms were absorbed by beds of cation resin, anion resin and activated aluminum oxide in the BLVWS. The mobile species and oxidation state of several radionuclides are being studied in detail using a variety of chemical manipulations.

3.1 Methods

Water samples were collected from the trench, two of the sampling wells and the seepage springs to determine the charge-form speciation of the mobile radionuclides and to characterize the chemistry of the waters. The details of the sampling procedure were published in the FY82 Annual Report (Robertson et al., 1983). In the trench, 10 to 20 L of water were pumped at a flow rate of 0.5 to 1.0 L/min through a 0.4-micron Nuclepore® membrane filter. The filtered water passed directly through the Battelle large volume water sampler (BLVWS), which consists of a 2.5-cm-thick x 15-cm-diameter bed of cation exchange resin (Dowex 50 x 8, H⁺ form, 200-400 mesh), a 2.5-cm-thick x 15-cm-diameter bed of anion exchange resin (Dowex 1 x 8, Cl⁻ form, 200-400 mesh), and a 0.7-cm-thick x 15-cm-diameter bed of fine-grained activated aluminum oxide. The resin beds are separated by fiber glass filters. The initial 0.4-micron Nuclepore filter removes radionuclides associated with particulates greater than 0.4 micron in diameter, the resins remove soluble cationic and anionic species and the activated aluminum oxide removes the nonionic species. The wells and springs were sampled with a larger version of the BLVWS employing dual resins and aluminum oxide beds because much greater volumes of water were needed to measure the lower radionuclide concentrations. Typically, 50 to 100 L of well water and 1000 to 5000 L of spring water were pumped at a flow rate of 1 to 8 L/min through the sampler.

Water samples were simultaneously collected for detailed measurements of temperature, pH, Eh, dissolved oxygen, alkalinity, dissolved organic carbon (DOC), major cations and anions, trace elements, and dissolved organic constituents. Temperature, pH, Eh, dissolved oxygen and alkalinity were measured onsite during the sampling. Aliquots of 0.4-micron-filtered water were frozen for later laboratory analysis of major anions by ion chromatography. For major cation and trace element analysis, a separate aliquot of filtered water was acidified by adding 20 mL of concentrated Ultrex hydrochloric acid per liter of water. For the sulfide analysis, an inline filtered sample of water was collected in a glass bottle containing a sufficient quantity of an antioxidant solution. Sulfide analysis was performed by a sensitive polarographic technique. Another aliquot of water was also collected and preserved by filtration for dissolved organic carbon constituent analysis (Section 4.0).

3.2 Physicochemical Speciation

The analytically determined radionuclide concentrations and charge-form distributions in the trench, well and spring waters samples collected in January, February, March, May, June, and September of 1983 are given in Appendix A.

Most of the radionuclides in the influent water are removed during residence in the disposal basin and trench probably by sorption onto the soil. Only trace amounts of several radionuclides have been observed to migrate to the seepage springs. The group of mobile radionuclides includes isotopes of Co, Tc, I, Ru, Sb, Fe, and Ni. Previous measurements of the particulate, cationic, anionic and nonionic charge-form distribution of the radionuclides in the trench, well, and spring waters indicated that the migrating radionuclides are primarily in anionic and nonionic forms. This was further confirmed by the distributions determined in these samples.

3.3 Radionuclide Concentrations in Trench Waters

The influent water was sampled at the inlet and end of the extension trench to determine differences that may exist in radionuclide concentrations and water chemistry at the opposite ends of the trench.

Typically, the radionuclides entering the disposal facility predominantly in particulate forms were observed to have higher total concentrations at the first or second manway compared to the last manway. For example, ^{59}Fe , found at manway #1 primarily in a filterable ($0.4\ \mu\text{m}$) particulate form (Figure 2), had a total ^{59}Fe concentration, particulate plus dissolved forms, that was 2 to 7 times higher than at the last manway. The particulate ^{59}Fe appears to settle out of solution during the several hours of transport through the trench. This same behavior was observed for other radionuclides that are predominantly in particulate forms in the influent water (e.g., ^{54}Mn , $^{58,60}\text{Co}$, ^{95}Zr , ^{95}Nb , $^{103,106}\text{Ru}$, and $^{141,144}\text{Ce}$).

The concentrations of those radionuclides entering the disposal facility predominantly in soluble forms tend to be more homogeneously distributed throughout the trench. For example, Figure 3 shows that the concentrations of ^{137}Cs in trench water samples collected from the first or second manway are nearly the same or slightly lower than for water samples collected from the last manway. Apparently, a small fraction of the soil-sorbed ^{137}Cs inventory in the trench is leached into solution during transport of the water from the inlet end of the trench to the terminal end. This same behavior is observed for other predominantly soluble radionuclides (e.g., ^{125}Sb , ^{131}I).

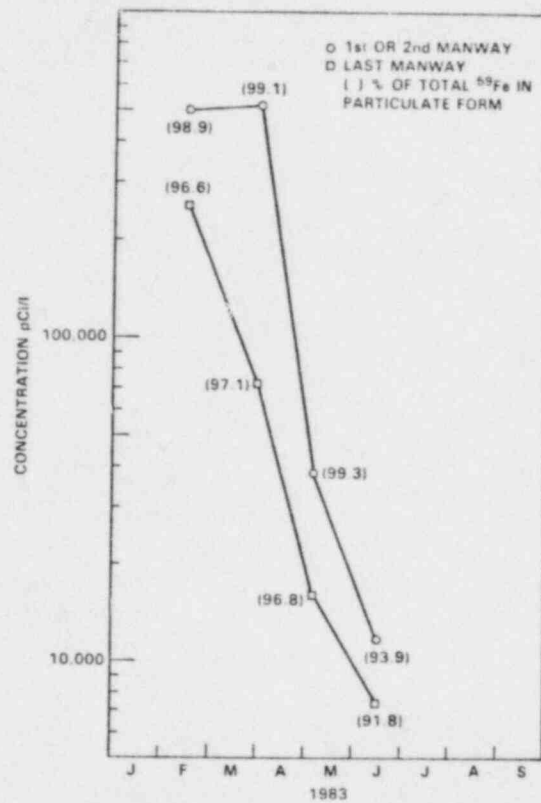


Figure 2. Comparison of ⁵⁹Fe concentrations found in opposite ends of trench.

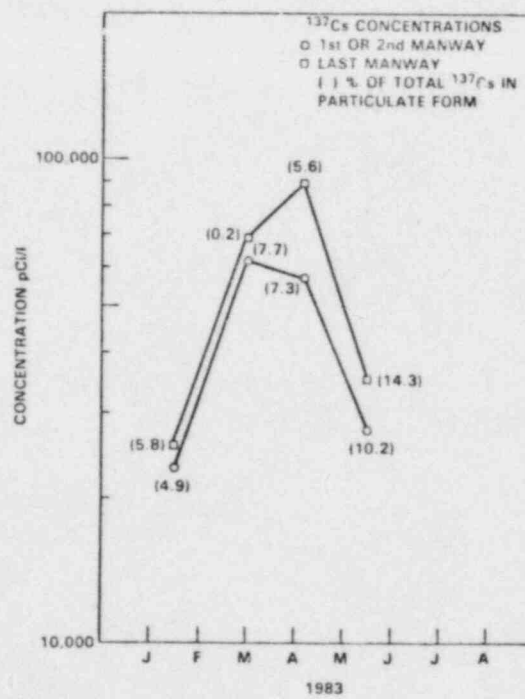


Figure 3. Comparison of ¹³⁷Cs concentrations found in opposite ends of trench.

3.4 Charge-Form Distribution

The disposal basin/trench system apparently removes most of the radionuclides in the discharged influent water because only trace amounts of several radionuclides have been observed to migrate to the wells and seepage springs.

Comparison of the distribution of particulate, cationic, anionic, and nonionic forms of the radionuclides in the trench, well, and spring waters indicates that the migrating radionuclides are primarily those in anionic and nonionic forms. The mobile radionuclides include isotopes of Co, Tc, I, Ru, Sb, and to a lesser degree of Fe and Ni. Radionuclides that occur predominantly in cationic charge-forms (^{54}Mn , ^{59}Fe , ^{95}Nb , ^{140}Ba , ^{140}La , ^{144}Ce , and ^{137}Cs) are present in the trench water. However, these radionuclides have not been observed to migrate to the springs; only trace amounts of these radionuclides are found in Well #1 in cationic forms, a distance of only 30 m from the trench.

The behavior of several of the more important radionuclides during their migration through the soil bank is discussed below.

3.4.1 Cobalt-60

Cobalt-60 has been shown to be one of the few radionuclides to migrate in groundwaters from a variety of low-level waste disposal sites (Means et al., 1978; Killey et al., 1984; Kirby, 1981; Eddy and Wilbur, 1980; Stone, 1982). This ubiquitous mobility appears to be related to the occurrence of the anionic cobalt species, possibly occurring as natural or man-made organic complexes.

The concentrations of ^{60}Co in the trench, Well #1 and spring waters in 1983 are listed in Appendix A, and are shown graphically in Figure 4. The numbers in parentheses in Figure 4 are the percent of the total "soluble" ^{60}Co represented by the anionic form(s).

The ^{60}Co in the trench water is typically partitioned between particulate and cationic, with traces of anionic species. In February, March, and May, relatively high concentrations of particulate ^{60}Co were discharged to the facility, whereas the discharge of soluble ^{60}Co during 1983 remained fairly constant at about $15,000 \pm 4000$ pCi/L. As the water moves from the trench to the springs, the selective removal of particulate and cationic ^{60}Co by the soil is clearly evident, resulting in the anionic ^{60}Co being the dominant charge-form in the spring water.

If it is assumed that no interconversion of cationic and anionic forms of ^{60}Co occurs during transport from the trench to the springs, the average decontamination factors for the cationic ^{60}Co for the soil column between the trench and Well #1 was 144 ± 42 and 514 ± 209 for the soil column between the trench and the springs (Table 1). The decontamination factor (DF) in this specific case is defined as the radionuclide concentration in the trench water divided by its concentration in the well or spring water. The DF represents the removal of radionuclides from solution in the groundwater by reactions with the soil phases,

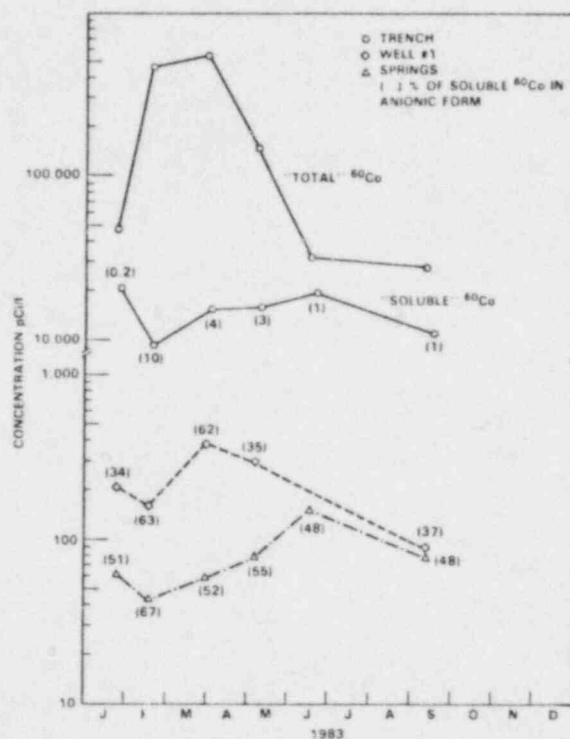


Figure 4. Temporal variability of ⁶⁰Co concentrations in trench, well, and spring waters.

Table 1. Comparison of decontamination factors for cationic and anionic forms of ⁶⁰Co.

	Trench to Well #1 Decontamination Factors		Trench to Springs Decontamination Factors	
	Cationic ⁶⁰ Co	Anionic ⁶⁰ Co	Cationic ⁶⁰ Co	Anionic ⁶⁰ Co
January	149	0.7	730	1.6
February	154	10	718	33
March	133	2.9	627	19
May	85	5.4	459	12
June	--	--	275	2.2
September	201	3.1	274	2.5
Average	144 ± 42	4.4 ± 3.6	514 ± 209	12 ± 12

dilution by groundwater, and also removal by radioactive decay as the groundwater moves from the trench to the wells or springs. If only the anionic species of ^{60}Co in the trench, well, and spring waters are considered, the average decontamination factors for these soil columns are only 4.4 ± 3.6 and 12 ± 12 , respectively. Thus, it is apparent that the anionic ^{60}Co is about 30 to 40 times more mobile in this groundwater system than the cationic ^{60}Co . During the January sampling, however, the anionic ^{60}Co was about 460 times more mobile than the cationic ^{60}Co .

Thermodynamic data indicate that Co^{2+} would be the dominant solution species with a much smaller contribution from $\text{Co}(\text{OH})^+$ (Rai and Serne, 1978). A possible form for the anionic ^{60}Co is $\text{Co}(\text{OH})_3^-$, although this specie is calculated to be approximately 8 orders of magnitude lower in concentration than Co^{2+} . The discrepancy may be due to organically complexed ^{60}Co , which would appear as a soluble anionic form in our separation scheme. Organically complexed anionic ^{60}Co has been identified and shown to be quite mobile in a contaminated groundwater plume at the Chalk River Nuclear Laboratories (Killey et al., 1984), and Means et al. (1978) have shown that ^{60}Co complexed with EDTA is relatively mobile in groundwaters near the Oak Ridge National Laboratory LLW disposal area.

The relatively high mobility of ^{60}Co observed in ground waters was unexpected since laboratory studies using inorganic ^{60}Co tracers indicated that cobalt was efficiently adsorbed onto soils. The affinity of a radionuclide for the soil phases in an aquifer is usually expressed in terms of an equilibrium distribution coefficient (K_d). In radioactive waste management, K_d 's have normally been measured by radiochemical analysis such that the K_d is determined as the ratio:

$$K_d = \frac{\text{dps/gm soil}}{\text{dps/mL solution}}$$

where dps stands for disintegrations per second of the radionuclide of interest. When K_d values for cobalt are measured in the laboratory using ^{60}Co tracers, the most likely chemical form of the ^{60}Co tracer would probably be Co^{2+} ion. Laboratory derived K_d values for cobalt on the order of 10^4 for weathered Conasauga shale and Nevada Test Site alluvium, and 10^3 for bentonite have been reported (Erdal et al., 1977; Means et al., 1978), and these probably reflect the adsorption of the cationic ^{60}Co tracers used in these experiments. Means et al. (1978) have shown that when cobalt is complexed with 10^{-5} M EDTA, the K_d for cobalt on the weathered shale is reduced by about four orders of magnitude compared with uncomplexed ^{60}Co .

At this site, it is possible to estimate in situ Kds for ^{60}Co at Well #1 and Well #3 by dividing the ^{60}Co concentration on the soils collected from these wells by the ^{60}Co concentration in the ambient groundwater. To estimate an in situ Kd requires several assumptions: first, that the water and soil are in equilibrium with respect to the radionuclides, and second, that the input of radionuclides is constant or at least that any fluctuations are averaged out in the trench. It is also necessarily assumed that the water entering the trench eventually travels to the spring via the groundwater intercepted by the well and does not travel by some other route. These calculations give "apparent" Kds of 35 and 9, respectively, for soils from Well #1 and Well #3. These "apparent" Kds are for a mixture of anionic and cationic ^{60}Co . Undoubtedly, the "pure" anionic ^{60}Co would have an even lower Kd. By using a more accurate "apparent" Kd (determined from in situ measurements) it is possible to estimate more precisely the transport rate for ^{60}Co migration from the trench to the wells or springs than could be achieved from using laboratory-derived Kd values. Using a simplified retardation equation, which ignores the effects of dispersion and diffusion:

$$V_{^{60}\text{Co}} = \frac{V_{\text{GW}}}{1 + \frac{P}{n} \cdot Kd}$$

where

$V_{^{60}\text{Co}}$ = velocity of advancing ^{60}Co front (m/d)

V_{GW} = velocity of the groundwater (60 m/d)

P = bulk density of site soil (1.9 gm/cm³)

n = porosity of site soil (0.3)

Kd = "apparent" distribution coefficient measured at Well #3 (Kd = 9)

we can compute that the velocity of the advancing ^{60}Co front would be about 1.1 m/d. This migration rate seems to be in accordance with the observed behavior of ^{60}Co at this facility.

Gee and Campbell (1980) measured an average batch Kd value of 3560 ± 810 for ^{60}Co on a similar soil contacted by five different water compositions. If a Kd of 3560 is used in the retardation equation, the estimated velocity of the advancing ^{60}Co front in this groundwater system would be only 0.0028 m/d. In the 19 years of operation of this facility the ^{60}Co would have moved only 19 m from the trench if a Kd of 3560 were used. Obviously, this is not the case. However, Gee and Campbell (1980) also showed that EDTA-complexed ^{60}Co had a Kd of only 1.2, which would make this organically complexed form very mobile if it were present in this groundwater system. Traces of EDTA and other complexing agents, as well as naturally occurring organic complexing materials have been identified in trench water and sediments (Robertson et al., 1983), thus it appears likely that either natural or man-made organic complexation is the mechanism producing the mobile anionic ^{60}Co species observed to migrate in the groundwater.

3.4.2 Strontium-90

Strontium-90 measurements made in January, 1983 of trench, well, and spring water samples indicate that the dissolved cationic form is always predominant (Table 2).

The ^{90}Sr concentration in Well #1 water (30 m from the trench) was actually higher than that observed in the trench, suggesting that gradual dissolution of the soil-adsorbed ^{90}Sr inventory in the trench was occurring or that the ^{90}Sr input to the trench was episodic. The soil bank between Well #1 and the springs is effective in removing over 90% of the ^{90}Sr that migrates to Well #1, and a spring water concentration of 1036 pCi/L was observed. The most probable form of the ^{90}Sr in these waters is Sr^{2+} , and ion exchange may be the principal mechanism of strontium adsorption on these soils (Rai and Serne, 1978).

Table 2. Physicochemical forms of ^{90}Sr in trench, well and spring waters in January 1983.

	pCi/L		
	Trench Water	Well #1 Water	Spring Water
Particulate	425 ± 41	--	--
Cationic	6,582 ± 740	16,485 ± 1,870	1,036 ± 117
Anionic	266 ± 25	71 ± 7	<0.1
Nonionic	47 ± 13	222 ± 3	0.45 ± 0.05

The observed ^{90}Sr transport behavior in this system appears to be more in keeping with predicted migration based on equilibrium distribution coefficients. Gee and Campbell (1980) reported an average ^{90}Sr Kd of 123 ± 13 for two similar soils in contact with groundwater of similar composition to this study site. Using this value in the retardation equation, the ^{90}Sr transport rate is predicted to be about 0.08 m/d. This would require about 10 years for the ^{90}Sr migrating front to travel from the trench to the springs, which appears to be in agreement with field observations.

3.4.3 Nickel-63

The physicochemical speciation and behavior of ^{63}Ni in this groundwater system is of interest because of its listing in 10 CFR 61 as a radionuclide of primary concern in low-level waste management. This concern is based on the long half-life of ^{63}Ni (100 year) and the fact that it is an abundant radionuclide in nuclear power plant low-level wastes.

Therefore, a suite of water and soil samples collected in June, 1981, were analyzed for ^{63}Ni to determine its concentrations and physicochemical forms. Water from the trench and the springs was sampled using the system previously described. The ^{63}Ni absorbed on the filters, resin, and Al_2O_3 was leached with hot concentrated HCl, radiochemically separated, electrodeposited on stainless steel discs, and counted in a specially constructed anticoincidence shielded, low-level beta proportional counter. The results of these analyses are shown in Table 3.

Table 3. Nickel-63 concentrations and chemical speciation, June 1981.

	<u>Trench Water</u> pCi/L	% of Total
Particulate	199.6 ± 28	59.1
Cationic	107.4 ± 38.4	31.8
Anionic	16 ± 16	4.7
Nonionic	15 ± 15	4.4
	<u>Spring Water</u> pCi/L	
Cationic	<0.8	
Anionic	<1.9	
Nonionic	1.2 ± 0.2	

The ^{63}Ni in the trench water was partitioned mainly between particulate and cationic forms. The cationic and anionic forms were nondetectable in the spring water, but a measurable fraction appeared in a nonionic form that was retained on the activated Al_2O_3 . The total decontamination factor afforded by the soil column was calculated to be 280. The trench sediments contained 6 to 30×10^4 pCi/g of ^{63}Ni and this sediment-sorbed ^{63}Ni appeared to be tightly bound.

3.4.4 Iron-59,55

The ^{59}Fe was predominantly present in the trench water in a particulate form, and during the 1983 sampling periods the particulate ^{59}Fe accounted for 91.1% to 99.3% of the total (see Figure 2 and Appendix A). The particulate ^{59}Fe concentrations entering the crib were highest during the February, March, and May samplings. The soluble cationic and anionic species during 1983 represented 0.3% to 3% and 0.3% to 2.8% of the total ^{59}Fe , respectively. Iron-59 has never been detected in the spring water, but trace quantities were found in Well #1 variably partitioned between particulate, anionic, and cationic forms. The anionic ^{59}Fe did not migrate to the springs, and DFs for Well #1 and the springs during 1983 ranged from 2.6×10^3 to 1.0×10^4 and 2.1×10^5 to 2.5×10^6 , respectively.

For the January 1983 samples, ^{55}Fe was removed from the filters, resins, and aluminum oxide with nitric acid, radiochemically purified, electro-deposited, and then counted on the intrinsic germanium detector. The results are shown in Table 4.

Table 4. Iron concentrations and physicochemical forms, January 1983.

	pCi/liter		
	Trench Water	Well #1 Water	Spring Water
Particulate	3,270 ± 16	--	--
Cationic	5,538 ± 22	8.01 ± 0.22	2.14 ± 0.06
Anionic	116 ± 0.1	1.96 ± 0.05	4.43 ± 0.18
Nonionic	40 ± 0.2	3.03 ± 0.07	<0.01

The ^{55}Fe behavior did not exactly duplicate that observed for ^{59}Fe during the January, 1983, sampling. The ^{55}Fe was present in the trench and in Well #1 water mainly in a dissolved cationic form. In the spring water, the ^{55}Fe was the only iron radionuclide observed and was 67% anionic. The difference in speciation between the ^{55}Fe and the ^{59}Fe may be due to the difference in half-lives and their relative inventories in the trench sediments. The relatively short half-life of ^{59}Fe (45 days) does not permit its extensive accumulation in trench sediments and its presence in the trench water reflects fairly recent additions. In contrast, the ^{55}Fe (2.7 years) inventory in the trench sediments must be much greater, allowing the ^{55}Fe more time to engage in possible dissolution and complexation processes which could result in somewhat different chemical forms compared to the ^{59}Fe .

3.4.5 Ruthenium-103,106

The concentrations and charge-form distributions of ^{106}Ru in trench, Well #1, and spring water samples collected between January and September, 1983, are tabulated in Appendix A and are graphically illustrated in Figure 5. Soluble ^{106}Ru concentrations in trench water ranged between 500 to 1150 pCi/L during the 1983 sampling period. Concentrations at Well #1 and the springs were about 5 to 10 times lower,

respectively than the trench water. Ruthenium tends to migrate rather easily through the soil bank between the trench and the springs. Ruthenium-103,106 present in the trench waters is variably distributed between particulate, cationic, anionic, and nonionic species. During migration in the groundwater, much of the cationic $^{103,106}\text{Ru}$ is preferentially sorbed to the soil in comparison to the anionic form(s). Once the $^{103,106}\text{Ru}$ reaches Well #1, the anionic form(s) in the well water generally accounts for about 60% to 90% of the total $^{103,106}\text{Ru}$. Based on thermodynamic data, the predicted soluble anionic $^{103,106}\text{Ru}$ species that would be prevalent in an oxidizing groundwater environment would be RuO_4^- , RuO_4^{2-} , RuO_4^0 , and $\text{Ru}(\text{OH})_2^{2+}$ (Rai and Serne, 1978). Table 5 shows the decontamination factors between the trench and the springs for "total" ^{106}Ru , soluble ^{106}Ru , and anionic ^{106}Ru species.

The anionic ^{106}Ru species obviously are the most mobile forms in this groundwater system, having an average decontamination factor between the trench and the springs of 6 ± 2 compared to DFs of 15 ± 8 and 57 ± 42 for soluble ^{106}Ru and total ^{106}Ru , respectively. Estimation of in situ Kds for ^{106}Ru at Well #1 and Well #3 is possible by dividing the ^{106}Ru concentrations in the soils collected from these wells by the ^{106}Ru in the ambient groundwater. These calculations give apparent Kds of about 10 and 6, respectively, for soils from Well #1 and Well #3. Using the

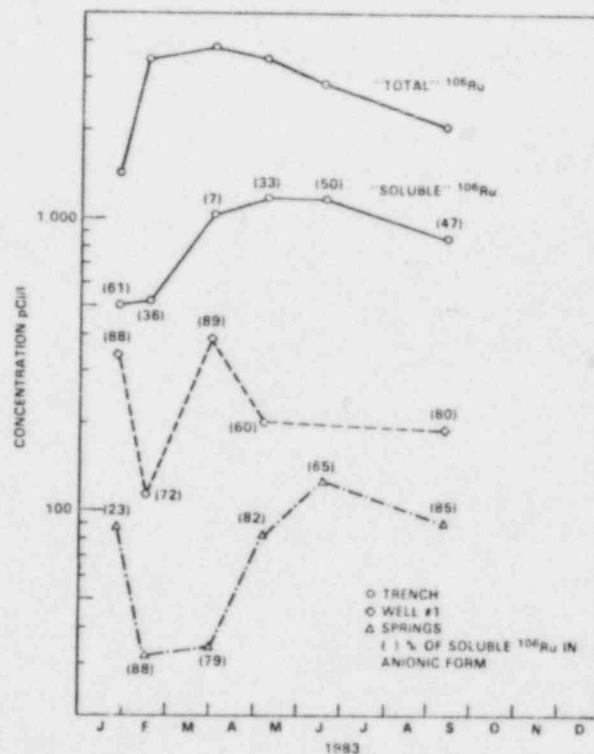


Figure 5. Temporal variability of ^{106}Ru in trench, well, and spring waters.

Table 5. Decontamination factors between the trench and springs for various forms of ruthenium-106.

Date	DF "Total" ^{106}Ru	DF "Soluble" ^{106}Ru	DF Anionic ^{106}Ru
January	32	11	<9
February	110	16	<9
March	110	30	3
May	43	14	6
June	23	9	7
August	23	10	5
Average	57 ± 42	15 ± 8	6 ± 2

retardation equation and an apparent Kd of about 6 for the anionic ^{106}Ru , this form of ^{106}Ru appears to be moving at a rate of about 1.6 m/d or slightly faster than the anionic ^{60}Co .

Laboratory batch sorption experiments on site soils produced Kd values from 40 to 752 within the pH range of 7 to 9 expected for groundwater (Ames and Rai, 1978). Using these Kds in the retardation equation would give ^{106}Ru transport rates ranging from 0.25 to 0.013 m/d, which would be at least 6 to 120 times slower than the observed transport rates.

Coles and Ramspott (1982) observed similar behavior for ^{106}Ru migrating in groundwater near the Cambrian nuclear test site at the Nevada Test Site. They concluded that a mobile fraction of the ^{106}Ru was moving at about the same velocity as the groundwater (based on comparison with the tritium transport rates), despite the fact that a laboratory-derived equilibrium Kd value of 976 would indicate that very little migration should have occurred. This again points out a problem associated with the unqualified use of laboratory-derived Kds and the need for field verification of radionuclide migration rates.

3.4.6 Antimony-125

Antimony (Sb) is very mobile in this groundwater system and exhibits very interesting charge-form distributions. Figure 6 illustrates the temporal variations in the ^{125}Sb concentrations in trench, Well #1, and spring waters during the 1983 sampling period.

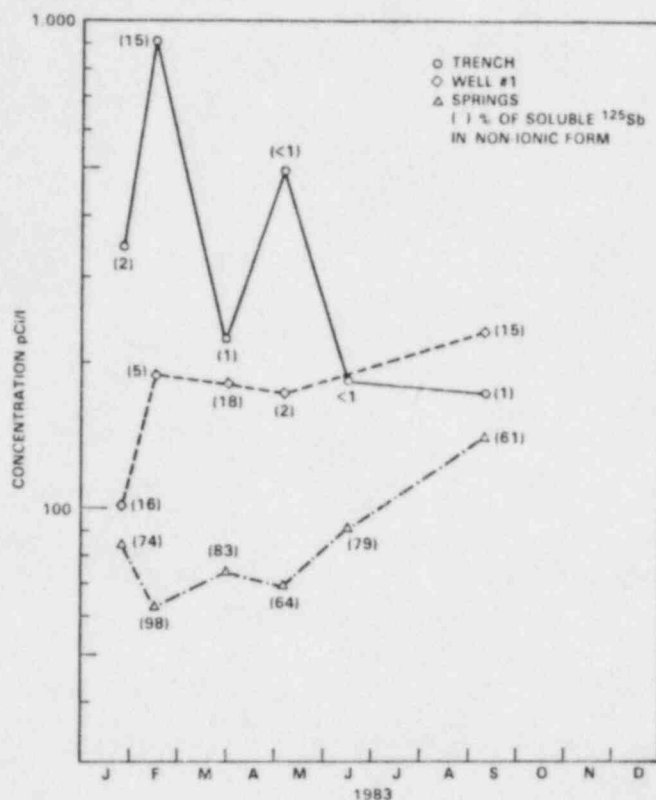


Figure 6. Temporal variability of ¹²⁵Sb in trench, well, and spring waters.

Antimony-125 in the trench water is normally present primarily in a soluble anionic form, with a very small fraction being present as a soluble nonionic species. During migration in the groundwater, the anionic ¹²⁵Sb appears to be converted to a nonionic form(s), which is also very mobile. At the springs the ¹²⁵Sb is typically partitioned as follows: $76 \pm 13\%$ nonionic, $23 \pm 13\%$ anionic, and generally less than 1% cationic. These observations are in agreement with the predicted chemical forms estimated to be present in oxygenated groundwater, namely the nonionic species HSbO_2^0 and $\text{Sb}(\text{OH})_3^0$ (Rai and Serne, 1978).

Decontamination factors between the trench and springs average only 6 ± 5 , 3 ± 2 , and 0.02 ± 0.03 for "total" ¹²⁵Sb, soluble ¹²⁵Sb and nonionic ¹²⁵Sb, respectively. The DF of 0.02 for nonionic ¹²⁵Sb obviously indicates that the nonionic form(s) of this radionuclide observed at the springs is formed by conversion of other species during transport from the trench to the springs.

In situ "apparent" Kds of 10 and 4 for Well #1 and Well #3 soils, respectively, were measured for the migrating ¹²⁵Sb. Assuming the lower

"apparent" K_d is most representative of the species migrating to the springs, a transport rate of about 2.4 m/d for the migrating ^{125}Sb species is estimated. This is probably a minimum rate of movement for the nonionic ^{125}Sb species.

3.4.7 Cesium-137

Cesium-137 is the least mobile of the radionuclides studied. During all sampling periods, the majority of the ^{137}Cs in the trench water was present as a soluble cation; however, as the water percolates through the soil the ^{137}Cs becomes tightly fixed onto the soils in the immediate vicinity of the trench. The ^{137}Cs was occasionally detectable in the well waters but never detectable in the spring water samples. Average decontamination factors of $3.4 \pm 2.6 \times 10^4$ to $>7 \pm 6 \times 10^5$ for the Well #1 and spring water, respectively, were afforded by the soil column during the 1983 sampling. The twenty-fold greater DF for ^{137}Cs at the springs is simply due to the fact that a much larger water sample was taken at the springs, thereby increasing the detection limit by twenty-fold. The ^{137}Cs presumably enters the trench water as soluble Cs^+ ion and the principal retention mechanism on soils is expected to be cation exchange on the clay minerals (kaolinite, montmorillonite, and illite) present in these soils (Rai and Serne, 1978).

The actual transport behavior of ^{137}Cs at this site is in accordance with predicted behavior. An average batch K_d of 2380 ± 1000 for ^{137}Cs adsorption onto site soils has been measured (Gee and Campbell, 1980). Using this value in the retardation equation gives a transport rate prediction of 0.0042 m/d. After 19 years of usage of this system the ^{137}Cs would have migrated only about 29 m from the trench. Field observations suggest that this may be a reasonable transport rate, since only very trace amounts of ^{137}Cs have been detected in Well #1 water (1-4 pCi/L) and Well #1 soil (<0.1 pCi/g) collected from below the perched water table. Well #1 is located approximately 30 m from the trench/crib. Cesium-137 has never been detected in water at the springs, a distance of about 250 m from the trench. Thus, the ^{137}Cs appears to be strongly sorbed onto the soil in the immediate vicinity of the trench, and is one of the least mobile of the radionuclides studied.

3.4.8 Iodine-129,131

Iodine-131 (8.04 days) can be useful tracer of the long-lived ^{129}I (1.59×10^7 year), which has been shown to be mobile in slightly contaminated groundwaters (Barracough et al., 1981; Eddy and Wilbur, 1980). The radioiodine in the trench, well, and spring waters occurs

predominantly in soluble anionic forms, and appears to be quite mobile in this system (see Appendix A). Further studies (Robertson et al., 1983) have recently shown that about 42% to 48% of the soluble anionic ^{131}I is in the form of iodide (I^-), and about 49% to 57% occurs presumably as an organically bound anionic complex.

The concentrations of the short-lived ^{131}I in the influent water to the crib are highly variable and for this reason it is difficult to ascertain decontamination factors for the soil column between the trench and Well #1 or the springs. The input of ^{129}I to the crib is probably more uniform with time, and its measurement could give a better estimate of the decontamination factor afforded by the soil column between the trench and the springs. The January 1983, samples of resins and aluminum oxide were subsequently analyzed for ^{129}I . The results are shown in Table 6.

The charge-form distribution of the ^{129}I is essentially the same as for ^{131}I , i.e., predominantly anionic. The ^{129}I concentrations were very low in the trench, well, and spring water samples, and a decontamination factor of about 40 was afforded by the soil column between the trench and the springs. This is approximately the DF observed for ^{131}I when accounting for losses due to radioactive decay.

Table 6. Iodine-129 concentrations and charge-forms in trench, well, and spring waters, January 1983.

	Concentration, pCi/L		
	Trench Water	Well #1 Water	Spring Water
Particulate	<0.06	--	--
Cationic	<0.08	<0.04	0.003
Anionic	0.30 ± 0.10	0.23 ± 0.04	0.0068 ± 0.001
Nonionic	<0.04	<0.01	<0.001

3.4.9 Niobium-95

Niobium-95 is a convenient tracer in this system for the much longer-lived ^{94}Nb . Niobium-94 is of concern in low-level waste management

because it is calculated to be a major constituent in aged neutron-activated metal components of reactor pressure vessel internals. Based on the results to date in this groundwater system, the ^{95}Nb does not appear to be very mobile. The input of ^{95}Nb to the trench during the 1983 sampling periods is shown in Figure 7. In the trench water sampled in 1983, the ^{95}Nb was partitioned as follows: 51 to 94% particulate, 2% to 36% cationic, and 5% to 13% anionic. The trench water samples collected in January, June, and September contained predominantly cationic ^{95}Nb , whereas the February, March, and May trench water samples contained predominantly anionic ^{95}Nb species. In water from Well #1, the ^{95}Nb in the anionic state accounted for 18% to 98% of the total (see Appendix A). During the ensuing movement from Well #1 to the springs, the soluble cationic and anionic ^{95}Nb were removed by absorption on the soil, and ^{95}Nb was usually not detectable in water from the springs. The DF factors for Well #1 and the springs ranged from 180 to 14,000 and 1.4×10^5 to $>1.0 \times 10^6$, respectively.

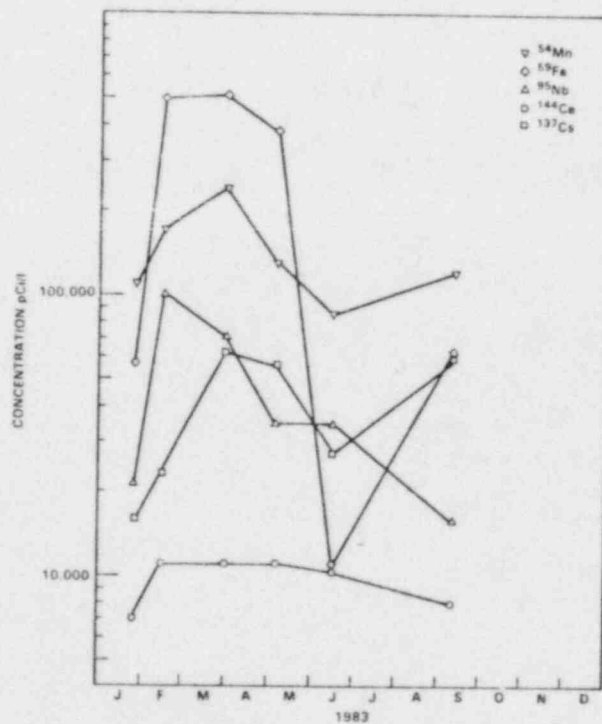


Figure 7. Temporal variability of several radionuclides in trench water sampled near the inlet to the trench.

3.4.10 Manganese-54

In the trench water the ^{54}Mn typically occurs primarily as cationic and particulate forms (see Appendix A). Input to the trench during the 1983 sampling periods is shown in Figure 7. During the 1983 sampling of Well #1, traces of ^{54}Mn were detected in a mixture of cationic and anionic forms. Neither of these forms of ^{54}Mn migrated to the springs. The DFs for Well #1 and the springs during 1983 ranged from 2.6×10^3 to 1.1×10^5 and $>1.1 \times 10^6$ to $>3.9 \times 10^6$, respectively. The dominant solution species of manganese in shallow oxygenated aquifers is probably Mn^{2+} , with smaller amounts of $\text{Mn}(\text{HCO}_3)^+$ and MnSO_4^0 present (Morgan, 1967).

3.4.11 Other Radionuclides

A number of other radionuclides of lesser significance in the management of low-level wastes are also present in this system. Cobalt-58 is a shorter-lived activation product formed by an (n,p) reaction with stable nickel. Its behavior relative to ^{60}Co , which is formed by an (n,r) reaction with cobalt, is often slightly different. This may be due to its formation from a different parent material, to kinetic differences, and/or to differences in half-lives that result in a much greater inventory of ^{60}Co in the trench sediments relative to ^{58}Co . If microbial or very slow geochemical processes are affecting the chemical speciation of the radiocobalt, then these effects would probably have a more pronounced effect on the longer-lived ^{60}Co .

Zirconium-95 is the precursor of ^{95}Nb and its behavior is somewhat similar. Zirconium-95 is present in the trench water mainly in particulate form, with the anionic form being the dominant soluble species. In Well #1, trace amounts of ^{95}Zr were observed, occurring from 38% to 97% as an anionic form. The ^{95}Zr has only occasionally been detected in the spring water, giving a DF ranging from 6.7×10^4 to $>3.7 \times 10^5$. Thermodynamic data (Rai and Serne, 1978) suggest that $\text{Zr}(\text{OH})_5^-$ would be the dominant solution species with smaller amounts of $\text{Zr}(\text{OH})_4^0$ and $\text{Zr}(\text{OH})_3^+$, which appears to be in good agreement with the field observations.

Barium-140 is often present in the trench water and occurs primarily as a cation. A trace of ^{140}Ba was detected in an anionic form in Well #1 during the January sampling. It has never been detected at the springs. Its short half-life does not make this an important radionuclide in waste management.

Several rare earth radionuclides, ^{140}La , ^{141}Ce , ^{144}Ce , and ^{154}Eu , are present in the trench water, mainly in particulate forms. The rare

earths could generally not be detected in well or spring waters, indicating efficient retention by the soil.

Antimony-124 exhibits behavior similar to the longer-lived ^{125}Sb in the well and spring water. However, the ^{124}Sb appears to be associated with particulates in the trench water to a greater degree than the ^{125}Sb . This may be due to the fact that the ^{124}Sb is a relatively short-lived neutron activation product and the ^{125}Sb is a longer-lived fission product.

3.5 Chemistry Of Trench And Groundwater

During the sampling for radionuclide charge-form measurements, field analyses of pH, Eh, temperature, dissolved oxygen, and alkalinity were conducted. Three separate aliquots of filtered water samples were taken for the following analyses: 1) a 250 mL sample of unacidified water for anion analysis by ion chromatography, 2) a 250 mL sample preserved with 25 mL of antioxidant solution for sulfide analyses by a polarographic method, and 3) a 2 L sample acidified with 20 mL of Ultrex HCl for analyses of trace elements.

The results of these measurements are given in Appendix B and the concentrations of several important constituents are plotted versus time in Figure 8. The chemistry of the influent water to the trench was somewhat variable during the 1983 sampling period. As shown in Figure 8a, the pH of the influent water ranged from 7.20 to 9.25, whereas the Well #1 and spring water pH consistently remained near 8.0. Obviously, the 30 m of soil between the trench and Well #1 provide an efficient buffering system for the percolating ground water.

The Eh of the trench water also showed considerable variability, ranging from +155 mV to +405 mV. The Well #1 and spring waters tended to be more oxidizing than the trench waters, and ranged from +300 mV to +430 mV and +275 mV to 435 mV, respectively. Normally, the Eh of the trench water at the influent and terminal ends of the trench was similar, but during the May, 1983, the water sampled at the first manway had an Eh of +155 compared to +315 for water sampled from the last manway. It is not known if this was due to rapid fluctuations of the Eh in the influent water, or if in situ changes occurred in the trench.

The temperature of the trench water remained fairly uniform, ranging between 17 to 23°C, whereas the Well #1 and spring waters showed a temperature increase that correlated with the summer months. The dissolved oxygen content of the waters generally showed an expected

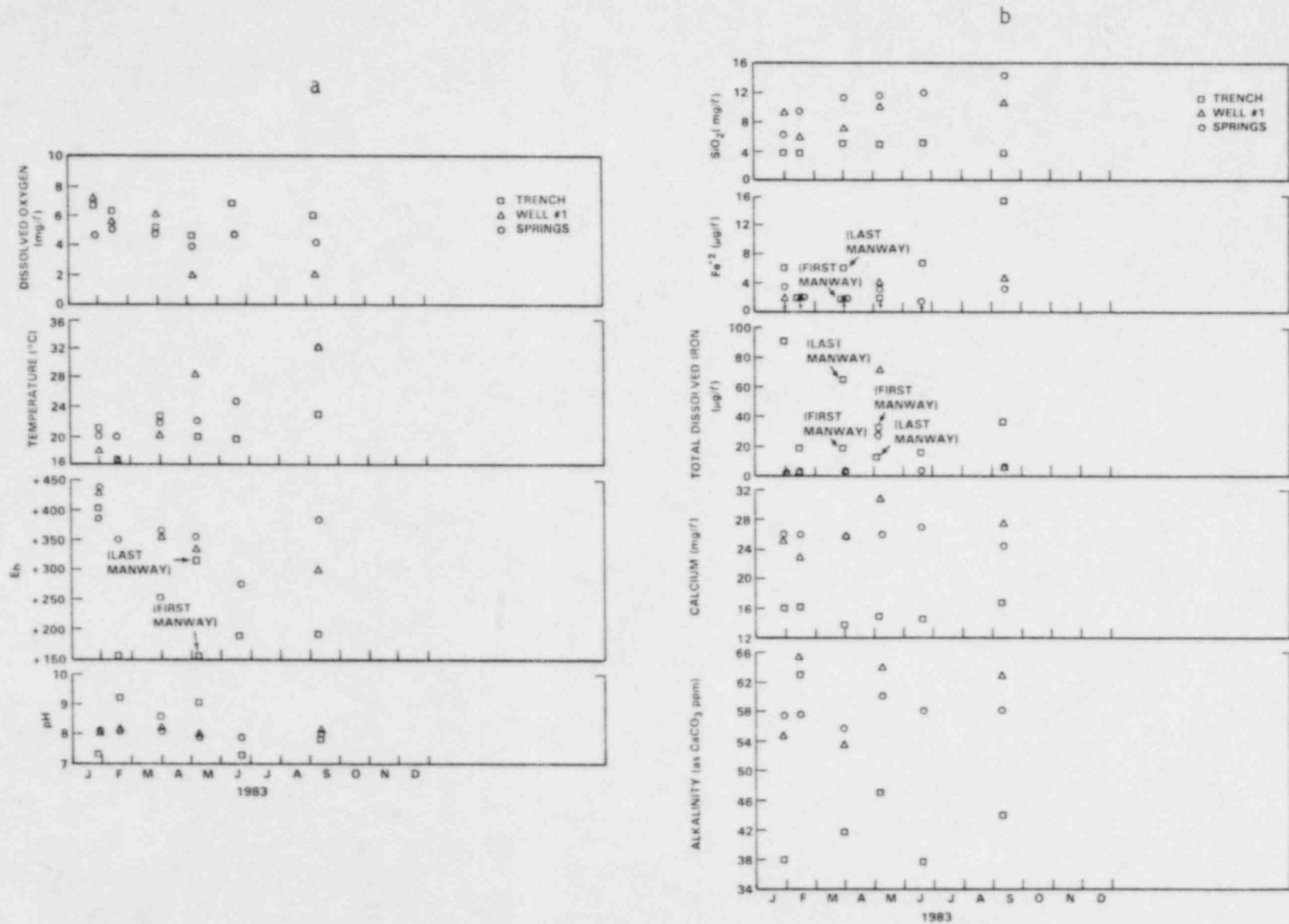


Figure 8. Temporal variability of alkalinity, calcium, total iron, ferrous iron, and silica in trench, well, and spring waters.

inverse relationship with the water temperature, reflecting the reduced solubility of oxygen in water as a function of temperature.

The alkalinity of the trench waters was substantially lower than that of Well #1 and spring water (see Figure 8b), except during the February 1983 sampling when a relatively high alkalinity was correlated with the highest pH (9.25) observed in the trench water. The calcium concentrations showed a relationship similar to that observed for the alkalinity, suggesting dissolution of calcite as the trench water percolates through the soil bank (Section 8.0). Silica (SiO_2) also increases in concentration as the trench water moves through the soil (see Figure 8b), suggesting dissolution of silicate minerals in the soils (Section 8.0).

The concentrations of "total" iron in the trench water was highly variable and ranged from 12 to 91 $\mu\text{g/L}$, whereas Fe^{2+} concentrations were generally low, ranging between <2 to 15 $\mu\text{g/L}$. Ferrous iron concentrations in well and spring waters were always very low. Likewise, sulfide concentrations were very low, again indicating the well-oxygenated nature of all of these waters.

3.6 Conclusions

- 1) Most of the radionuclides in the influent water are removed during the residence time in the disposal basin and the trench, probably by sorption onto the soil.
- 2) Only trace amounts of several radionuclides have been observed to migrate to the springs. The group of mobile radionuclides includes the isotopes of Co, Tc, I, Ru, Sb, Fe, and Ni and the mobile forms are the anionic and nonionic charge forms.
- 3) Typically radionuclides that enter the trench predominantly in particulate forms have higher total concentrations at the beginning of the trench than at the end, whereas radionuclides that enter the trench predominantly in soluble forms tend to be more homogeneously distributed.
- 4) Although the chemistry of the influent water was somewhat variable during the sampling period, the chemistry of the groundwater was quite consistent indicating an effective buffering system in the groundwater.

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4. CHARACTERIZATION OF ORGANIC SPECIES

Because organic compounds may play an important role in mobilizing radionuclides in groundwater, the organic content in water samples from the low-level disposal site has been analyzed. The goal of this research is to identify organic compounds that may chelate radionuclides in the groundwater, thereby enhancing their mobility.

4.1 Sample Collection and Preparation

Water samples were collected from the trench, monitoring wells, and springs in January, February, March, May, and June for dissolved organic constituent analysis. The trench waters were sampled at three locations along the trench referred to as manways 1, 2, and 10, respectively. All except the January water samples were filtered through 0.4-micron Nucleopore filters in the field and collected in acid-cleaned amber glass bottles. The samples were immediately transported to the laboratory where they were stored at 4°C prior to sample analysis.

4.2 Analysis Scheme for Organic Species

Each water sample was filtered in the laboratory just before analysis using a 0.4- μ m silver membrane filter to remove any particulate matter that may have formed during storage and to kill any bacteria in the samples. An aliquot of filtered water (200-1500 mL) was concentrated to near dryness by rotary evaporation prior to organic derivatization. The concentrated water sample was then transferred to a tapered reaction vial and evaporated to complete dryness at 40°C under nitrogen. The resulting residue was methylated with 1 mL of BF_3 /methanol (14% w/v) in a sealed reaction vial at 100°C for 40 min. After cooling, 1 mL of chloroform was added to the vial. This mixture was then transferred to a test tube containing 3 mL of 1 M KH_2PO_4 buffer solution (pH 7) and vortexed. Part of the chloroform layer (0.6 mL), which contained the methylated hydrophilic organics, was then removed and evaporated to complete dryness under nitrogen at room temperature. The residue was immediately redissolved in 100 mL of chloroform and analyzed by GC and by GC-MS.

Gas chromatography analyses were performed on a Hewlett-Packard 5880 gas chromatograph equipped with a 60-m x 0.25-mm I.D. fused silica capillary column coated with a 0.25- μ m film of Se-52. From an initial temperature of 40°C, the column temperature was programmed to increase by 20°C per minute for 3 min. to a final temperature of 100°C. This was followed by an increase in temperature of 8°C per minute for 25 min. until a temperature of 300°C was reached. The sample was maintained at 300°C for 10 min.

Gas chromatography-mass spectroscopy analyses were performed on a Hewlett-Packard 5985 GC-MS instrument in the electron-impact (70-eV) mode. The gas chromatograph on the 5985 instrument was equipped with a 60-m x 0.25-mm I.D. fused silica capillary column coated with 0.25- μ m of Se-54; the column was programmed to increase the temperature at a rate of 5°C per minute starting at 40°C and terminating at 300°C, where it was maintained isothermally for 8 min. A splitless injection system was used to introduce the sample into the GC-MS instrument. A mass range of 50 to 400 amu was scanned every 1.0 s by computer (HP-2100MX equipped with the HP-7920 large disc drive).

The organic species identified by the GC-MS analyses were quantified by GC analyses using internal and external standardization methods. Pure compounds representative of the various compound classes identified by GC-MS were selected as standards and methylated. A specific amount of each standard was co-injected with each sample to confirm the GC-MS identifications. For quantitation, each standard was injected onto the gas chromatograph prior to and following sample analyses. The response factor of each standard was calculated under analytical conditions identical to those of the sample analyses.

4.3 Organic Content of the Trench Waters

Numerous carboxylic acids were identified in the trench waters (Table 7), generally at ppb levels. Four classes of carboxylic acids are represented: dicarboxylic acid (ranging from C₂ to C₁₃); monocarboxylic acids (ranging from C₇ to C₂₁); oxygenated acids (ranging from C₂ to C₅); and aromatic acids (Table 8). The monocarboxylic and dicarboxylic acids are common bioorganic species; the oxygenated and aromatic acids are typical products of microbial activity. In general, the water sampled from Manway 1 contained the most acids (24.5 to 64.2 ppb), followed by Manway 2 (22.1 to 24.5 ppb) and Manway 10 (11.4 to 22.1 ppb). Comparison of the concentration of total acids with the total organic carbon values (Table 8) reveals that the low molecular weight (MW) organic acids account for only a small percentage of the total dissolved organic carbon (0.1 to 2.6%). The bulk of the organic carbon content of the waters is undoubtedly composed of higher MW, relatively non-volatile species which are not detected using GC analysis.

The most likely constituents of the remaining organic carbon are fulvic and humic acids. The relative over-abundance of these non-volatile, higher MW organic species suggests that these species will prove to be much more important in mobilizing radionuclides at this low-level disposal site than the low MW organic acids. Research has indicated that these high molecular weight organic species can be important in

Table 7. Hydrophilic organics in waters from the low-level waste disposal site (ppb x 100).

Hydrophilic Organics*	Manways**			Wells**		Springs**
	No. 1 (2/15/83)	No. 2 (5/5/83)	No. 10 (6/16/83)	No. 1 (2/15/83)	No. 3 (2/17/83)	(6/20/83)
Dicarboxylic acids						
Oxalic acid	272	93	16	BD***	BD	13
Malonic acid	185	59	42	BD	BD	12
Succinic acid	245	90	97	BD	BD	41
Methyl succinic acid	37	BD	41	BD	BD	9
Pentanedioic acid	136	29	53	BD	BD	27
Hexanedioic acid	417	89	80	BD	53	68
Heptanedioic acid	64	16	29	BD	BD	10
Octanedioic acid	58	65	86	BD	34	24
Nonanedioic acid	231	153	112	6	87	38
Decanedioic acid	122	23	32	11	16	21
Undecanedioic acid	106	20	24	13	20	14
Dodecanedioic acid	101	12	13	14	9	12
Tridecanedioic acid	87	26	BD	44	54	12
Monocarboxylic acids						
Heptanoic acid	6	8	11	BD	BD	3
Octanoic acid	102	BD	BD	BD	BD	9
Nonanoic acid	202	29	24	BD	BD	18
Decanoic acid	79	29	36	BD	11	21
Undecanoic acid	18	BD	17	BD	BD	6
Dodecanoic acid	125	100	101	BD	66	54
Tetradecanoic acid	341	150	135	17	144	56
Pentadecanoic acid	125	64	52	13	51	24
Hexadecanoic acid	1,525	554	465	252	638	183
Heptadecanoic acid	43	30	24	18	61	8
Octadecanoic acid	1,000	172	218	207	177	54
Octadecenoic acid	166	BD	BD	32	BD	BD
Nonadecanoic acid	41	BD	14	11	8	6
Heneicosenoic acid	BD	BD	BD	13	BD	BD
Oxygenated acids						
2-Hydroxy propanoic acid	14	35	BD	BD	BD	14
2,2-Dimethoxy ethanoic acid	109	38	230	BD	BD	16
2,2-Dimethoxy propanoic acid	92	147	24	BD	BD	70
4-Oxo-pentanoic acid	55	56	17	BD	BD	24
Aromatic acids						
Benzoic acid	131	31	17	BD	BD	21
Phthalic acid	318	4	193	BD	25	78

*Methylated, BF₃/methanol

**Sampled 2/15/83 through 6/20/83

***BD indicates compound is below detection level

Table 8. Hydrophilic organics in trench waters.

Hydrophilic Organics*	Concentration (ppb) in Manway					
	No. 1		No. 2		No. 10	
	(2/15/83)	(3/30/83)	(5/5/83)	(6/16/83)	(2/15/83)	(6/16/83)
Dicarboxylic acids (C ₂ to C ₁₃)	20.6	3.6	6.8	5.9	3.4	6.3
Monocarboxylic acids (C ₇ to C ₂₁)	37.7	18.5	11.4	12.3	6.7	11.0
Oxygenated acids (C ₂ to C ₅)	2.7	1.4	2.8	1.2	0.5	2.7
Aromatic acids	3.2	1.0	3.5	2.7	0.8	2.1
Total acids (low MW hydrophilic organics)	64.2	24.5	24.5	22.1	11.4	22.1
Total organic carbon	2,500	16,700	16,100	15,200	1,700	1,800

*Methylated, BF₃/methanol

binding radionuclides (Carlsen et al., 1984; Means et al., 1978); especially, since large quantities of specific chelating agents such as EDTA were not identified in these waters in contrast to the relatively large quantities (ppm levels) of synthetic chelating agents such as EDTA, HEDTA and ED3A that have been identified at other low-level disposal sites (Means et al., 1978; Rees and Cleveland, 1982; Toste et al., 1983, 1984).

Comparison of the low MW organic acid content of the trench sediments characterized in FY82 (Robertson et al., 1983) with the trench waters reveals several significant differences. First, citric acid, a tri-carboxylic acid found in biological organisms and often used as a chelating agent for radionuclides, was present in the sediments but was absent in the trench waters. Second, dicarboxylic acids were predominant in the sediments, whereas monocarboxylic acids were most abundant in the trench waters. Third, the trench waters contain a much greater variety of monocarboxylic and dicarboxylic acids than the sediments. On the other hand, the sediments contained a much greater variety of oxygenated and aromatic acids. Finally, there is a greater abundance of acids in the sediments, which suggests a rich microbial biota in the

sediments and vigorous organic production. It appears likely that the abundant biological activity of the sediments was the source of the organic acids in the trench waters, as well as in the groundwaters.

4.4 Organic Species in the Groundwaters

Many of the carboxylic acids identified in the groundwaters collected from Wells #1 and #3 (Table 9) are similar to those present in the trench waters; however, a number of acids identified in the trench waters were not identified in the groundwaters. These absent organic compounds include a number of small dicarboxylic acids. The most significant group of organic compounds absent in the groundwater are the oxygenated acids. Their absence is probably due to their preferential degradation or their reactivity, e.g., sorption on the soil because of their greater polarity. Aromatic acids occur in lower concentrations in the groundwaters than in the trench. The concentrations of the low MW acids in the groundwaters are lower than those in the trench waters despite relatively high Total Organic Carbon concentrations in the groundwater (Table 9). As was observed in the trench waters, the low MW organic acids identified in the groundwaters using GC-MS analysis constitute a small percentage of the TOC in the groundwater (0.05% to 0.14%). The percentage of TOC accounted for by the low MW fraction in the groundwater is much lower than the percentage in the trench waters.

Table 9. Hydrophilic organics in groundwaters.

Hydrophilic Organics*	Concentration (ppb)		
	Well #1		Well #3
	(2/15/83)	(5/6/83)	(2/17/83)
Dicarboxylic acids (C ₂ to C ₁₃)	0.9	3.7	2.7
Monocarboxylic acids (C ₁₀ to C ₂₁)	5.6	7.9	11.6
Oxygenated acids (C ₂ to C ₅)	0	0	0
Aromatic acids	0	1.4	0.3
Total acids (low MW hydrophilic organics)	6.5	13	14.3
Total organic carbon	13,000	19,100	9,900

*Methylated, BF₃/methanol

The higher MW, nonvolatile organic species may be even more important to radionuclide mobility in the groundwaters than in the trench.

4.5 Organic Content of the Spring Water

The spring water contains the same assortment of carboxylic acids identified in the trench and well waters (Table 7). The relative abundances of the various classes of acids are similar to those of the groundwaters (Table 10). The TOC values of the springs waters are relatively high, in some cases higher than those of the groundwaters. Once again, the low MW organic acids identified constitute a small fraction of the waters' TOC (0.06% to 0.19%).

Table 10. Hydrophilic organics in spring water.

Hydrophilic Organics*	Concentration (ppb)		
	(2/17/83)	(5/9/83)	(6/20/83)
Dicarboxylic acids (C ₂ to C ₁₃)	1.4	2.1	3.0
Monocarboxylic acids (C ₇ to C ₂₁)	12	4.8	4.4
Oxygenated acids (C ₂ to C ₅)	0	0.3	1.2
Aromatic acids	0	0.6	1.0
Total acids (low MW hydrophilic organics)	13.4	7.8	9.6
Total organic carbon	8,000	4,200	17,000

*Methylated, BF₃/methanol

4.6 Conclusions

- 1) The low MW hydrophilic organic compounds in water samples from the trench, wells, and springs consist of carboxylic acids, generally at ppb levels.
- 2) Four classes of carboxylic acids are represented and are bioorganic and microbial diagenesis products.

- 3) The organic acids in the trench waters arise from the rich biological activity of the trench muds.
- 4) The low MW hydrophilic organic compounds (carboxylic acids) of the trench, well, and spring waters constitute only a small percentage of the total organic carbon in the water (0.05% to 2.6%) suggesting that if organic species contribute to radionuclide transport then the high MW organic species will be most important.

4.7 References

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5. COLLOID ANALYSIS

The presence of colloidal material in nuclear generating plant operating systems has been the focus of recent utility research (Matijevic, 1982; Berry and Diegle, 1979; Chen and Theus, 1982) because the corrosion product colloids become neutron activated in the reactor vessel and subsequently deposited throughout the operating systems. This results in increased exposures during maintenance operations. The colloidal material is also not filterable using normal commercial cartridge systems. Attempts have been made to apply ultrafiltration to radioactive waste systems in order to achieve better decontamination levels (Palino et al., 1982).

Colloidal material is also naturally present in soil systems in inorganic form as hydrous oxides of iron and aluminum and in organic form as macromolecules, typically classified as humic or fulvic acids. It has been suggested that radionuclides present in low-level waste sites may be migrating associated with colloids (Killey et al., 1984; Champ and Merritt, 1981) of inorganic or organic nature.

The objective of this task is to investigate the nature and contribution of colloids in the influent and groundwaters at a low-level disposal site in order to assess the potential for colloid associated migration.

5.1 Methods

A set of water samples was obtained from the influent and from Well #1, which is closest to the trench. These samples were processed sequentially through membrane filters ranging from 8.0 to 0.1 μm . Several liters of the filtrate from the finest membrane filter was then subjected to ultrafiltration sequentially through filters with pore sizes ranging from 0.01 to 0.001 μm . The filter pore sizes, μm filter types, and volumes subjected to filtration are shown in Table 11. The filtering process took several weeks, since the ultrafilters have very slow flow rates. During the ultrafiltration, the water samples were maintained in a refrigerated condition. The material retained on each pore size filter was rinsed with distilled water, packaged, and subjected to gamma spectrometric analysis. The colloidal range extends from 0.1 to 0.001 μm .

5.2 Results

The radionuclide concentrations on the separated particulate matter are shown in Tables 12 and 13. In the influent water, a much more detailed characterization was possible since the radionuclide activity levels are

Table 11. Sequential filter series used for particulate and colloid separation.

Pore Size (μm)	Filter Type	Diameter (mm)	Volume Filtered
8.0	Nuclepore	144	25 to 40 L
3.0	Nuclepore	144	25 to 40 L
1.0	Nuclepore	144	25 to 40 L
0.4	Nuclepore	144	25 to 40 L
0.1	Nuclepore	144	25 to 40 L
0.01	Amicon	76	6 to 8 L
0.005	Amicon	76	6 to 8 L
0.003	Amicon	76	6 to 8 L
0.001	Amicon	76	6 to 8 L

much higher. Particulate associated radionuclides in the colloidal size range were observed in both the influent and Well #1 waters. The particulate radionuclide concentrations (pCi/L) at Well #1 were so low that only a limited number of radionuclides were in concentrations above detection limits (Table 13). Figure 9 illustrates percentage distributions for several radionuclides measured in the trench water. The distributions of ^{60}Co , ^{54}Mn , and ^{59}Fe are quite similar with the greatest particulate associated activity occurring in the largest particle size of 8.0 μm . All three radionuclides also have an increased percentage of the radionuclide associated with the smallest filter sizes, (1.0 or 0.4 in the case of ^{59}Fe μm , 0.01 μm , and 0.001 μm) compared with intermediate pore sizes. Another group of radionuclides in the influent water, ^{103}Ru , ^{106}Ru , ^{124}Sb , and ^{137}Cs , have a smaller percentage (30% or less) of their total particulate activity associated with the largest filter. This latter group of radionuclides have increases in the percentage distribution versus particulate size in the size range retained on the 0.4- μm filter. All these latter group of radionuclides except ^{137}Cs also have peaks in the percentage distribution at 0.01 μm , and ^{137}Cs shows increasing concentrations in the smallest sizes from >0.005 to >0.001 μm .

In the well water (Figure 10), a very different particulate size distribution is observed than that found for the trench water. A concentration maximum for ^{54}Mn , ^{60}Co , ^{103}Ru , and ^{137}Cs was measured in the 1.0- μm

Table 12. Source water particulate matter, September 1983
(Radionuclide concentrations in pCi/L.)

Isotope	8.0 μm^*	3.0 μm^*	1.0 μm^*	0.4 μm^*	0.1 μm^*	0.01 μm^*	0.005 μm^*	0.003 μm^*	0.001 μm^*
Sc-46	17 \pm 4	6.5 \pm 0.9	5.9 \pm 0.9	4.7 \pm 0.9	1.6 \pm 0.6	3.4 \pm 0.9	<0.1	<0.3	<0.4
Cr-51	1000 \pm 20	352 \pm 6	370 \pm 10	150 \pm 10	53 \pm 9	126 \pm 8	<1	<5	40 \pm 10
Mn-54	2828 \pm 7	613 \pm 1	698 \pm 1	494 \pm 1	247 \pm 1	461 \pm 1	53.1 \pm 0.4	111 \pm 1	490 \pm 1
Fe-59	12340 \pm 30	5709 \pm 6	7723 \pm 7	8078 \pm 8	2834 \pm 6	4711 \pm 6	19.1 \pm 0.5	9.0 \pm 0.9	11 \pm 1
Co-56	<4	3.6 \pm 0.9	4 \pm 1	3.6 \pm 0.9	2.3 \pm 0.6	2.8 \pm 0.9	0.3 \pm 0.1	0.6 \pm 0.3	0.6 \pm 0.5
Co-57	6 \pm 1	2.4 \pm 0.3	4.7 \pm 0.3	6.0 \pm 0.3	1.9 \pm 0.2	3.4 \pm 0.3	0.39 \pm 0.06	3.3 \pm 0.1	3.5 \pm 0.1
Co-58	271 \pm 4	58.6 \pm 0.9	74.2 \pm 0.9	13.8 \pm 0.8	5.3 \pm 0.6	7.4 \pm 0.8	<0.1	<0.4	2.0 \pm 0.5
Co-60	5440 \pm 10	1101 \pm 2	1278 \pm 1	261.1 \pm 0.8	87.5 \pm 0.5	145 \pm 1	13.9 \pm 0.3	58.0 \pm 0.5	145 \pm 1
Zn-65	94 \pm 7	24 \pm 1	32 \pm 1	14 \pm 1	6.8 \pm 0.7	10 \pm 1	0.5 \pm 0.3	1.3 \pm 0.4	<0.6
Zr-95	1680 \pm 10	876 \pm 2	1275 \pm 3	1416 \pm 3	573 \pm 2	993 \pm 3	25.1 \pm 0.4	75 \pm 1	251 \pm 1
Nb-95	3140 \pm 10	1718 \pm 2	3969 \pm 4	5018 \pm 5	2157 \pm 4	3065 \pm 4	68.8 \pm 0.5	274 \pm 1	781 \pm 4
Ru-103	960 \pm 5	562 \pm 1	823 \pm 2	1010 \pm 2	476 \pm 2	906 \pm 1	9.9 \pm 0.3	16.3 \pm 0.5	24 \pm 1
Ru-106	540 \pm 30	214 \pm 6	315 \pm 5	337 \pm 5	139 \pm 4	269 \pm 6	5 \pm 1	6 \pm 3	<4
Sb-124	99 \pm 3	26.4 \pm 0.6	35.0 \pm 0.7	56.6 \pm 0.8	41.7 \pm 0.8	80 \pm 1	0.4 \pm 0.1	<0.3	<0.4
Sb-125	14 \pm 6	5 \pm 1	15 \pm 1	31 \pm 1	24.2 \pm 0.9	44 \pm 1	0.9 \pm 0.3	<0.5	<0.8
I-131	81 \pm 9	29 \pm 2	90 \pm 30	<40	<40	61 \pm 6	9 \pm 1	30 \pm 10	300 \pm 250
Cs-134	6 \pm 4	<0.8	1.1 \pm 0.6	<0.6	<0.4	<0.6	0.9 \pm 0.1	2.1 \pm 0.3	3.8 \pm 0.3
Cs-137	106 \pm 3	19.2 \pm 0.6	18.2 \pm 0.5	9.5 \pm 0.4	9.7 \pm 0.7	18.6 \pm 0.6	21.0 \pm 0.3	61.0 \pm 0.4	89.4 \pm 0.6
Ce-141	2624 \pm 5	1364 \pm 2	1892 \pm 2	2360 \pm 3	1008 \pm 2	1646 \pm 3	74 \pm 1	213 \pm 1	110 \pm 1
Ce-144	4860 \pm 20	2256 \pm 4	2900 \pm 40	4020 \pm 40	1745 \pm 3	2860 \pm 4	131 \pm 1	381 \pm 1	224 \pm 1
Eu-152	<5	<1	<1	<1	<0.8	<1	<0.3	<0.5	<0.6
Eu-154	<20	<4	6 \pm 3	11 \pm 3	5 \pm 2	8 \pm 4	1.9 \pm 0.5	4 \pm 1	10 \pm 1
Eu-155	16 \pm 6	6 \pm 1	12 \pm 1	18 \pm 1	7.4 \pm 0.9	13 \pm 1	1.5 \pm 0.3	12.1 \pm 0.5	13.0 \pm 0.5

*Pore size of filter in micrometers

Table 13. Well #1 particulate matter, September 1983.
(Radionuclide concentrations in pCi/L.)

Isotope	8.0 μm^*	3.0 μm^*	1.0 μm^*	0.4 μm^*	0.1 μm^*	0.01 μm^*	0.005 μm^*	0.003 μm^*	0.001 μm^*
Sc-46	<0.02	<0.02	<0.06	<0.03	<0.02	<0.07	<0.05	0.23 \pm 0.07	<0.2
Cr-51	<0.2	<0.2	<0.6	<0.3	<0.2	<0.7	<0.7	<1	<5
Mn-54	0.09 \pm 0.02	0.36 \pm 0.03	0.39 \pm 0.06	0.35 \pm 0.03	0.23 \pm 0.02	<0.05	0.10 \pm 0.05	0.15 \pm 0.05	0.9 \pm 0.1
Fe-59	0.23 \pm 0.06	0.18 \pm 0.05	0.2 \pm 0.1	0.20 \pm 0.06	0.08 \pm 0.05	<0.1	0.3 \pm 0.1	<0.2	<0.5
Co-56	0.07 \pm 0.03	0.04 \pm 0.02	0.11 \pm 0.06	0.07 \pm 0.03	0.06 \pm 0.03	0.23 \pm 0.08	0.34 \pm 0.07	0.25 \pm 0.08	0.7 \pm 0.2
Co-57	<0.02	<0.02	<0.05	<0.02	<0.02	<0.05	0.54 \pm 0.05	0.11 \pm 0.05	0.16 \pm 0.08
Co-58	<0.02	0.03 \pm 0.02	<0.06	<0.03	<0.02	<0.07	<0.07	<0.07	<0.2
Co-60	1.38 \pm 0.05	1.48 \pm 0.05	2.2 \pm 0.1	1.7 \pm 0.1	1.05 \pm 0.04	0.64 \pm 0.08	0.64 \pm 0.07	0.80 \pm 0.08	7.2 \pm 0.2
Zn-65	<0.05	<0.05	<0.09	<0.05	<0.04	<0.1	<0.1	<0.1	<0.2
Zr-95	0.15 \pm 0.04	0.08 \pm 0.04	0.2 \pm 0.1	0.35 \pm 0.05	0.08 \pm 0.04	0.2 \pm 0.1	0.2 \pm 0.1	<0.1	<0.3
Nb-95	0.09 \pm 0.03	0.05 \pm 0.02	0.13 \pm 0.07	0.30 \pm 0.04	0.08 \pm 0.03	0.11 \pm 0.07	<0.07	<0.1	<0.3
Ru-103	1.83 \pm 0.05	0.90 \pm 0.03	3.0 \pm 0.1	2.77 \pm 0.06	0.91 \pm 0.04	0.51 \pm 0.08	0.67 \pm 0.08	1.2 \pm 0.1	23.0 \pm 0.5
Ru-106	1.2 \pm 0.2	0.7 \pm 0.2	1.1 \pm 0.5	1.7 \pm 0.2	0.6 \pm 0.2	<0.5	0.6 \pm 0.5	0.8 \pm 0.5	10 \pm 1
Sb-124	0.05 \pm 0.03	<0.03	<0.05	<0.03	0.06 \pm 0.03	<0.1	<0.1	<0.1	<0.2
Sb-125	0.15 \pm 0.06	<0.05	<0.1	<0.06	<0.05	<0.1	<0.1	<0.1	<0.3
I-131	2.2 \pm 0.8	2.08 \pm 0.08	3.1 \pm 0.2	2.0 \pm 0.1	0.9 \pm 0.1	0.5 \pm 0.3	1.5 \pm 0.3	2.0 \pm 0.8	110 \pm 50
Cs-134	<0.02	<0.02	0.09 \pm 0.05	0.06 \pm 0.03	0.04 \pm 0.02	<0.07	0.13 \pm 0.07	<0.07	0.3 \pm 0.1
Cs-137	0.22 \pm 0.02	<0.02	<0.05	0.04 \pm 0.02	<0.02	<0.05	5.6 \pm 0.1	0.18 \pm 0.05	0.6 \pm 0.1
Ce-141	<0.04	<0.04	<0.1	0.10 \pm 0.05	<0.04	<0.1	<0.1	<0.1	<0.7
Ce-144	0.4 \pm 0.2	<0.1	<0.4	0.9 \pm 0.2	<0.1	<0.3	<0.3	<0.3	<0.7
Eu-152	<0.06	<0.06	<0.1	<0.07	<0.06	<0.2	<0.2	<0.2	<0.3
Eu-154	<0.1	<0.1	<0.3	<0.1	<0.1	<0.3	0.8 \pm 0.3	<0.3	<0.5
Eu-155	0.12 \pm 0.08	<0.07	<0.2	0.3 \pm 0.1	<0.07	<0.2	2.8 \pm 0.2	0.3 \pm 0.2	1.1 \pm 0.3

*Pore size of filter in micrometers

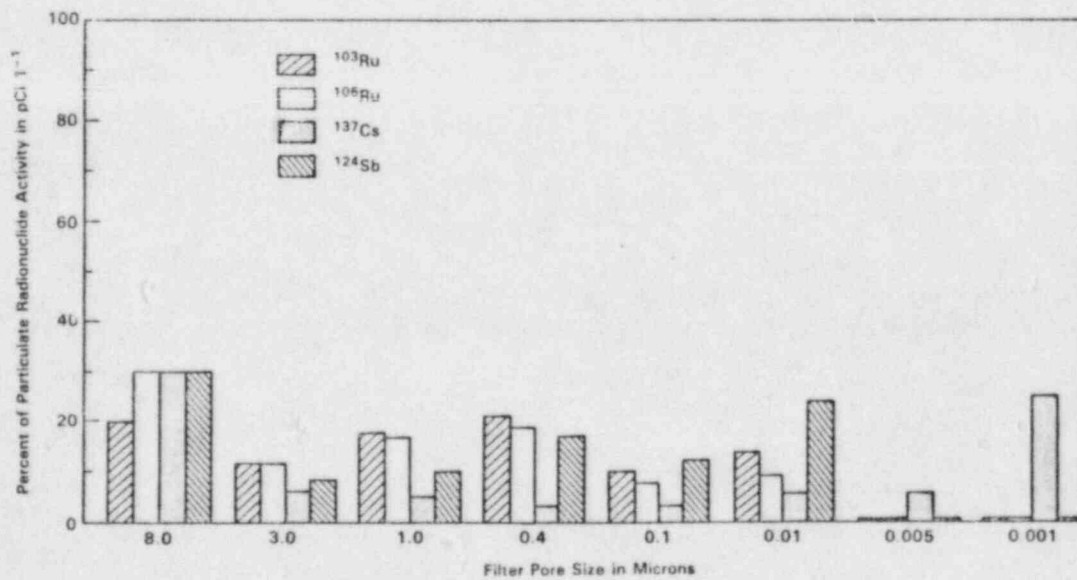
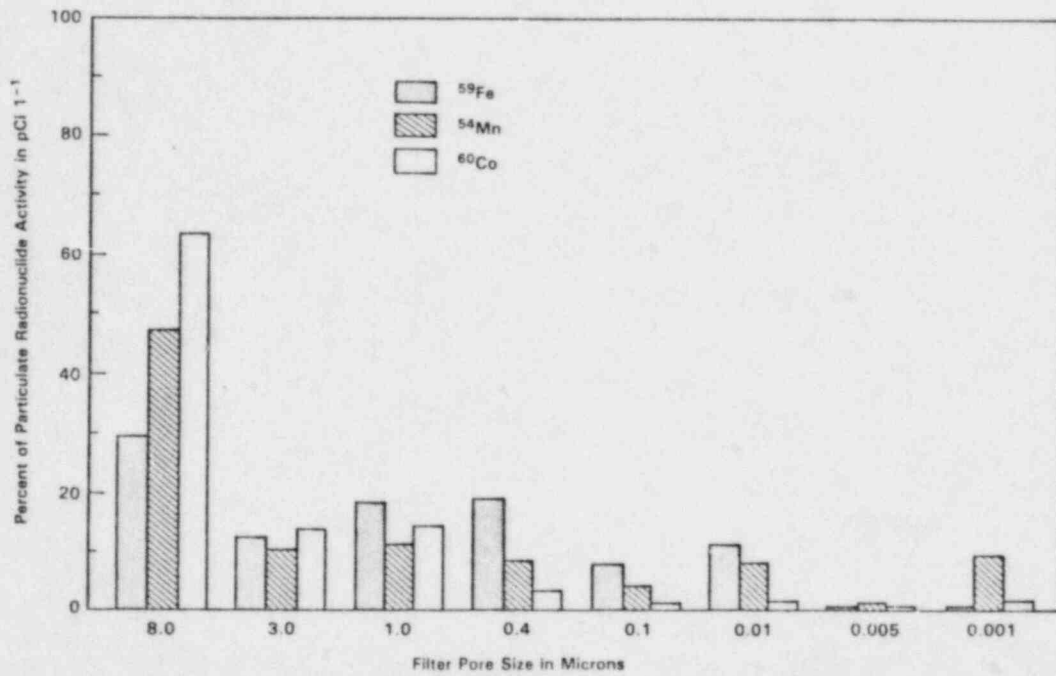


Figure 9. Percentage distributions for selected radionuclides found in the trench water.

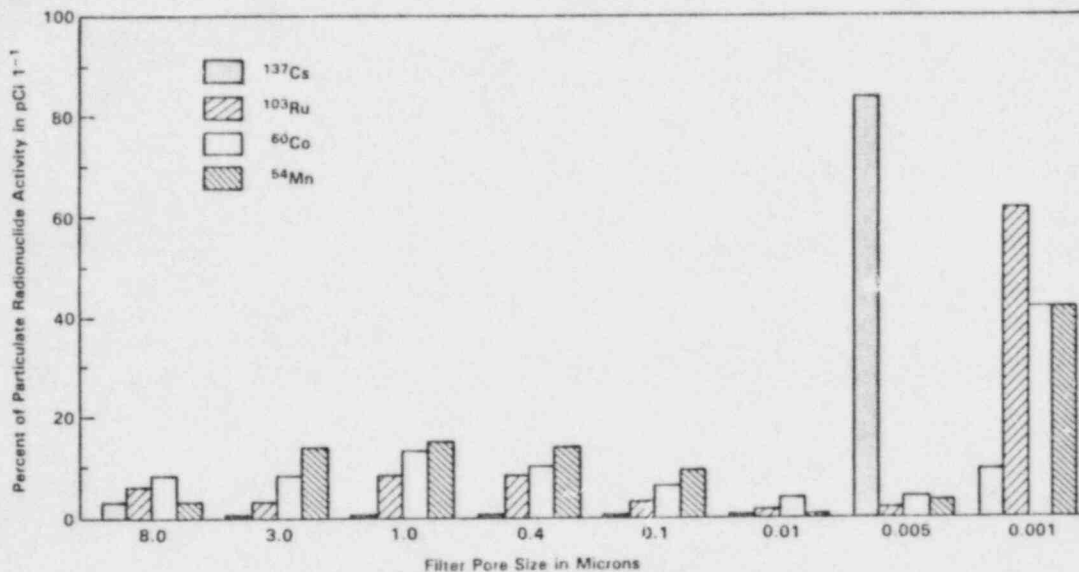


Figure 10. Percentage distributions for radionuclides found in well water.

size range and strikingly increased percentages of radionuclides were observed associated with the finer particulate material. A maximum was measured for ¹³⁷Cs on the 0.005- μ m filter, whereas the other three radionuclides, ⁵⁴Mn, ⁶⁰Co, and ¹⁰³Ru, show steadily increasing concentrations from 0.01 μ m to 0.001 μ m.

The percentage of the particulate associated activity, on a pCi/L basis, found in the colloidal size range (<0.4 to >0.001 μ m) is shown for both the trench water and well water in Table 14. The fraction of activity in the colloidal size range increases for all radionuclides above detection limits in this set of samples. For all radionuclides measured in Well #1, the percentage of particulate associated activity in the colloidal range is greater than 50% of the total particulate activity, and for ¹²⁴Sb and ¹³⁷Cs the colloidal particulates constitute 75% and 97% of the total particulate associated activity.

Although the colloidal particulates contain a substantial portion of the total particulate associated activity, the particulate activity is typically a small fraction of the total activity. Tables 15 and 16 show the distribution of radionuclide activity between the larger particulates, the colloidal particulates, and the solution phases. In the trench water, radionuclide distributions are quite variable, with the soluble fraction ranging from 1% to greater than 99% for ¹⁰³Ru and ¹³⁷Cs, respectively. The activity associated with the colloidal fraction ranges from <1% to 27% for ¹³⁷Cs and ¹⁰³Ru, respectively.

Table 14. Percentages of the total particulate associated with radionuclide activity in the colloidal range (<0.4 μm to >0.001 μm).

Radionuclide	Trench Water	Well #1
^{54}Mn	13%	53%
^{59}Fe	18%	--
^{60}Co	5%	59%
^{106}Ru	23%	--
^{103}Ru	29%	75%
^{124}Sb	36%	--
^{137}Cs	56%	97%

Table 15. Percentages of radionuclides in influent water in particulate, colloidal, and soluble fractions.

Radionuclide	>0.4 μm	<0.4 to >0.001 μm	Soluble
^{54}Mn	6%	2%	92%
^{60}Co	50%	3%	4%
^{103}Ru	62%	27%	1%
^{106}Ru	60%	18%	22%
^{137}Cs	<1%	<1%	>99%

Table 16. Percentages of radionuclides at Well #1 in particulate, colloidal, and soluble fractions.

Radionuclide	>0.4 μm	<0.4 to >0.001 μm	Soluble
^{60}Co	4%	7%	89%
^{103}Ru	10%	4%	95%
^{106}Ru	2%	4%	94%
^{131}I	0.7%	9%	91%
^{137}Cs	<1%	13%	87%

In the Well #1 waters, the soluble fraction represents typically 90% or greater of the total activity for the detected radionuclides. The colloidal fraction contains from 4% to 13% of the total activity as shown in Table 16.

5.3 Conclusions

- Radionuclides associated with particulates in the colloidal size range are present in the influent and in the first monitoring well; however, with the limited program undertaken it was not possible to determine whether the radionuclides in the colloidal fraction at Well #1 had migrated to the well as colloidal particulates, or whether the radionuclides had migrated in solution and been sorbed from solution onto natural soil colloids present at Well #1.
- The colloidal fraction is a larger percentage of the total particulate at Well #1 compared to the source, however, the soluble fraction constitutes 90% or greater of the total activity at Well #1.

- A much more comprehensive program would be necessary to define whether colloidal particulates are indeed migrating in the groundwater.

5.4 References

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6. QUANTITATIVE DETERMINATION OF SORPTION SUBSTRATES

Modeling of radionuclide adsorption onto sediment using a surface-coordination modeling approach requires estimates of the quantity of the important substrates present in these samples. There are adequate reasons to believe that the primary sorption substrates are oxides of Fe, Mn, Al, and Si plus the reactive portion of particulate organic carbon that occur as coatings on detrital and authogenic minerals and their agglomerates (Niehof and Loeb, 1972; Suarez and Langmuir, 1976; Jenne, 1977; Hunter and Liss, 1979; Hunter, 1980). The available evidence indicates that both coprecipitated and adsorbed impurities within the oxides, as well as the effect of clay mineral surfaces upon which much of the oxide precipitates have nucleated, contribute to the noncrystalline nature of naturally occurring oxides (Jenne, 1968, 1977). The amorphous and microcrystalline nature of these mixed oxide coatings preclude obtaining useful characterization with either x-ray or electron diffraction. Thus, chemical extractions offer the only available method to estimate the quantities of these oxides occurring as coatings.

It appears likely that because of the well known reactivity of manganese and iron oxides, both in terms of coprecipitation and sorption and their relative high intrinsic adsorption coefficients, those two oxides are likely to be the primary adsorption substrates. Amorphous aluminum oxides are also important in the sorption of certain anionic species such as borate (Sims and Bingham, 1967, 1968a, 1968b).

There are numerous selective dissolution techniques, most of which call for a single extraction period and all of these suffer from a lack of selectivity for a particular solid phase (Luoma and Jenne, 1976, p. 343; Guy et al., 1978). The only way to obtain assurance that the quantity of metal extracted represents the amorphous phase in its entirety and does not include an appreciable quantity of crystalline oxides of the primary component is to carefully analyze extraction curves obtained by determining the amount of oxidic elements dissolved as a function of time. In addition, the choice of extractant must be made very carefully since many extractants are quite unspecific for the amorphous Fe and Mn phases.

Recently Chao and Zhou (1983) suggested that acidic hydroxylamine hydrochloride, which is the classical extractant for poorly crystalline Mn oxides, be used to estimate amorphous Fe oxides as well. We selected this extractant because of the opportunity it provided to (1) obtain both Fe and Mn with a single extraction, (2) extract amorphous iron slowly enough to permit it to be quantified by curve stripping or equation-of-line techniques, (3) evaluate the possible existence of more

than one amorphous Fe oxide phase based on their dissolution rates as evidenced by multiple slopes of the extraction curve, and (4) evaluate interelemental relationships in the extracts for evidence for the existence of multiple amorphous Fe oxide phases.

Luoma and Bryan (1981) estimated the reactive particulate organic content of estuarine sediment with 0.1 N NaOH and 1 N ammonia using adsorption at 460 nm in a 1-cm cell to quantify the amount of organic carbon extracted. We elected to use 0.5 N KOH for extraction of reactive particulate organic carbon. The KOH was used instead of NaOH to minimize attack on layer silicates (Dudas and Harward, 1971).

6.1 Experimental Method

The original samples of the unconsolidated glaciofluvial samples were collected with a bailer during well drilling. These samples were initially used for gamma counting. Samples recovered from successive depths were composited as necessary to provide an adequate quantity of sediment from above and below the water table for each of the two wells. The depths from which the samples were composited and their size distribution are shown in Figure 11. The dried sediment was sieved

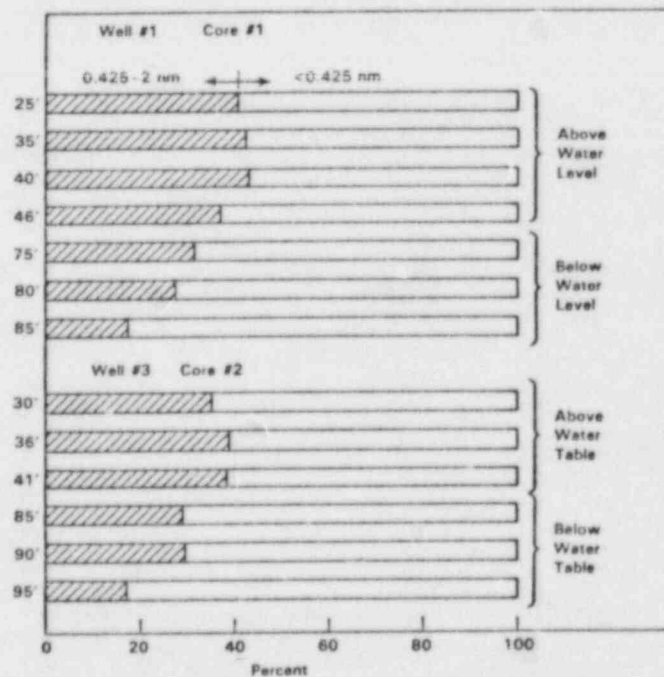


Figure 11. Distribution of <2.0-mm fraction of sediment.

through a 2-mm (No. 10) sieve, with light pressure from a ceramic pestle, if necessary, to break up large aggregates. Both fractions were weighed. The >2-mm fraction was stored. Weighed portions of the <2-mm fraction were sieved onto a <425- μ m (No. 40 sieve) and >425- μ m fractions and their air-dry weights determined. Oven-dry (110°C) moisture content was determined on duplicate (<10 g) samples of each fraction.

For the extractions, 0.50 ± 0.02 g of <425- μ m sediment was added to 95 mL of 0.25 M $\text{NH}_2\text{OH}\cdot\text{HCl}$ in 0.25 M HCl , which had been preheated to 50°C, with stirring for a selected time interval. It was then cooled for 10 minutes in a water bath. The suspension was brought up to 100 mL in a volumetric flask with deionized water, centrifuged and filtered through a 0.22- μ m membrane filter (Gelman). Samples, together with blank solutions, were analyzed by inductively-coupled plasma spectrophotometry.

6.2 Results

These sediments are coarse textured as shown in Figure 11 and Table 17. As shown in Table 18, these samples contain very low amounts of inorganic carbon with organic carbon values that are normal for coarse-textured sediments (Jenne, 1977). The fraction of the organic carbon that is extractable in alkali, hence, presumed to be available for reactions with radionuclides varies between 60% and 76% of the

Table 17. Gravimetric analysis.

Depth		Total	Whole Sample				<2 mm Portion					
			>2 mm		<2 mm		0.425 to 2 mm			<0.425 mm		
m	ft		g	%	g	%	<wt g	% of 2 mm	% of Total	<wt g	% of 2 mm	% of Total
Sample 3-1												
9.1	30	249.59	30.90	12.4	218.61	87.6	76.79	35.1	30.8	141.32	64.6	56.6
11.0	36	246.59	45.43	18.4	201.13	81.6	77.90	38.7	31.6	122.71	61.0	49.8
12.5	41	283.60	59.57	21.0	224.01	79.0	85.17	38.0	30.0	138.34	61.8	48.8
Total							239.86			402.37		
Sample 3-2												
25.9	85	300.25	109.51	32.2	230.60	67.8	66.97	29.0	19.7	163.32	70.8	48.0
27.4	90	240.77	76.78	31.9	163.95	68.1	48.91	29.8	20.3	114.82	70.0	47.7
29.0	95	250.67	44.60	17.8	206.02	82.2	36.45	17.7	14.5	169.40	82.2	67.6
Total							152.33			447.54		

Table 18. Physical and chemical characterization of the <425- μm (40-mesh) fraction of the sediment sample.

Sample Number	Depth (ft)	Surface Area	Avg.	Carbon				Reactive Particulate Organic Carbon Relative to Wt			
				Organic		Inorganic		% of Tot. Wt.	% of TOC		
				Duplicates	Avg.	Duplicates	Avg.				
		---	m ² /g	---	wt %	---	---	---	---		
2-1	25-46	12.65, 12.71	<u>12.68</u>	0.040	0.027	<u>0.034</u>	0.143	0.070	<u>0.106</u>	0.024	<u>70.6</u>
2-2	75-85	3.85, 4.46, 3.86	<u>4.06</u>	0.010	0.030	<u>0.025</u>	0.110	0.040	<u>0.075</u>	0.019	<u>76.0</u>
3-1	30-41	12.69, 11.59	<u>12.14</u>	0.01	0.020	<u>0.015</u>	0.080	0.110	<u>0.095</u>	0.009	<u>60.0</u>
3-2	85-95	3.23, 2.93	<u>3.08</u>	0.020	0.012	<u>0.016</u>	0.110	0.060	<u>0.085</u>	0.012	<u>75.0</u>

Note: Total carbon determined by microcombustion and infrared analysis, Schwartzkopf Microanalytical Laboratory, 56-19 37th Avenue, Woodside, NY 11377; Surface area by BET using nitrogen; reactive particulate organic carbon extracted with 0.5 M KOH at 90°C for 30 min.

total organic carbon. The sediment samples from below the water table (2-2 and 3-2) have a significantly smaller amount in the <425 μm fraction than the two samples from above the water table (2-1 and 3-1). It is also apparent from the BET surface area values (Table 18) that the median diameter of the <425 μm fraction is larger in the samples from below the water table. The surface area measurements were determined from 15 injections of ultra-pure N_2 into a Quantasorb BET surface area analyzer. The range of the 15 observations was $\pm 4.2\%$ of mean with a coefficient of variation of 2.25%, the 95% confidence interval calculates to be 1.25%. Accuracy determinations obtained with 3 replicates are given in Table 19.

Table 19. Accuracy of surface area measurements.

Standards		Expected*	Measured
Material	Lot No.	$\bar{X} \pm \text{s.d.}$	$\bar{X} \pm \text{s.d.}$
		----- m^2/g -----	-----
Al_2O_3	7008	1.97 ± 0.014	2.01 ± 0.045
SiO_2	8008	5.15 ± 0.044	4.84 ± 0.11
TiO_2	5008	13.0 ± 0.5	12.46 ± 0.28

*Mean surface area and standard deviation determined by manufacturer based on six samples of the given lot.

A single 30-min. hydroxylamine hydrochloride extraction was carried out initially to obtain a preliminary estimate of the concentrations of the amorphous oxides and extractable trace elements. These data are given in Table 20 along with error estimates. The combined reliability of the estimated concentrations is demonstrated in Figure 12. Table 20 lists the quantities of Fe, Al, Si, and Mn extracted with time as micromoles per gram of dry soil.

It was initially hypothesized that extraction of amorphous Fe and Mn oxides with time would proceed according to first-order kinetics, i.e., the rate of dissolution was proportional to the amount of material present. Thus, the data were initially plotted as the logarithm of the total dissolved metal normalized to unit extraction time, as a function of extraction time (Figure 13). These plots indicated that there was

Table 20. Chemical analyses of Fe, Mn and associated metals in 30-min extractions with 0.25 M NH₂OH·HCl at 50°C from the <.425-μm fraction.

Sample 2-1

Element	Replicate Extractions	Mean	Instrument Precision
----- mg/kg -----			
Fe	7721, 7351	7536	±16
Mn	147, 158	153	±4.5
Al	1997, 2056	2027	±8
Si	2475, 2489	2482	±32
B	11, 11.9, 15.3	13.6	±3.7
V	-----	<21.2	-
Cr	5.4, <3.0	-----	±1.7
Cu	11.4, 7.6	9.4	±2.1
Zn	86.9, 78.1	82.5	±4.3
Mo	9.2, <8.9	-	3.3
Co	-----	<11.5	-
Ni	-----	<13.5	-

Sample 2-2

Element	Replicate Extractions	Mean	Instrument Precision
----- mg/kg -----			
Fe	3519, 3407	3463	±50
Mn	99, 102	100	±2.4
Al	1275, 1353	1314	±22
Si	1378, 1447	1412	±31
B	-----	<12	-
V	-----	<11.6	-
Cr	-----	<3.6	-
Cu	5.9, 6.0	6.1	±1.20
Zn	138, 90.9	114	±4.5
Mo	-----	<12.0	-
Co	-----	<3.8	-
Ni	-----	<9.1	-

Sample 3-1

Element	Replicate Extractions	Mean	Instrument Precision
----- mg/kg -----			
Fe	9149, 10655	9902	±117
Mn	172, 191	182	3.6
Al	2400, 2587	2494	±35
Si	2990, 3576	3283	±37
B	-----	<13.0	-
V	-----	<12.0	-
Cr	-----	<6.4	-
Cu	8.9, 10.7	9.8	±1.2
Zn	69.6, 109	89.3	±4.1
Mo	-----	<11.0	-
Co	-----	<6.9	-
Ni	-----	<6.4	-

Sample 3-2

Element	Replicate Extractions	Mean	Instrument Precision
----- mg/kg -----			
Fe	2189, 1864	2026	±21
Mn	66.1, 65.4	65.7	±1.9
Al	844, 851	848	±8
Si	849, 865	857	±18
B	-----	<10.4	-
V	-----	<11.2	-
Cr	-----	<8.5	-
Cu	6.1, 4.3	5.2	±1.4
Zn	113, 83.2	98.3	±5.7
Mo	-----	<11.4	-
Co	-----	<7.8	-
Ni	-----	<10.2	-

NOTE: Precision cannot be determined for below detection values.

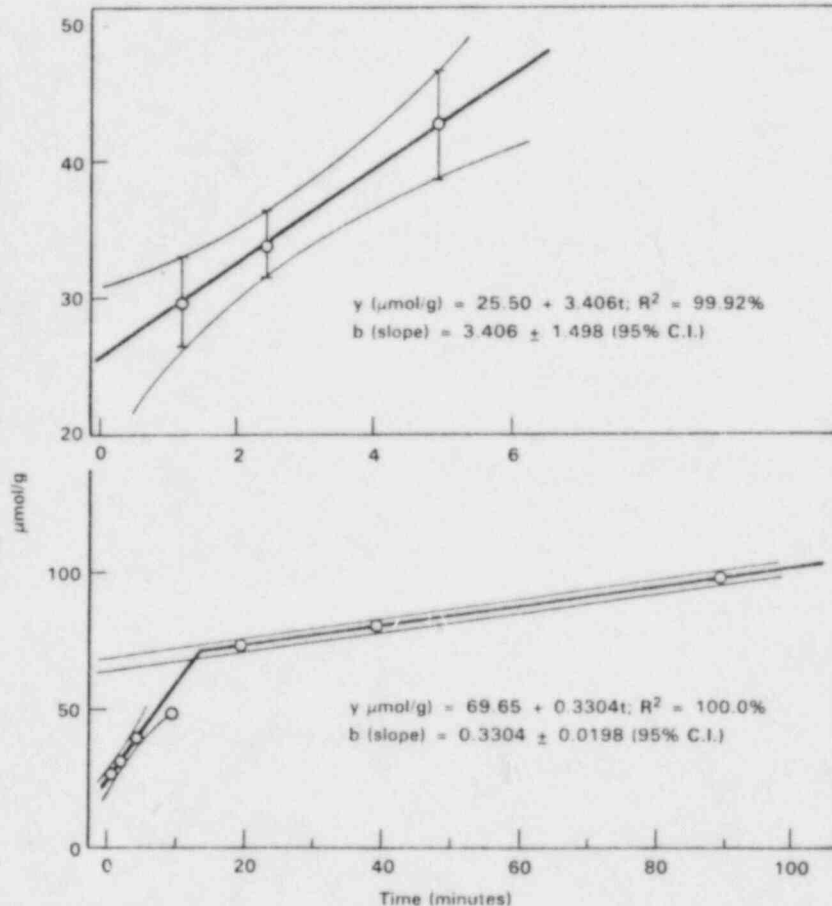


Figure 12. Confidence interval of regression equations for Fe concentrations at selected extraction time intervals.

more than one component and, perhaps, as many as three. However, a greater number of extraction times would be required to permit an interpretation to be made using this approach. This, as well as the possibility of zero-order kinetics, led us to utilize linear time plots.

Replicates of a single time point have not yet been carried out, but it appears that the 0.5-g sample size was adequate, however, based upon the relative constancy of Si or Al to Fe ratios.

Regression of the concentration of one element against another (not shown) provides independent evidence that at least two components of extractable Fe are present in these sediments. For sample 2-1 the first component has a larger Fe content than the second, i.e., the Fe:Al and Fe:Si ratios are about 3:1 and 5:2, respectively, in the first component, compared with about 5:4 and 1:1, respectively, in the second

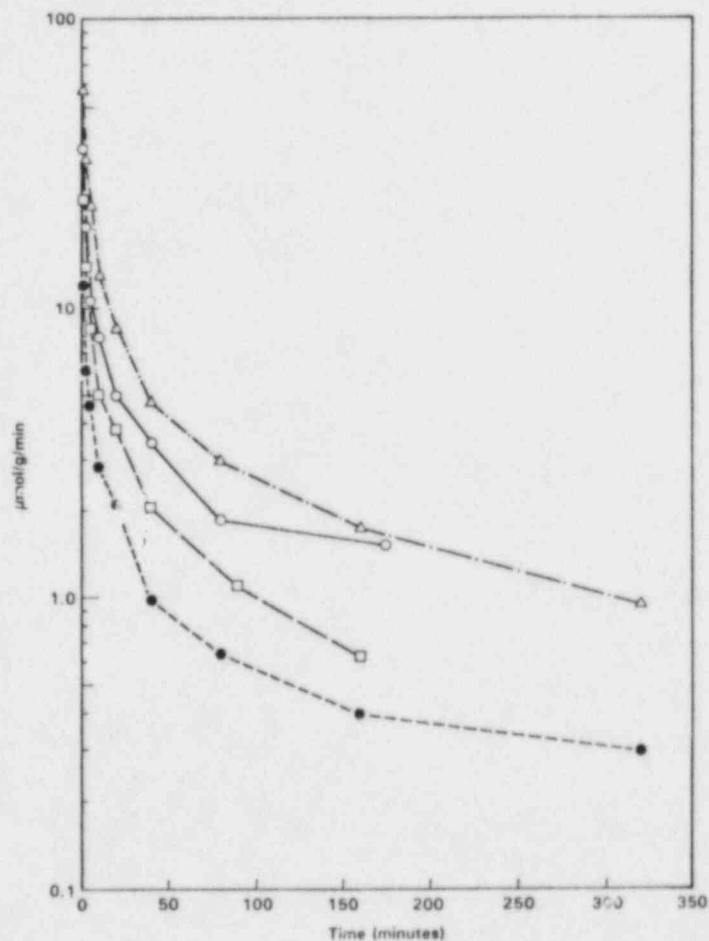


Figure 13. Iron extraction from trench sediments.

component. Similar results were obtained for sample 3-1 from above the water table. In this case, the Si:Al ratio is about 2:1, Fe:Si is 2:1 and 1:1, and Fe:Al is 3:1 and 2:1, respectively, for the first and second components.

Analyses of the extraction data, assuming either zero-order or first-order dissolution kinetics and using either linear or log regression equations (of the quantity of oxide phases extracted vs. time), were used to estimate the total quantity of each of the three oxide phases identified. Our approach further assumed a homogeneous mixture of Fe, Al, and Si oxides in which the rates of dissolution for each oxide were independent. These results are summarized in the first three columns of Table 21. The estimated quantities of material in Phase 1 are virtually instantaneously soluble. As a first approximation, this Phase 1 material is the quantity of amorphous oxide used to interpret the adsorption data in Section 7.

Table 21. Estimated quantities and approximate atomic ratios of Fe, Al, and Si in amorphous oxides extraction from the <425- μm fraction.

Sample Phase	Quantity $\mu\text{mol/g}$			Extraction Time (min)			Estimated Composition			
	Fe	Al	Si	Fe	Al	Si	Fe _x	Al _y	Si _z	
2-1	1	39.9	36.8	22.0	0	0	0	0.40	0.37	0.22
	2	60.0	20.2	27.5	29.8	29.0	34.9	0.56	0.18	0.26
	3	182.9	96.5	187.4	192	246	224	0.39	0.21	0.40
2-2	1	25.5	24.0	23.7	0	0	0	0.35	0.33	0.32
	2	44.2	13.1	16.2	14.2	13.7	12.8	0.60	0.18	0.22
	3	59.2	70.0	94.7	179	267	261	0.26	0.31	0.42
3-1	1	65.2	45.7	36.4	0	0	0	0.44	0.31	0.25
	2	98.2	22.4	45.0	17.1	13.5	17.2	0.59	0.14	0.27
	3	149.6	95.0	178.1	202	265	228	0.35	0.22	0.42
3-2	1	12.2	12.9	8.8	0	0	0	0.36	0.38	0.26
	2	13.1	7.7	8.4	10.3	16.0	17.8	0.45	0.26	0.29
	3	70.9	58.1	91.0	216	305	302	0.32	0.26	0.41

The estimated amounts of Fe in each phase by both the linear and log regression methods are listed in Table 22.

Table 22. Iron estimated by two regression methods.

	Linear - - - - $\mu\text{mol/g}$ - - -	Log - - -
Phase 1	25.5	25.6
Phase 2	44.2	42.7
Phase 3	59.2	63.2
Total	128.9	131.5

The two methods estimate very similar quantities in the three phases.

If each phase represents a mixed amorphous $Fe_xAl_ySi_z$ solid phase, then the slopes of the regression equations can be interpreted as suggesting possible mineral compositions as is also shown in columns 4 to 6 of Table 21. In each case, the rate of release of Fe compared to Al or Si in the first phase is about 1.5 to 2 times that in the second phase.

The general characteristics of the phases are as follows: Phase 1 has moderate Fe and Al and low Si; Phase 2 has high Fe, low Al and Si; and Phase 3 has moderate Fe and Si and low Al. Interpretation of these data in FY84 and future experiments may determine whether single Fe, Al, and Si oxides or mixed $Fe_xAl_ySi_z$ oxides represent a more realistic interpretation of the extraction data. Information on whether the adsorbing phases in soils are single or mixed amorphous phases may be significant for the interpretation of adsorption data on real soils in Section 7.

6.3 Conclusions

- The hydroxylamine hydrochloride extraction technique of Chao and Zhou (1983) used for time sequence extractions proved to be an excellent method for determining the quantities and possibly the composition of amorphous oxide phases in several soils studied.
- This extraction technique together with curve-stripping and sequential regression of quantities extracted vs. time (for various time intervals) may prove useful with other sediments from other locations to estimate the quantities of amorphous oxide sinks responsible for radionuclide adsorption.
- Three amorphous oxide phases, each differing in its solubility and Fe:Al:Si composition, were identified in each of the four sediment samples examined.

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7. ADSORPTION

A potentially important mechanism for retardation or removal of radionuclides in groundwater is adsorption onto amorphous manganese and iron oxides, and organic compounds that have a high adsorption affinity for cationic or anionic forms of radionuclides. For each individual radionuclide, these soil/sediment constituents have distinct adsorption affinities (binding constants) and adsorption capacities (quantity of binding sites per gram of sediment). Experimental measurement of the adsorption affinity and capacity of these individual sinks for a particular radionuclide, combined with determination of the quantities of individual sinks present in a particular soil or sediment provides the information base necessary to test the validity of surface coordination modeling approaches to describing adsorption phenomena in real soil systems. In its simplest form, the surface coordination modeling approach describes the total adsorption of a radionuclide on all the sinks present in a real soil or sediment as a linear combination of the adsorption on the individual sinks, where the adsorption on each sink is described by the product of the adsorption affinity, the capacity of the individual sinks, and the quantity of the sink present in the soil or sediment.

The objective of this task is to 1) experimentally determine the adsorption behavior of the ^{235}Np on the sediments, 2) test the hypothesis that iron oxides dominate the adsorption behavior of $\text{Np}[\text{V}]$ on these sediments, and 3) test the alternate hypothesis that $\text{Np}[\text{V}]$ adsorption on the sediments can be described as linear combination of Np adsorption on the individual sinks quantified in Section 6 of this report.

7.1 Adsorption On Amorphous Iron Oxides

To test the hypothesis that Np adsorption behavior is dominated by amorphous iron present in the sediment, the Np adsorption on synthetic amorphous iron oxyhydroxide must first be characterized. The neptunium adsorption on amorphous iron oxyhydroxide ($\text{Fe}_2\text{O}_3 \cdot \text{H}_2\text{O}(\text{A})$) was investigated at 25°C for $\text{Np}[\text{V}]$ concentrations between 4.5×10^{-11} and 4.5×10^{-13} M in 0.1 M NaNO_3 solutions in contact with air. The $\text{Fe}_2\text{O}_3 \cdot \text{H}_2\text{O}(\text{A})$ was prepared and aged in a teflon container using the procedure described by Benjamin (1979). The suspension was used within 24 hours for adsorption experiments. Adsorption experiments were performed in individual polyethylene tubes containing iron suspension in contact with air ($P(\text{CO}_2) = 10^{-3.5}$ atm). Suspensions were in a 0.1 M NaNO_3 supporting electrolyte. The time required for $^{235}\text{Np}[\text{V}]$ adsorption to reach equilibrium was determined for suspensions of 10^{-2} to 10^{-3} M total iron, at $\text{pH} = 5.7$ and 6.7. Contact times were varied between 1 and 96 hr. Since the

adsorption of Np[V] had reached 95% of its 96-hr value within three to four hours, with essentially no change after 24 hr, 3- to 4-hr contact times were used to determine the adsorption for a fixed initial Np concentration on a range of amorphous iron concentrations that bracket those found in the sediments to be investigated in FY84.

In the range of pH 5 to pH 9, over which the adsorption experiments were conducted, the aqueous speciation of Np[V] is dominated by the neptunyl oxyanion, NpO_2^{2+} , except above pH = 8.5 where $\text{NpO}_2(\text{CO}_3)^-$ begins to dominate (Figure 14). The species distribution was calculated using MINTEQA2 (Felmy et al., 1984) using the stability constants shown in Table 23.

For each adsorption curve (Figure 15), the adsorption "edge" is defined as the narrow band of pH for which the percent adsorption increases

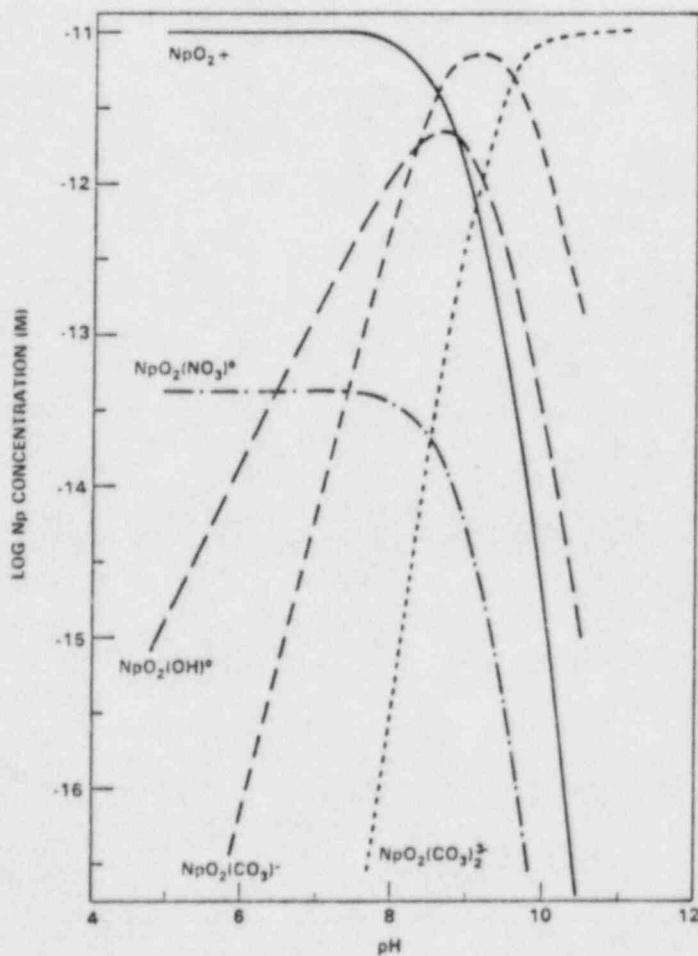


Figure 14. Neptunium species distribution over a range of pH.

Table 23. Aqueous neptunium reactions and stability constants used for determining adsorption constants.

	Reaction	PK	Reference
Hydroxide	$\text{NpO}_2^+ + \text{H}_2\text{O} = \text{NpO}_2(\text{OH})^0 + \text{H}^+$	8.85	*Kraus and Nelson, 1948
		8.75	Schmidt et al., 1980
		8.89	Sevast'yanova and Khalturin, 1976
		9.12	Maya, 1983
Nitrate	$\text{NpO}_2^+ + \text{NO}_3^- = \text{NpO}_2\text{NO}_3^0$	1.6	*Danesi et al., 1971
	$\text{NpO}_2^+ + 2\text{NO}_3^- = \text{NpO}_2(\text{NO}_3)_2^-$	1.4	*Danesi et al., 1971
Carbonate	$\text{NpO}_2^+ + \text{CO}_3^{2-} = \text{NpO}_2(\text{CO}_3)^-$	-4.5	*Maya, 1983
	$\text{NpO}_2^+ + 2\text{CO}_3^{2-} = \text{NpO}_2(\text{CO}_3)_2^{3-}$	-7.1	*Maya, 1983

*Used for determination of adsorption constant.

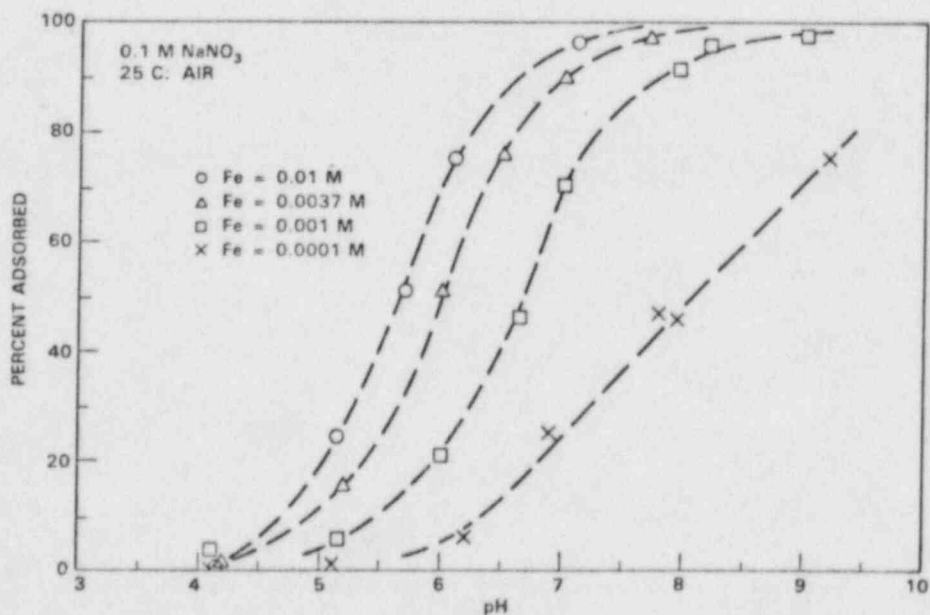


Figure 15. NpO_2 adsorption on amorphous Fe oxyhydroxide: Total Np = 4.7×10^{-12} M.

rapidly with pH. For the range of conditions studied, the adsorption edges occur between pH 5.5 and 6.5. For pH 9.5 the quantity of Np adsorbed approaches 100% even in the presence of atmospheric CO₂. The shift of the adsorption edge to higher pH as the quantity of iron available for adsorption decreases is characteristic behavior for adsorption of cations onto oxides (Leckie et al., 1980; Benjamin and Leckie, 1981) The data in Figure 16 have been described by a single reaction and adsorption constant (Girvin et al., 1984).

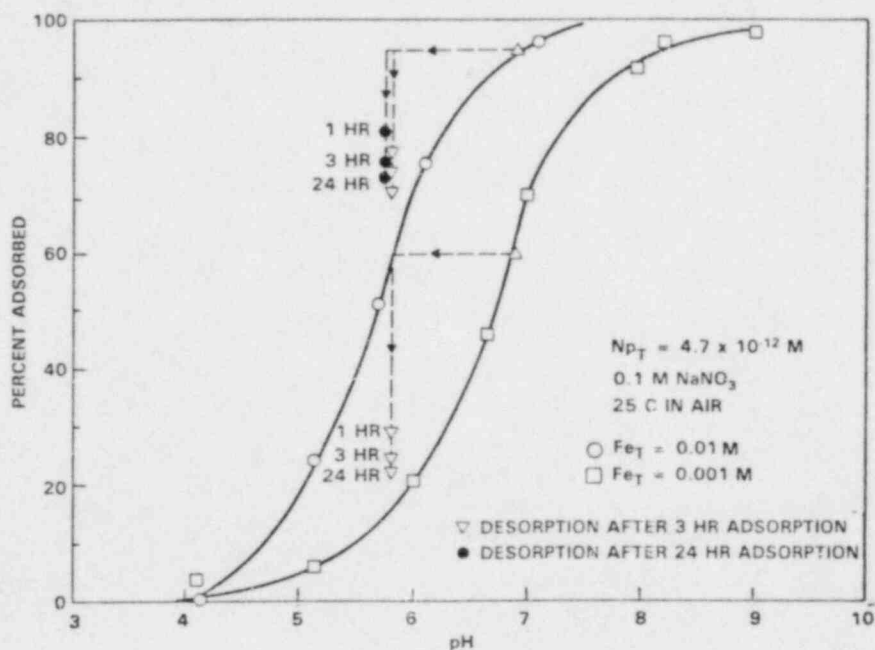


Figure 16. Adsorption/desorption of NpO₂ on amorphous Fe
Desorption path by pH adjustment.

7.2 Desorption From Amorphous Iron Oxide

The reversibility of the Np adsorption reactions were examined by determining the extent of the Np desorption resulting from shifts in pH (Figure 16). The solid lines are four-hour adsorption curves for 10⁻² and 10⁻³ M iron suspensions. Separate adsorption experiments for 3- and 24-hr contact times were conducted in the 10⁻² M iron suspension. The pH of these samples was then adjusted and the desorption followed for 24 hr. A large fraction of the Np was desorbed rapidly, within three hours, from both the 3- and 24-hr contact samples. This is followed by a slower desorption component, which is approaching the 4-hr adsorption curve at that final pH. Despite the apparent trend, the differences in the desorption between the 3- and 24-hr contact time samples are not

significant since the differences are less than or equal to the experimental error of $\pm 3\%$ on the percent adsorption scale. The data show that the adsorption and desorption rates differ and that 75% to 90% of the Np, which is going to desorb at the new pH, desorbs within 24 hr. However, without additional data, e.g., at 96 to 144 hr, we cannot establish the complete reversibility of the Np adsorption/desorption reaction.

7.3 Adsorption on the Sediment

The testing of the validity of the linear surface coordination model approach to describing adsorption of Np on the sediment will use the quantities of individual sinks in the sediment (Section 6) and the adsorption affinities of individual sinks obtained from literature adsorption data or obtained experimentally. Compositated sediment samples ($< 425 \mu\text{m}$) 2-1, 2-2, 3-1, and 3-2 as described above (Section 6.1) were used for adsorption experiments. The data characterizing these sediments are given in Tables 17, 18, and 21.

Adsorption experiments were conducted using Np[V] at several values of pH and solid to solution ratios that encompass actual field conditions at the site. These experiments were conducted using a synthetic spring water, the composition of which is given in Table 24. The cation and anion concentrations given are average values of analyses on spring waters collected during 1981 and are in good agreement with 1983 spring water composition (Appendix B). The quantity of aqueous Np adsorbed varies as a function of pH; the solid:solution ratio, S ; the initial aqueous Np concentration, C_I ; and time, t . The time to reach equilibrium is given as t_{equil} . The batch adsorption experiments were conducted in individual acid-leached polyethylene bottles at 25°C . Samples were gently shaken throughout each experiment except when daily pH adjustments were made by adding HCl or NaOH. The final solution composition of all samples were determined by ICP, IC, and total organic carbon (TOC) analyses.

To encompass the natural pH of the sediment-water system ($\text{pH} = 8.3$), adsorption experiments were performed at pH values between 6.0 and 10.0. Unless otherwise noted, the pH of individual samples were initially adjusted to the desired pH and allowed to equilibrate for a period of 120 hr prior to the addition of the pure ^{235}Np spike. During this "pre-adsorption equilibration period" samples were gently shaken at 25°C , except during the daily pH adjustment. This pre-adsorption equilibration period allows sediment surfaces to readjust, via surface hydration, protonation, and dissolution/precipitation reactions, to the new experimentally imposed pH before adsorption experiments are initiated. The pH drift (diagonal lines) and the pH adjustment

Table 24. Composition of synthetic spring water used in soil adsorption experiments based on ICP, IC, and TOC analyses.

Constituent	Concentration (mg/L)
Ca ²⁺	15.2
Mg ²⁺	4.0
Na ⁺	4.2
K ⁺	5.9
Sr ²⁺	0.01
SO ₄ ²⁻	14.3
Cl ⁻	2.3
PO ₄ ³⁻	0.4
Al	0.006
Si	4.6
Fe	0.003
Mn	<0.0001
Alkalinity	58 (as HCO ₃ ⁻)
DOC	1.1 ppm

(vertical lines) made during this pre-adsorption equilibration period are shown in Figure 17 for the pH 6, 9, and 10 samples. The pH drift and adjustments over the time span of the adsorption experiments are shown from 120 hr to 312 hr. The pH drift begins to decrease after 120 hr and is relatively minor between 250 and 312 hr, except for the pH 6 samples. Figure 18 shows the change in the dissolved Al concentration during the pre-adsorption equilibration period and the adsorption experiments. At pH = 6 there is very little change in the Al concentration with time, with respect to the concentration of Al in the synthetic spring water, which is indicated by the asterisk along the vertical concentration axis. At pH = 8, 9, and 10, greater than 75% of the total observed change in concentration occurs during the first

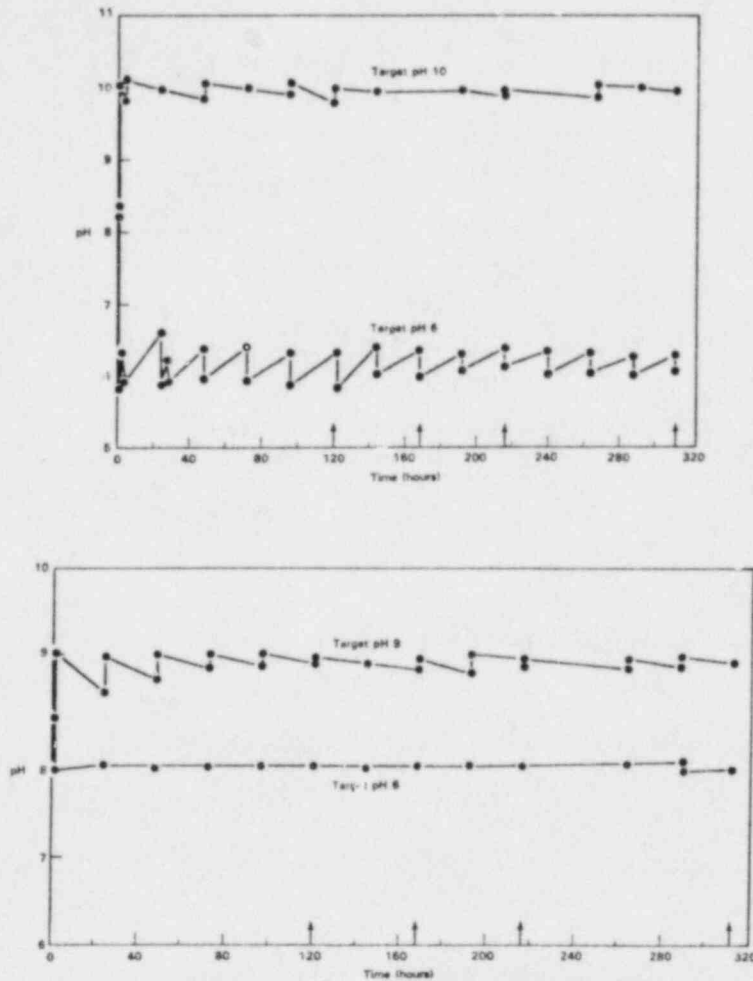


Figure 17. Drift and adjustment of pH during the pre-adsorption equilibration.

120 hr. These results suggest that the 120-hr pre-adsorption equilibration period is a sufficiently long time to avoid starting the adsorption experiments during the initial rapid readjustment period following the initial pH adjustment of the samples.

The adsorption rate as a function of pH (Figure 19) has been determined for two adsorption experiments using sediment 2-1. The time plotted here is the time after the start of the adsorption experiments, i.e., for the pre-equilibrated samples add 120 hr to the times in Figure 19. These data indicate that for $C_I = 1.0 \times 10^{-12}$ M, adsorption reaches a steady state after a 96-hr contact time for all pH values. Similarly, the pH = 8, non-pre-equilibrated samples also reached a steady state after 96 hr, however the percent adsorbed is consistently 10% higher for these samples. The non-pre-equilibrated samples were spiked with Np and

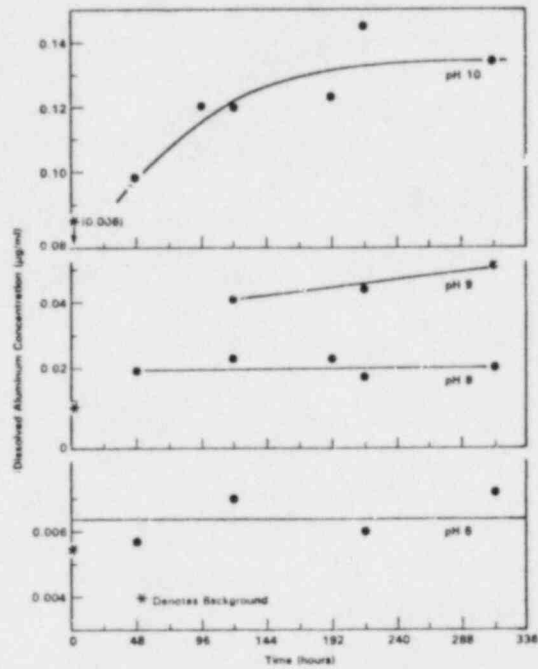


Figure 18. Changes in dissolved Al concentrations during adsorption experiments.

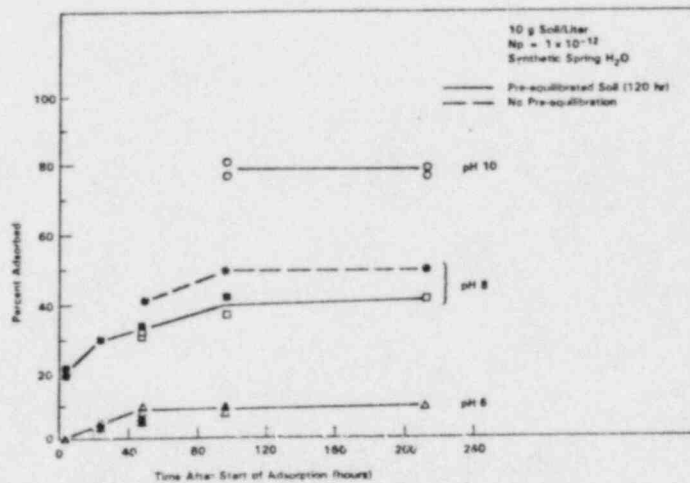


Figure 19. Adsorption rate as a function of pH.

pH adjusted 12 hr after the synthetic spring water was added. The pH during this 12-hr period was 8.3, i.e., the natural pH of the sediment water system. A comparison of the amount adsorbed on pre-equilibrated and non-pre-equilibrated samples will be made in FY84 at pH = 6 and 10 to determine the magnitude of the difference as a function of pH. That this difference exists and may exist at other values of pH and that the time to reach a steady state is the same for pre-equilibrated and non-pre-equilibrated samples raises questions regarding the necessity for, and the effect on the sediment surfaces of, pre-equilibration. In FY84 experiments will be conducted using Np-soil contact times of 100 hr and higher solid:solution ratios. The 100-hr contact time should ensure steady state conditions.

The adsorption data for a contact time of 192 hr are shown in Figure 20 for solid:solution ratios of 10 and 100 g of sediment/liter. In FY84 additional data will be obtained for the latter ratio to substantiate the expected pH dependence indicated by the dashed line. The point indicated by an "x" was not pre-equilibrated and shows a higher percent adsorbed for that pH than the pre-equilibrated sediments. The superposition of these data on the curves from Figure 15 is shown in Figure 21. Based on the rapid hydroxylamine hydrochloride extraction data in Table 21 the quantity of amorphous iron in sediment 2-1 (Phase 1) is 40 μmol s Fe/g of sediment. Based on this value, the 10 and 100 g/L data in Figure 21 correspond to 4×10^{-4} and 4×10^{-3} M Fe, which allows a direct comparison to be made between the amorphous iron and sediment adsorption edges. Although more data are required to complete the 100 g/L curve the available data are consistent with oxide control of Np[V] adsorption in the sediment (since the sediment TOC concentrations are low).

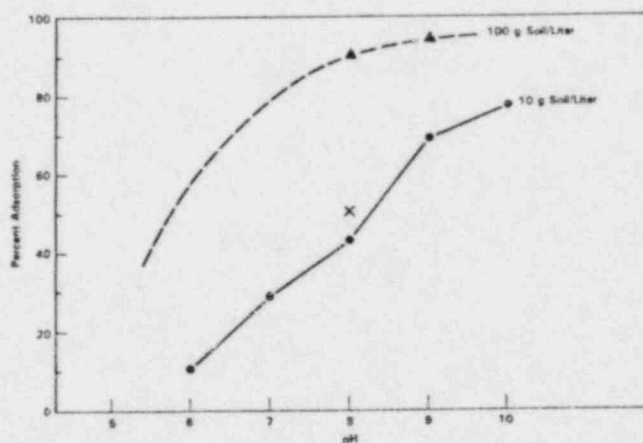


Figure 20. Effect of solid:solution ratio on adsorption.

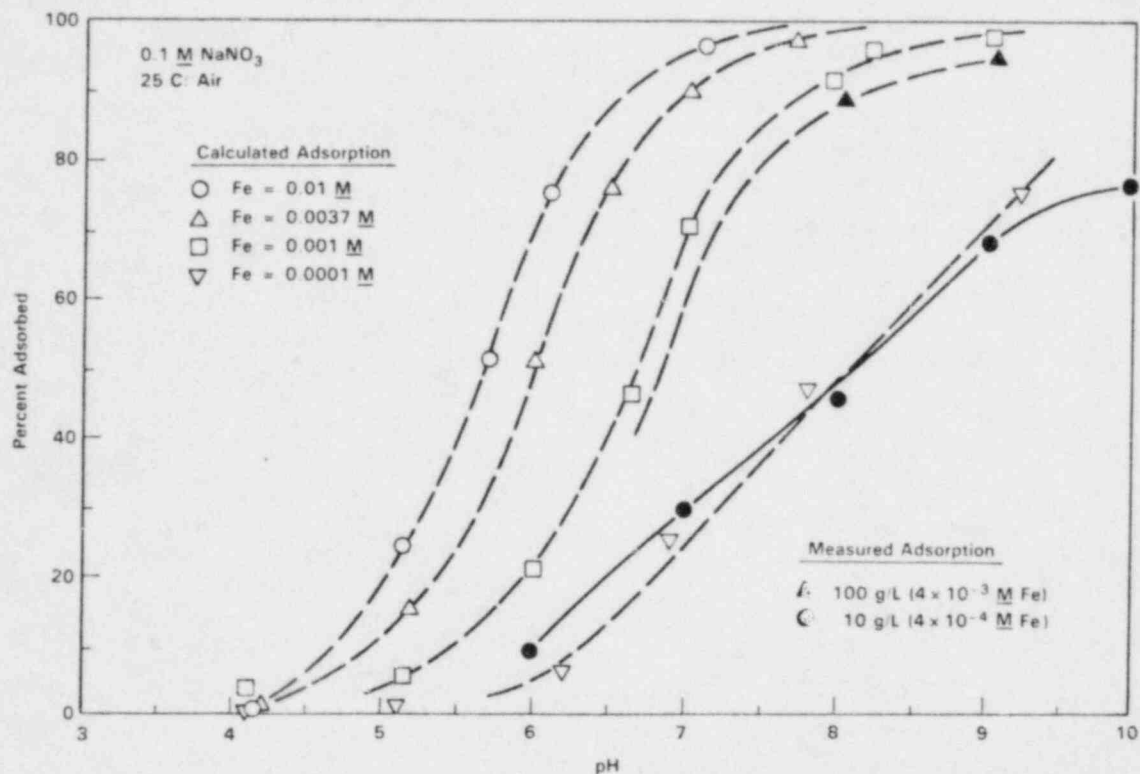


Figure 21. Comparison of calculated and measured adsorption.

The thenoyl-trifluoro-acetone (TTA) extraction technique (Burney et al., 1974) was used to determine the oxidation state of Np in the solution contacting the sediments. These oxidation state analyses were performed on the filtered ^{235}Np solutions before they contacted the soil. In addition, oxidation state analyses were performed on suspension filtrates that had been in contact with the soil for 100, 192, and 600 hr. In all cases, greater than 97% of the Np in solution was Np[V]. Within the $\pm 4\%$ error of these extractions no Np[IV] was observed. These results represent a necessary but not sufficient condition for establishing that the Np bound to the soil is Np[V]. It is possible that Np[V] in solution is adsorbed and reduced on the soil surface to Np[IV]. Since equilibrium is reached in these experiments, exchange of bulk solution and bound Np may introduce some Np[IV] into solution. However, since these experiments were performed in contact with atmospheric oxygen, Np[IV] leaving the solid and entering solution could be rapidly oxidized to Np[V] (J. L. Swanson, PNL, personal communication). Thus, the absence of Np[IV] in the solution in contact with the soil, even at 600 hr, is consistent with, but does not prove, that the adsorbed Np is in the +5 oxidation state.

7.4 Conclusions

- Np[V] is strongly bound to amorphous iron ($\text{Fe}_2\text{O}_3 \cdot \text{H}_2\text{O}$) and is described by surface coordination models of adsorption.
- Np[V] adsorption on $\text{Fe}_2\text{O}_3 \cdot \text{H}_2\text{O}$ approaches complete reversibility within 24 hr.
- The Np adsorption data obtained to date on these low organic carbon soils are consistent with the hypothesis that amorphous iron oxides or amorphous oxides in the soil determine the adsorption behavior of the neptunyl oxy cation NpO_2^+ .
- Oxidation state analysis of Np in soil suspensions is consistent with, but does not prove, the hypothesis that Np[V] adsorbed from solution exists as Np[V] while bound to these soils.

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8. GEOCHEMICAL MODELING

Geochemical models can be used to predict the speciation of radionuclides in ground water and to provide a more generic interpretation of the data collected at the low-level disposal site. For example, the aqueous speciation of the radionuclides can be used to identify the particular complexes that are mobile in the ground water. This information can in turn be used to examine the possible effects of alterations in the aqueous chemistry on radionuclide mobility. Geochemical models can also be used to identify solid phases that are in equilibrium or are oversaturated with respect to the ground waters. X-ray analysis of soils can be used to identify the major phases and the crystalline phases, but much more complex and time-consuming techniques must be used to identify the much smaller amorphous phases that can be very important in determining the solution chemistry and retardation of radionuclides. Geochemical models can complement the complex techniques and can be very important tools in identifying solid phases in the soils. Geochemical models can also be used to determine the detection limits and components necessary for identification of aqueous species and solid phases.

The objective of this task is to use the geochemical model, MINTEQ to 1) identify the specific aqueous species of the radionuclides that are mobile in these groundwaters, and 2) identify solid phases that may be controlling the radionuclide concentrations in the groundwaters.

8.1 Method

Water samples collected in March and June 1981 (Robertson et al., 1981) and in January, February, March, May, June, and September 1983 (Section 3.0), which included both analyses for radionuclide charge-form and inorganic composition, were used in the thermodynamic speciation modeling. Although the charge-form speciation for several radionuclides was determined in February 1974 (Robertson and Perkins, 1974) and in September 1981 (Robertson et al., 1981), water quality data from which the aqueous speciation of the radionuclides could be calculated were not available. In all, 37 different radionuclides or isotopes have been identified in trace quantities in the waters at the site. Several of the radionuclides were isotopes of the same element, and for some of those, thermodynamic data were not available. The charge-form speciation was calculated for a total of 22 different radionuclides.

The MINTEQ geochemical model (Felmy et al., 1984), a descendant of the MINEQL (Westall et al., 1976) and WATEQ3 (Ball et al., 1980) geochemical models, was used to calculate the aqueous speciation of most of the

radionuclides. The charge-form distribution was computed from the sum of the activity of each species in a particular charge-form. MINTEQ contains well referenced thermodynamic data for Cs, Fe, I, Mn, Na, As, Ba, Sr, Ni, and Zn (Ball et al., 1980); Cr documentation is in preparation (Schmidt, 1984). The speciation calculations for Ce, Co, Zr, Eu, Ru, Pm, Tc, Np, and Sb were calculated by hand using the thermodynamic data in available reviews (Rai and Serne, 1978; Rai and Zachara, 1984). These hand calculations are not considered to be as accurate as the MINTEQ model calculations because 1) only those complexes expected to be dominant are considered, 2) the hand calculations are not iterative, and 3) the thermodynamic data from compilations used have not been subjected to further critical review. When the concentration of a constituent is reported as less than a detection limit, the concentration of that component is estimated as equal to one-half the reported detection limit.

Neither the analytical nor the thermodynamic based charge-form speciation is a priori assumed to be correct. The accuracy of the analytical distribution is limited by 1) lack of replicated analytical charge-form analyses, 2) variability in chemistry of the source waters, 3) improper partitioning on the resins, and 4) counting errors. The magnitude of the total errors is not possible to ascertain from the available data. Duplicates of plutonium determinations yielded $\pm 9.0\%$ of the mean (Robertson and Abel, 1980). A validation study of the BLVWS is currently under way. The accuracy of the geochemical model calculations depends on 1) the accuracy of the thermodynamic data, 2) inclusion of all relevant solute complexation reactions, 3) correctness of the computer code, 4) accuracy of the dissolved constituent analyses, and 5) the extent to which local equilibrium is obtained. Errors in the charge-form distributions determined analytically or by modeling cannot be reliably estimated at this time. Thus, it is necessarily assumed that coherence of results between the two techniques is evidence that both approaches are correct for that element and the water chemistry involved. Where the two approaches do not yield coherent results, we give possible reasons for the discrepancy.

8.2 Charge-Form Speciation

Of the 22 radionuclides for which analytical charge-form speciation data were available, very good agreement between the analytical and calculated charge-form speciation was obtained for 13 (Appendix Tables C.1 through C.6). These radionuclides are ^{24}Na , ^{54}Mn , ^{76}As , ^{82}Br , ^{90}Sr , ^{99}Mo , $^{99\text{m}}\text{Tc}$, $^{131,133,135}\text{I}$, $^{134,137}\text{Cs}$, ^{140}Ba , $^{141,144}\text{Ce}$, ^{151}Pm , and ^{239}Np . These radionuclides tend to have less complicated redox and fairly well known complexation chemistry and, with the exception of Mn, As, and Tc, are not redox sensitive. Cesium, for example, only forms

two aqueous inorganic complexes in natural and polluted waters (CsOH^0 and CsCl^0), hence, Cs occurs in solution predominantly as the uncomplexed species Cs^+ . Similarly, I, Na, Sr, Ba, Br, and Pm form only weak inorganic complexes, hence, are predicted to occur predominantly as I^- , Na^+ , Sr^{2+} , Ba^{2+} , Br^- , and Pm^{3+} . This corresponds very well to the analytical speciation measurements for these radionuclides in both the influent and groundwater except that approximately 50% of the I was retained on the cation resin at one sampling date (June 1981) and $\geq 16\%$ of the Pm was retained on the alumina bed in the one sample taken in February 1974. More recent data (Robertson et al., 1983) suggest that some of the anionic iodine is present in an organic form. The dominant species of Mo, Tc, and Np from thermodynamic calculations are the monovalent oxy complexes, MoO_4^- , TcO_4^- and NpO_2^+ . This also corresponds very well to the analytical charge-form measurement for these radionuclides (Appendix Table C.6). Thermodynamic-based speciation calculations for cerium indicate that nearly 100% of the cerium occurs as the $\text{Ce}(\text{OH})_2^{2+}$ complex; the Ce^{4+} and CeOH^{3+} complexes only become important at much lower pH values. The significant portion of anionic Ce found in the influent sample is unexplained, although, this sample calculates to be oversaturated with respect to Ce oxide.

Although Mn is a redox sensitive element, it is known to equilibrate with Fe (Jenne, 1968), which is probably the major redox mediating couple of natural and polluted waters. Thus, Mn^{2+} is the dominant species in most fresh waters, with small amounts of the MnHCO_3^+ and MnSO_4^0 complexes formed. The predominance of the cationic form agrees with the results of the analytical charge-form speciation.

For nine of the radionuclides, significant discrepancies were noted between calculated and analytical charge-form speciation. One possible explanation for the observed discrepancy is organic complexation of the radionuclides. A significant limitation to the thermodynamic speciation discussed above is that only the formation of inorganic complexes is accounted for. This is due to the general lack of thermodynamic data for natural organic complexes. The organic analyses of the influent, trench, well, and spring waters (Robertson, et al., 1981; Section 4.0) indicates that only a few percent of the dissolved organic content was of low molecular weight and volatile (i.e., analyzable by GC/MS) and could be identified as specific compounds. Thus, compilations of complexation constants for specific organic compounds, as in Smith and Martell (1976), are of limited value in estimating the effect of organic complexation. The total organic carbon content of these waters varies from approximately 1 ppm to almost 15 ppm, which is in the range of natural waters where organic complexation of transition metals is recognized to be significant.

Organic complexation may explain the discrepancy between the analytical and calculated charge-form distributions for Fe, Co, Ni, and Zn and possibly Cr, Zr, and Eu (Appendix Tables C.7 to C.10). Both Fe(II) and Fe(III) form very strong complexes with fulvic and humic acids in soils (Schnitzer, 1969; Malcolm et al., 1970) and with many other organic compounds such as those identified in the influent water samples. Citrate was identified as a major constituent in the September 1981 influent sample. Using the log K_r value given in Martell and Smith (1977) for the formation of the Fe(II) and Fe(III) citrate complexes, we estimated that a citrate concentration of 0.65 mg/L (i.e. about twice the concentration measured in the September 1981 sample) would result in agreement between the calculated charge-form speciation and the analytical charge-form speciation for the June 1981 sample, because the citrate complex has an anionic charge. Since the concentration of citrate is only a small fraction of the total organic carbon, it is possible that the remaining organic matter could account for the observed complexation. The observed dominance of the anionic charge-form in trench, well, and spring waters suggest that the iron is forming anionic complexes with the fulvic and humic acids, resulting in a reduction in the percentage of the iron in cationic charge-form. Iron, cobalt, nickel, and zinc all are recognized to form complexes with fulvic and humic acids in soils although not with the same strength (Schnitzer, 1969; Malcolm et al., 1970). As with iron, if cobalt is assumed to form an anionic complex with the fulvic and humic acids in the groundwater, then the observed discrepancy between the calculated and analytical charge-form distribution could be explained. Europium speciation in the groundwater (Appendix Table C.8) indicates a pattern similar to that shown by iron and cobalt suggesting that europium may be forming organic complexes in the groundwater. The thermodynamic speciation (Rai and Serne, 1978) suggests that Eu would form only cationic complexes, yet the analytical speciation results indicate a predominance of the anionic charge-form in the groundwaters just as was noted for iron and cobalt and ascribed to organic complexation. The potential for organic complexation of europium in the groundwaters needs to be evaluated.

Although it might also be assumed that zinc and nickel would form similar anionic complexes with the fulvic and humic acids, the discrepancy between the calculated and analytical charge-form speciation cannot be explained by the presence of an anionic complex. The discrepancy in the zinc speciation results is due to an increase in the cationic charge-form in the analytical measurements over that predicted from the thermodynamic speciation (Appendix Table C.10). The thermodynamic data for zinc are considered to be reliable and redox does not effect zinc; therefore, the most likely cause for the discrepancy is complexation by dissolved organic compounds, although the nature of the complex formed needs to be identified. Likewise the discrepancy noted

between the analytical and calculated speciation for nickel is due to the presence of a cationic form in the influent and trench water, which is not predicted based on the thermodynamic data (Appendix Table C.9). The role of organic complexation of nickel needs to be further evaluated and the form of the complex identified. The formation of a positively charged organic complex would also explain the discrepancy that has been observed between the analytical and calculated speciation for Cr (Appendix Table C.10). Although Cr is not significantly complexed by natural organic material (Nakayama et al., 1981), Cr does form citrate and other chelator complexes in accordance with other +III metals. Thus the discrepancy between the calculated and analytical speciation in the influent and trench waters may be due to organic complexation with chelators that are present; whereas, the agreement between the charge-form distribution at the spring is indicative of the low complexation by fulvic and humic acids.

Redox sensitive elements are generally in thermodynamic disequilibria in surface and groundwater (Jenne et al., 1980; Jenne, 1981) because such redox reactions generally have slow reaction rates. Antimony, Sb, was the only one of the redox sensitive radionuclides to show poor agreement between the calculated and analytical charge-form speciation (Appendix Table C.11). The analytical and calculated charge-form distributions for manganese, technetium, and arsenic, which are all redox sensitive, agree very well. Manganese and technetium have been discussed above. Arsenic occurs as an anionic complex in both the oxidation states observed in the pH range of the influent and ground waters (7.0 to 9.5) in accordance with the analytical results. Thus the charge-form distribution cannot be used to determine if arsenic is in redox equilibrium. The thermodynamic speciation for antimony using the measured platinum electrode potentials suggests that the oxidation state of the Sb would be +III in the influent and +V in the groundwater (Rai and Serne, 1978). However, the analytical charge-form speciation shows more than half of the Sb was retained on the anionic and neutral absorbants; this would occur if a significant fraction of the Sb was in the +V oxidation state (Sb(OH)_6^-) in the influent. In the groundwater, the Sb was predominantly in the nonionic charge-form, which suggests it was in the +III oxidation state (HSbO_2^0). If the analytical results are assumed to be accurate, this would suggest that the antimony is being reduced in the groundwaters, which is opposite of the predicted behavior of the redox couple in oxygenated groundwater. These results suggest that redox equilibria for antimony may not occur for this system or that the analytical charge-form speciation is in error.

Ruthenium is the final radionuclide for which calculated and analytical charge-form distributions were not in agreement (Appendix Table C.12). The dominant charge-form in the influent and trench waters was cationic.

On several sampling dates a significant percentage of the ruthenium occurred in the anionic charge-form, especially in the influent. In the groundwater and the spring water samples, the dominant charge-form determined analytically was anionic. Based on the thermodynamic data tabulated by Rai and Serne (1978) and assuming equilibrium with the redox potential measured using the platinum electrode, ruthenium in all the waters would be in the +IV oxidation state occurring predominantly as the $\text{Ru}(\text{OH})_2^{2+}$ complex. Only when the pe is above 16.81 would the +VII oxidation state be dominant and the dominant aqueous species would be the anionic RuO_4^- complex. Because the thermodynamic data for ruthenium complexes are generally considered to be poor (Baes and Mesmer, 1976) and thermodynamic data for ruthenium complexes with common ions such as sulfate, nitrate, and carbonate do not exist, it is very likely that the observed discrepancies are due at least in part to the poor quality of the thermodynamic data.

8.3 Solid Phases

As the waters from the influent infiltrate through the bed and surficial aquifers to the springs, radionuclides are removed differentially from the water. Radionuclides can be removed from the water either by precipitation of "pure" phases, co-precipitation in solid-solutions, or sorption onto the surface of existing solids. The role of sorption is being examined in another of the project tasks (Section 7.0). The geochemical model can be used to determine the solid phases that are in equilibrium with or oversaturated in these waters. The geochemical model can also be used to evaluate the analytical data by testing for oversaturation of the solution with respect to certain well recognized solubility controls, solids known to readily precipitate from oversaturated solutions, or solids identified in the sediments.

Quartz and calcium-sodium feldspar were identified by x-ray diffraction as the major minerals in the soils at the site (Robertson et al., 1983). The saturation indices for the silicates in the spring water (Appendix Table C.13) indicate that the solution is slightly oversaturated with respect to the highly crystalline phase, high-temperature-forming quartz; however, it is in equilibrium with the more amorphous silicates, cristobolite and chalcedony. This concurrence between the calculated solid phase and the observed solid phase implies that the analytically determined silica values are not in error. The saturation indices for the silicates indicate that the influent and trench waters are undersaturated with respect to the silicates and that the saturation indices increase as the water travels from the trench to the springs (Appendix Table C.13). This suggests that for this groundwater system, the silicates are dissolving and that the undersaturation with respect to quartz identified in the soils is due to slow kinetics of dissolution.

The saturation indices for the two end members of the sodium feldspar series, albite and anorthite, indicate that the influent, trench, and groundwaters of the site are undersaturated (Appendix Table C.14). Undersaturation of the groundwater with respect to the feldspar is due to the very slow kinetics of dissolution (Correns, 1969; Stumm and Morgan, 1981). The increase in the saturation indices as the groundwater travels from the trench to the springs suggests that the feldspars are also dissolving.

The solubility calculations for the groundwaters (i.e., the well and spring waters) indicate that these waters are in equilibrium with calcite, CaCO_3 , (Appendix Table C.13) that was identified in the soils (Robertson et al., 1983). The presence of calcite in the groundwater is particularly important because several transition elements that form carbonate solid phases are known to co-precipitate with calcite when their concentrations are below the levels at which their distinct carbonate phases would form (Holland et al., 1964; Jenne and Wahlberg, 1968; Kitano et al., 1968; Pesret, 1976; Lorens, 1981; Shanbhag and Morse, 1981). Some of the radionuclides that could potentially co-precipitate during calcite formation are ^{90}Sr , ^{140}Ba , ^{54}Mn , ^{65}Zn , ^{60}Co , and ^{54}Fe . The saturation indices for the influent and trench waters indicate that these waters generally are not in equilibrium with calcite. The influent water samples from March and June 1981 indicate undersaturation with respect to calcite. However, the trench waters are calculated to be undersaturated, oversaturated and in equilibrium with calcite on different sampling dates. From these data it is impossible to determine if calcite would precipitate from these waters.

Aluminosilicates, particularly the zeolites, are very effective in removing ions from solution through sorption and ion exchange. Thus the presence of aluminosilicates and aluminum hydroxides in the groundwater system could account for the observed removal of many radionuclides. For example, phosphorous is strongly sorbed to aluminum hydroxides such as gibbsite (Onishi et al., 1981). Radioactive cesium, cobalt, and strontium have all been observed to be removed from solution by ion exchange on or precipitation with aluminosilicates (Jenne and Wahlberg, 1968; Onishi et al., 1981). Cesium is especially strongly adsorbed by micas such as illite and biotite that have been identified in the soils. The saturation indices for both the influent and spring waters in the 1981 samples and the one 1983 sample (i.e., last manway 2-15-83), for which both aluminum and silica values were available, indicate that these waters are in equilibrium with allophane(a) (Appendix Table C.15). Halloysite, laumontite at the spring, kaolinite, montmorillonite, illite, and gibbsite are calculated to be oversaturated in the ground waters (Appendix Tables C.15 and C.16). The oversaturation, especially with respect to both gibbsite and kaolinite, suggests that the analytically determined aluminum concentrations may be too high, because the

aluminum activity in soils is commonly controlled by at least one of these solids phases (Lindsay, 1979). Even when the aluminum concentration is estimated to be one-half the detection limit in the 1983 samples (i.e. 15 or 25 mg/L) many of the aluminosilicates are calculated to be oversaturated in at least one of the water samples (Appendix Tables C.15 and C.16). All solutions are calculated to be oversaturated with respect to kaolinite and montmorillonite. Kaolinite is a weathering product of feldspar (Correns, 1969) and has been identified in the soils at the site (Robertson et al., 1983), therefore equilibrium with kaolinite would be expected. Overestimation of the concentration of aluminum in solution by up to an order of magnitude or more is a common analytical problem unless a $<0.1 \mu\text{m}$ membrane is used to filter the water sample because of the presence of aluminum colloid or clay particles that pass through the $0.45\text{-}\mu\text{m}$ membranes (Kennedy et al., 1974). A more accurate determination of the aluminum concentration in solution is necessary to determine which of the unidentified aluminosilicates is in equilibrium with the water; from this, the radionuclides that would most likely be removed from the groundwater can be identified.

Ferrihydrite is an important scavenger of both anionic complexes and radionuclides; thus, precipitation of ferrihydrite in the groundwater could exert control on the ^{59}Fe concentration as well as on other radionuclides such as antimony, cobalt, zinc, and zirconium (Jenne, 1968; Jenne and Wahlberg, 1968; Onishi et al., 1981). Using the analytically determined concentrations for total iron and assuming redox equilibria with the Eh measured using the platinum electrode, ferrihydrite was calculated to be oversaturated for several of the 1983 samples (Appendix Table C.17). Ferrihydrite precipitates very rapidly from waters high in iron concentration (Nordstrom et al., 1979); therefore, supersaturation with respect to this phase would not be expected. These results suggest that the iron determinations are in error. As with aluminum, overestimation of the concentration of iron in solution is a common analytical problem unless a $<0.1 \mu\text{m}$ -membrane is used to filter the water sample (Kennedy et al., 1974). If the pH and Eh of the spring and groundwater samples are assumed to be accurate, the total iron in solution is calculated to be in the range 2.0 to 5.0×10^{-4} mg/L assuming equilibrium with ferrihydrite. Only one set of samples (i.e., January 1983) has data on iron concentrations for the trench and groundwater. The calculated saturation indices suggest that the ferrihydrite is precipitating; however, given the sparsity of the data and the analytical errors in iron determination, this cannot be stated with certainty.

Several other solid phases are potentially solubility controls in these groundwaters and could exert controls on the concentrations of radionuclides in the groundwater; however, the analytical data are incomplete

with the concentrations of several of the constituents reported as less than some detection limit. These solid phases include otavite for control of cadmium concentrations, cerussite for control of lead, chloropyromorphite for lead and phosphate, $ZnSiO_3$ for zinc, $MnHPO_4$ for manganese and phosphate, wulfenite for lead and molybdenum, and CeO_2 for cerium. These solid phases have been identified as potential solubility controls in several groundwater systems (Jenne et al., 1980; Lindsay, 1979) and therefore could be active in this system also. The detection limits for many of the constituents in the waters need to be increased to at least 10^{-4} mg/L in order to be able to address the potential existence of these solubility controls.

8.4 Conclusions

- The mobile species of radionuclides are primarily anionic and nonionic. Most of the mobile species are oxyhydroxy complexes, although organic complexes may be important mobile species for iron, zinc, and cobalt.
- Those radionuclides that occur in most waters as the uncomplexed cation such as cesium, cerium, and manganese, appear to be the most greatly retarded.
- The ground waters appear to be in equilibrium with several solid phases. This can be important in controlling the concentrations of trace elements in solution and can act to remove certain radionuclides from solution by co-precipitation and sorption. These solid phases include calcite, several aluminosilicates, and ferrihydrite.
- Calcite precipitation could be an important mechanism in radionuclide retardation and this possibility needs to be evaluated.

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APPENDIX A

ANALYTICAL CHARGE-FORM SPECIATION FOR
RADIONUCLIDES

TRENCH - MANWAY #1 (1/26/83 @ 1400)

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	109.900	11.8	87.6	0.6	0.002
⁵⁹ Fe	57.030	96.7	2.3	1.0	0.02
⁵⁸ Co	3.670	23.0	76.3	0.5	<0.02
⁶⁰ Co	49.330	58.5	41.3	0.1	0.03
⁹⁵ Zr	11.230	90.6	3.0	6.0	0.4
⁹⁵ Nb	21.580	58.2	33.7	7.9	0.06
⁹⁹ Mo	121.200	1.2	1.1	97.7	<0.01
^{99m} Tc	17.930	<0.2	0.8	99.2	<0.01
¹⁰³ Ru	4.140	74.0	12.2	12.6	1.2
¹⁰⁶ Ru	1.440	>65	<14	<23	<0.4
¹²⁴ Sb	1,355	60.9	0.8	34.7	3.6
¹²⁵ Sb	344	<54	ND	98.0	2.0
¹⁴⁰ Ba	38.990	14.2	85.8	<0.1	<0.1
¹⁴⁰ La	113.880	63.6	36.3	0.1	<0.1
¹³⁷ I	31.980	2.7	2.9	94.0	0.5
¹³³ I	459.200	4.1	<0.05	95.5	0.4
¹³⁷ Cs	16.100	0.1	99.9	<0.1	<0.1
¹⁴⁴ Ce	7.010	~100	<4	<1	<0.06
¹⁵⁴ Eu	670	~100	<3	<40	<0.1

TRENCH - MANWAY #1 (2/15/83 @ 1145)

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	271.830	86.5	12.6	0.8	<0.007
⁵⁹ Fe	496.700	98.9	0.7	0.3	<0.005
⁵⁸ Co	46.800	97.9	2.0	<0.07	<0.02
⁶⁰ Co	73.000	98.1	1.7	0.2	0.07
⁹⁵ Zr	7.220	99.7	0.3	<1	<0.04
⁹⁵ Nb	35.700	90.0	4.3	5.4	0.2
⁹⁹ Mo	102.500	88.2	3.9	7.8	0.1
^{99m} Tc	28.950*	-	<1*	100*	<0.01
¹⁰³ Ru	3.150*	-	<0.4*	100*	<0.03*
¹⁰⁶ Ru	13.830	88.9	5.6	5.2	0.3
¹²⁴ Sb	3.490	>85	<12	<7	0.5
¹²⁵ Sb	10.000	93.0	0.7	5.7	0.8
¹³¹ I	<280	-	-	-	-
¹³³ I	9.380	-	4.9*	88.6*	0.2*
¹³⁷ Cs	13.470*	-	<0.9*	99.4*	0.7*
¹⁴⁰ Ba	23.000	4.9	96.1	<0.1	<0.01
¹⁴⁰ La	14.450*	-	98.6*	1.3	<0.08
¹⁴¹ Ce	12.680*	-	96.3	3.7	<0.01
¹⁴⁴ Ce	11,100	87.8	12.5	<0.5	<0.02
¹⁴⁴ Ce	11,000	85.9	14.2	<2	<0.05
¹⁵⁴ Eu	590*	-	71*	29*	<3

ND = NON-DETECTABLE
* = SOLUBLE FRACTION ONLY

TRENCH, 1ST MANWAY, MARCH 30, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	244.300	54.8	44.8	0.2	0.004
⁵⁹ Fe	519.500	99.1	0.6	0.3	0.01
⁵⁸ Co	80.210	94.9	5.1	0.02	0.01
⁶⁰ Co	567.100	57.2	2.6	0.1	0.02
⁹⁵ Zr	10.020	98.7	1.3	<0.2	0.003
⁹⁵ Nb	35.960	92.6	1.9	5.4	0.06
⁹⁹ Mo	70.940	73.2	22.7	3.9	0.09
^{99m} Tc*	13.400	-	3.5	96.4	0.1
¹⁰³ Ru	2.030	-	<0.6	100	<0.04
¹⁰⁶ Ru	18.200	82.9	8.3	7.8	0.9
¹⁰⁶ Ru	3.850	73.2	24.8	<2	2.0
¹²⁴ Sb	8.970	96.9	<0.4	3.1	<0.02
¹²⁵ Sb	223	<52	<14	>33	0.2
¹³¹ I	8.580	1.2	3.1	95.6	0.09
¹³⁷ Cs	62.070	7.7	92.3	<0.31	<0.005
¹⁴⁰ Ba*	43.900	-	95.9	<0.02	0.02
¹⁴⁰ La*	39.500	-	93.0	1.7	0.3
¹⁴¹ Ce	11,950	85.8	12.6	<0.1	0.5
¹⁴⁴ Ce	11,260	90.4	<0.7	0.4	<0.7
¹⁵⁴ Eu*	245	-	>95	<20	3.3

* = SOLUBLE FRACTION ONLY

FIRST MANWAY, JUNE 16, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	86.200	5.8	94.1	0.078	<0.002
⁵⁹ Fe	11.700	93.9	3.2	2.8	0.071
⁵⁸ Co	354	99.1	<8	0.90	<0.001
⁶⁰ Co	32.700	41.0	58.5	0.45	0.021
⁹⁵ Zr	475	56.2	43.7	0.11	0.002
⁹⁵ Nb	13.500	77.3	6.4	16.1	0.20
⁹⁹ Mo	35.200	51.2	35.5	13.1	0.15
^{99m} Tc	-	-	-	-	-
¹⁰³ Ru	7.430	65.1	13.4	20.0	1.5
¹⁰⁶ Ru	2.890	59.9	18.5	20.2	1.4
¹²⁴ Sb	256	82.4	<6	17.6	<0.2
¹²⁵ Sb	182	-	<33	87.9	<0.1
¹³¹ I	1.010	4.7	28.9	66.3	0.34
¹³⁷ Cs	27.500	10.2	95.8	0.007	<0.005
¹⁴⁰ Ba	7.730	9.0	91.0	<0.6	<0.05
¹⁴⁰ La	2.0E 10 ⁷	6.3	93.7	<0.1	<0.01
¹⁴¹ Ce	7.780	77.0	14.6	7.9	0.46
¹⁴⁴ Ce	10.400	78.0	13.7	7.8	0.47
¹⁵⁴ Eu	292	16.4	73.4	9.8	0.39

SECOND MANWAY, MAY 5, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵¹ Cr	3.170	93.6	5.8	0.38	0.60
⁵⁴ Mn	133.100	34.4	65.2	0.33	0.010
⁵⁹ Fe	37.900	99.3	0.3	0.33	0.006
⁵⁸ Co	4.410	33.4	66.2	0.24	0.06
⁶⁰ Co	115.400	86.4	13.1	0.45	0.05
⁶⁵ Zn	1.150	99.3	0.47	<0.01	0.26
⁹⁰ Zr	15.700	92.2	1.8	5.9	0.04
⁹⁵ Nb	35.300	94.4	2.7	2.9	0.02
⁹⁹ Mo	5.680	43.1	<35	36.8	<1
^{99m} Tc	453	<18	<19	99.6	0.3
¹⁰³ Ru	8.960	68.2	15.5	14.9	1.4
¹⁰⁶ Ru	3.490	66.3	21.4	11.2	1.1
¹²⁴ Sb	1.730	52.1	1.8	47.7	0.16
¹²⁵ Sb	493	<15	<20	~100	0.006
¹³¹ I	5.620	9.9	5.6	84.4	0.08
¹³⁷ Cs	57.500	7.3	92.7	<0.009	<0.004
¹⁴⁰ Ba	24.600	26.5	73.3	0.17	0.05
¹⁴⁰ La	815.000	18.8	81.0	0.19	0.002
¹⁴¹ Ce	6.260	96.2	3.7	<0.1	0.06
¹⁴⁴ Ce	11.380	97.8	2.2	<0.2	0.07
¹⁵⁴ Eu	<500	-	-	-	-

SECOND MANWAY, SEPTEMBER 9, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	122.200	6.9	93.1	0.05	0.008
⁵⁹ Fe	62.900	91.1	5.9	2.7	0.23
⁵⁸ Co	2.830	24.2	75.7	<0.1	0.1
⁶⁰ Co	29.100	63.1	36.4	0.33	0.16
⁶⁵ Zn	539	61.6	38.4	<0.9	<0.09
⁹⁰ Zr	7.110	80.3	10.2	9.3	0.19
⁹⁵ Nb	15.570	44.0	49.2	6.5	0.20
⁹⁹ Mo	1.110	<20	<40	~100	<3
^{99m} Tc	117	<7	<17	~100	<2
¹⁰³ Ru	3.010	60.8	16.0	20.1	3.1
¹⁰⁶ Ru	2.050	57.9	19.9	19.9	2.3
¹²⁴ Sb	525	80.4	<6	19.4	0.1
¹²⁵ Sb	172	<14	<48	~100	1.0
¹³¹ I	4.910	2.8	1.2	95.9	0.07
¹³⁷ Cs	59.950	9.9	90.1	<0.005	<0.0005
¹⁴⁰ Ba	25.700	5.5	94.6	<0.07	<0.008
¹⁴⁰ La	111.900	11.0	88.5	0.22	0.26
¹⁴¹ Ce	4.050	79.6	19.4	<0.1	0.9
¹⁴⁴ Ce	7.920	84.9	14.2	<0.2	0.9
¹⁵⁴ Eu	<270	-	-	-	-

TRENCH - LAST MANWAY (2/15/83 @ 1520)

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	137.500	41.1	58.3	0.6	<0.007
⁵⁹ Fe	250.600	96.6	1.7	1.6	<0.03
⁵⁸ Co	7.080	48.2	51.7	<0.2	<0.05
⁶⁵ Zn	3.390	96.7	3.2	<0.8	<0.008
⁶⁰ Co	214.200	92.0	7.1	0.73	<0.09
⁹⁵ Zr	7.780	39.8	21.2	35.7	3.2
⁹⁵ Nb	24.560	25.0	60.6	14.2	0.2
⁹⁹ Mo	18.900	6.5	0.8	92.7	<0.03
^{99m} Tc	1.970	18.0	<0.6	81.9	<0.04
¹⁰³ Ru	7.660	65.3	21.5	12.4	0.7
¹⁰⁶ Ru	630	100	ND	ND	ND
¹²⁴ Sb	1.760	54.7	2.8	37.6	4.8
¹²⁵ Sb	910	68.7	<8	26.4	4.8
¹³¹ I	7.930	0.8	3.7	95.4	<0.08
¹³² I	6.660	0.5	<2	99.0	0.5
¹³⁷ Cs	26.640	5.8	94.1	<0.05	<0.008
¹⁴⁰ Ba	31.560	1.0	98.6	0.04	<0.03
¹⁴⁰ La	54.590	11.2	87.5	1.3	<0.08
¹⁴¹ Ce	5.450	57.3	42.7	0.5	0.6
¹⁵⁴ Eu	1.990	94.4	0.9	5.1	<0.5

ND = NON-DETECTABLE

TRENCH, LAST MANWAY, MARCH 30, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	125.860	10.1	89.4	0.4	0.05
⁵⁹ Fe	72.250	97.1	0.76	2.1	0.05
⁵⁸ Co	4.737	<6	99.6	0.4	0.06
⁶⁰ Co	43.230	84.2	13.0	2.5	0.3
⁶⁵ Zn	—	—	—	—	—
⁹⁵ Zr	2.290	<20	4.5	94.7	0.7
⁹⁵ Nb	6.650	<3	3.3	94.3	2.3
⁹⁹ Mo	17.230	18.6	0.4	81.0	0.03
^{99m} Tc	3.310	11.7	0.4	87.8	<0.01
¹⁰³ Ru	3.010	32.6	14.4	49.0	3.9
¹⁰⁶ Ru	—	—	—	—	—
¹²⁴ Sb	585*	—	5.2	94.6	0.09
¹²⁵ Sb	380*	—	<23	99.1	0.9
¹³¹ I	11.110	0.8	0.5	98.6	0.07
¹³⁷ Cs	70.690	0.2	99.8	<0.01	<0.003
¹⁴⁰ Ba	55.690	1.0	98.7	0.2	<0.01
¹⁴⁰ La	71.570	0.3	98.6	0.9	0.1
¹⁴¹ Ce	2.010	43.8	54.2	<1	2.0
¹⁴⁴ Ce	1.250	<20	97.4	<7	2.4
¹⁵⁴ Eu	732	98.1	<20	<8	1.8

* = SOLUBLE FRACTION ONLY

LAST MANWAY, MAY 5, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵¹ Cr	1.050	71.3	26.0	2.8	<1
⁵⁴ Mn	153.100	16.8	82.8	0.44	0.02
⁵⁹ Fe	16.280	96.8	1.1	2.1	0.04
⁵⁸ Co	2.350	26.5	73.2	0.25	0.03
⁶⁰ Co	60.630	54.3	44.4	1.2	0.07
⁶⁵ Zn	371	96.7	<23	2.9	1.3
⁹⁵ Zr	14.640	88.8	4.3	6.9	0.01
⁹⁵ Nb	45.900	94.2	3.2	2.6	0.05
⁹⁹ Mo*	1.760	—	43.4	55.5	0.80
^{99m} Tc	—	—	—	—	—
¹⁰³ Ru	8.850	67.0	13.9	17.8	1.3
¹⁰⁶ Ru	5.210	65.3	22.3	11.4	1.0
¹²⁴ Sb	993	18.6	<4	79.8	1.6
¹²⁵ Sb	718	3.3	<18	95.5	1.1
¹³¹ I	3.990	13.0	6.4	80.5	0.07
¹³⁷ Cs	91.900	5.6	94.4	<0.002	<0.002
¹⁴⁰ Ba*	17.030	29.3	69.9	0.8	<0.05
¹⁴⁰ La	339.700	—	99.8	0.2	<0.02
¹⁴¹ Ce	7.061	95.1	2.9	1.8	0.14
¹⁴⁴ Ce	10.980	98.7	0.81	0.39	0.14
¹⁵⁴ Eu	197	<50	64	<36	<5

* = SOLUBLE FRACTION ONLY

LAST MANWAY, JUNE 16, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	98.800	2.8	97.1	0.07	0.002
⁵⁹ Fe	7.480	91.8	3.3	4.7	0.21
⁵⁸ Co	246	75.3	22.5	2.3	0.08
⁶⁰ Co	32.300	74.3	75.2	0.46	0.02
⁶⁵ Zn	364	45.5	52.5	2.1	0.01
⁹⁵ Zr	8.550	71.5	8.2	21.9	0.32
⁹⁵ Nb	24.200	37.3	48.0	14.5	0.23
⁹⁹ Mo	—	—	—	—	—
^{99m} Tc	—	—	—	—	—
¹⁰³ Ru	4.340	48.6	16.3	32.4	2.7
¹⁰⁶ Ru	1.620	56.3	4.8	35.9	3.0
¹²⁴ Sb	169	80.4	<25	19.5	<0.2
¹²⁵ Sb	168	—	—	~100	—
¹³¹ I	740	—	—	~100	—
¹³⁷ Cs	35.100	14.3	85.7	—	—
¹⁴⁰ Ba	8.210	3.5	96.4	0.07	0.01
¹⁴⁰ La	943.000	10.4	87.7	0.22	1.7
¹⁴¹ Ce	5.380	74.1	16.7	8.1	1.2
¹⁴⁴ Ce	7.540	77.0	13.8	8.0	1.2
¹⁵⁴ Eu	<360	—	—	—	—

WELL #1 (1/27/83 @ 1430)

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	90.6	0.08	0.4	99.4	0.1
⁵⁹ Fe	21.6	0.3	16.0	82.2	<1
⁶⁰ Co	12.2	<0.2	8.2	90.2	1.1
⁶⁰ Co	210	0.4	64.8	33.7	1.1
⁹⁰ Zr	19.9	0.09	8.0	90.4	<0.5
⁹⁵ Nb	118	0.08	1.9	97.5	0.3
⁹⁹ Mo	13,590	0.2	0.7	99.0	<0.01
^{99m} Tc	2,400	0.2	<0.8	99.0	<0.01
¹⁰³ Ru	300	0.3	11.7	86.7	1.2
¹⁰⁶ Ru	324	<0.2	10.9	87.6	1.3
¹²⁴ Sb	233	<0.2	0.5	91.0	9.0
¹²⁵ Sb	102	0.2	1.0	83.2	15.6
¹⁴⁰ Ba	40	1.5	<0.1	98.5	<0.1
¹⁴⁰ La	<6	-	-	-	-
¹³¹ I	11,120	0.4	4.9	94.7	<0.1
¹³³ I	21,740	0.5	<0.3	99.2	0.2
¹³⁷ Cs	<1	-	-	-	-
¹⁴⁴ Ce	<60	-	-	-	-
¹⁵⁴ Eu	<50	-	-	-	-

WELL #1(2/15/83 @ 1430)

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	7.88	37.6	60.7	1.8	<5
⁵⁹ Fe	19.5	80.0	17.4	<9	<6
⁶⁰ Co	<2	-	-	-	-
⁶⁰ Co	160	7.8	32.5	57.9	1.4
⁶⁵ Zn	2.7	<11	58.5	40.0	<19
⁹⁰ Zr	1.4	24.3	ND	37.9	<39
⁹⁵ Nb	7.4	9.1	22.2	53.0	4.1
⁹⁹ Mo	1,570	0.5	5.4	94.1	<0.2
^{99m} Tc	127	0.3	5.0	94.5	<0.2
¹⁰³ Ru	316	1.1	21.5	74.7	2.6
¹⁰⁶ Ru	114	1.5	21.0	71.9	6.3
¹²⁴ Sb	203	<0.3	2.1	96.1	1.9
¹²⁵ Sb	188	<0.2	0.5	94.7	5.2
¹³¹ I	1,320	0.8	4.5	94.6	0.08
¹³³ I	129	0.4	12.4	83.7	3.3
¹³⁷ Cs	1.8	27	49	10	15
¹⁴⁰ Ba	<7	-	-	-	-
¹⁴⁰ La	3.9	<15	61	<40	38
¹⁴¹ Ce	<3	-	-	-	-
¹⁵⁴ Eu	5.7	53	ND	48	<14

ND = NON-DETECTABLE

WELL #1, MAY 6, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	19.9	21.0	77.8	1.6	<0.2
⁵⁹ Fe	9.4	91.2	<5	8.8	<1
⁶⁰ Co	1.8	51.5	34.8	13.8	<2
⁶⁰ Co	292	5.6	60.7	32.7	0.96
⁶⁵ Zn	1.1	30	70	<47	<9
⁹⁰ Zr	1.4	55	<1	45	<4
⁹⁵ Nb	3.5	48	34	18	<0.03
⁹⁹ Mo	770	0.3	<25	99.7	<16
^{99m} Tc	166	0.2	<5	99.8	<2
¹⁰³ Ru	310	2.3	34.8	59.7	3.2
¹⁰⁶ Ru	200	3.1	30.4	58.1	3.4
¹²⁴ Sb	72.8	0.2	<0.4	98.1	1.7
¹²⁵ Sb	170	0.2	<0.8	97.4	1.7
¹³¹ I	1,440	3.2	5.1	91.5	0.1
¹³⁷ Cs	3.9	21.6	78.2	<5	<7
¹⁴⁰ Ba	<4	-	-	-	-
¹⁴⁰ La	426	<9	~100	<2	-
¹⁴¹ Ce	<0.6	-	-	-	-
¹⁴⁴ Ce	<2	-	-	-	-
¹⁵⁴ Eu	<2	-	-	-	-

WELL #1, MARCH 31, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	94.1	22.7	83.9	13.8	<0.1
⁵⁹ Fe	98.4	81.7	2.2	16.0	<0.003
⁵⁸ Co	6.6	86.7	13	<22	<1
⁶⁰ Co	381	19.6	29.1	50.1	1.1
⁶⁵ Zn	1.2	~100	-	-	-
⁹⁵ Zr	21.5	22.8	36.7	40.4	<0.9
⁹⁵ Nb	77.0	11.6	63.2	23.9	<0.08
⁹⁹ Mo	3,300	0.2	1.8	98.0	<0.06
^{99m} Tc	1,150	0.05	0.9	99.1	<0.007
¹⁰³ Ru	1,400	0.2	4.0	95.3	0.4
¹⁰⁶ Ru	395	0.8	9.9	88.6	0.7
¹²⁴ Sb	129	1.0	<0.9	84.3	14.7
¹²⁵ Sb	180	0.1	<0.6	82.2	18.1
¹³¹ I	6,865	0.6	2.4	97.0	0.04
¹³⁷ Cs	0.97	16	84	<50	<5
¹⁴⁰ Ba	7.1	11	89	<70	<5
¹⁴⁰ La	46	3.7	96.3	<4	<1
¹⁴¹ Ce	<20	-	-	-	-
¹⁴⁴ Ce	<2	-	-	-	-
¹⁵⁴ Eu	<10	-	-	-	-

WELL #1, SEPTEMBER 9, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	2.4	3.6	48.2	48.2	<2
⁵⁹ Fe	0.60	45	55	-	<17
⁵⁸ Co	0.27	<11	~100	-	<11
⁶⁰ Co	89.6	4.5	58.8	34.9	1.9
⁶⁵ Zn	<1	-	-	-	-
⁹⁵ Zr	3.5	3.4	<28	96.6	<2
⁹⁵ Nb	4.3	4.9	25.7	69.3	0.9
⁹⁹ Mo	2,810	0.06	<2	99.9	<0.07
^{99m} Tc	320	<0.02	<2	~100	<0.04
¹⁰³ Ru	271	1.1	10.7	74.8	1.2
¹⁰⁶ Ru	190	1.7	16.9	79.9	1.6
¹²⁴ Sb	210	<0.04	0.16	85.9	13.9
¹²⁵ Sb	233	0.1	0.68	84.6	14.7
¹³¹ I	25	<10	<10	~100	-
¹³⁷ Cs	<1	-	-	-	-
¹⁴⁰ Ba	<4	-	-	-	-
¹⁴⁰ La	350	<0.1	~100	<0.6	<0.6
¹⁴¹ Ce	<4	-	-	-	-
¹⁴⁴ Ce	<9	-	-	-	-
¹⁵⁴ Eu	<2	-	-	-	-

WELL #3 (2/17/83 @ 1030)

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	1.6	~100	ND	ND	ND
⁵⁹ Fe	3.5	~100	ND	ND	ND
⁶⁰ Co	1.8	<2	57	40	<11
⁶⁰ Co	125	3.0	50	43	3.1
⁶⁵ Zn	3.6	ND	100	ND	ND
⁹⁵ Zr	1.0	33	ND	68	ND
⁹⁵ Nb	<1	-	-	-	-
⁹⁹ Mo	<7	-	-	-	-
^{99m} Tc	<0.5	-	-	-	-
¹⁰³ Ru	13.6	0.9	15	82	3
¹⁰⁶ Ru	66.2	1.8	21.6	76.6	<3
¹²⁴ Sb	<0.5	-	-	-	-
¹²⁵ Sb	21.9	<1	<5	76	24
¹³¹ I	7.2	<1	4	94	<2
¹³² I	<2	-	-	-	-
¹³⁷ Cs	<1	-	-	-	-
¹⁴⁰ Ba	<3	-	-	-	-
¹⁴⁰ La	<2	-	-	-	-
¹⁴¹ Ce	<0.6	-	-	-	-
¹⁵⁴ Eu	<2	-	-	-	-

ND = NON-DETECTABLE

WELL #3 (1/26/83 @ 1300)

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	<1	-	-	-	-
⁵⁹ Fe	<1	-	-	-	-
⁶⁰ Co	0.74	1.0	29.7	58.0	12.3
⁶⁰ Co	165	0.5	63.8	34.2	1.3
⁹⁵ Zr	<1	-	-	-	-
⁹⁵ Nb	<1	-	-	-	-
⁹⁹ Mo	<5	-	-	-	-
^{99m} Tc	<0.2	-	-	-	-
¹⁰³ Ru	14.5	<0.1	8.4	91.1	1.4
¹⁰⁶ Ru	82.6	0.9	9.6	88.3	1.3
¹²⁴ Sb	<1	-	-	-	-
¹²⁵ Sb	22.5	0.4	1.7	96.0	1.8
¹⁴⁰ Ba	<1	-	-	-	-
¹⁴⁰ La	<1	-	-	-	-
¹³¹ I	5.0	<1	<2	>97	<1
¹³² I	<1	-	-	-	-
¹³⁷ Cs	<1	-	-	-	-
¹⁴⁴ Ce	<1	-	-	-	-
¹⁵⁴ Eu	<5	-	-	-	-

SPRINGS (1/31/83 @ 0900)

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	<0.1	-	-	-	-
⁵⁹ Fe	<0.2	-	-	-	-
⁵⁸ Co	0.28	<0.3	5.0	92.9	1.7
⁶⁰ Co	62.2	0.2	44.9	50.9	4.0
⁹⁵ Zr	<0.1	-	-	-	-
⁹⁵ Nb	<0.1	-	-	-	-
⁹⁹ Mo	48	<0.2	<12	>88	<0.3
^{99m} Tc	12	<0.1	<1	>99	<0.3
¹⁰³ Ru	39.2	0.1	13.0	83.2	3.6
¹⁰⁶ Ru	45.7	0.3	15.1	80.5	3.9
¹²⁴ Sb	6.2	<0.2	<1.6	26.0	74.2
¹²⁵ Sb	88.0	<0.05	0.6	23.3	76.1
¹⁴⁰ Ba	<0.1	-	-	-	-
¹⁴⁰ La	<0.5	-	-	-	-
¹³⁷ I	182	0.4	5.6	92.3	1.5
¹³⁷ I	7.2	<2	-	100	<8
¹³⁷ Cs	<0.1	-	-	-	-
¹⁴⁴ Ce	<0.4	-	-	-	-
¹⁵⁴ Eu	<0.3	-	-	-	-

SPRINGS (2/18/83 @ 1330)

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	<0.07	-	-	-	-
⁵⁹ Fe	<0.2	-	-	-	-
⁵⁸ Co	0.35	<0.1	10	89	<2
⁶⁰ Co	43	<0.07	26	67	7
⁹⁵ Zr	<0.2	-	-	-	-
⁹⁵ Zr	<0.1	-	-	-	-
⁹⁵ Nb	<0.1	-	-	-	-
⁹⁹ Mo	29.6	ND	ND	~100	ND
^{99m} Tc	3.6	ND	ND	~100	ND
¹⁰³ Ru	33	<0.03	3	92	5
¹⁰⁶ Ru	32	<0.2	4	88	7
¹²⁴ Sb	5.0	ND	ND	ND	100
¹²⁵ Sb	63	<0.00	2	<0.2	98
¹³⁷ I	341	<0.02	<0.7	91	8
¹³⁷ I	0.23	ND	ND	~100	ND
¹³⁷ Cs	<0.03	-	-	-	-
¹⁴⁰ Ba	<0.3	-	-	-	-
¹⁴⁰ La	<0.03	-	-	-	-
¹⁴¹ Ce	<0.1	-	-	-	-
¹⁵⁴ Eu	<0.3	-	-	-	-

ND = NON-DETECTABLE

SPRINGS, APRIL 1, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	<0.1	-	-	-	-
⁵⁹ Fe	<0.2	-	-	-	-
⁵⁸ Co	<0.2	-	-	-	-
⁶⁰ Co	58.0	0.3	40.6	51.9	7.2
⁹⁵ Zr	<0.3	-	-	-	-
⁹⁵ Zr	<0.2	-	-	-	-
⁹⁵ Nb	<0.2	-	-	-	-
⁹⁹ Mo	<3	-	-	-	-
^{99m} Tc	0.4	-	-	~100	-
¹⁰³ Ru	21.5	0.2	13.0	79.9	7.0
¹⁰⁶ Ru	33.8	0.3	14.7	78.7	6.2
¹²⁴ Sb	2.9	<0.3	<2	5.4	94.5
¹²⁵ Sb	74.0	<0.08	<0.02	17.0	83.0
¹³⁷ I	133	0.1	1.5	94.6	3.8
¹³⁷ Cs	<0.1	-	-	-	-
¹⁴⁰ Ba	<1	-	-	-	-
¹⁴⁰ La	<0.4	-	-	-	-
¹⁴¹ Ce	<0.1	-	-	-	-
¹⁴⁴ Ce	<0.6	-	-	-	-
¹⁵⁴ Eu	<0.3	-	-	-	-

SPRINGS, MAY 9, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	0.050	~100	-	-	-
⁵⁹ Fe	0.15	40	<50	80	<2
⁵⁸ Co	0.50	<0.8	35	86	<2
⁶⁰ Co	81.1	0.2	40.6	55.0	4.2
⁸⁵ Zn	<0.2	-	-	-	-
⁹⁵ Zr	0.06	<5	<18	~100	<2
⁹⁵ Nb	0.12	<3	<8	~100	<4
⁹⁹ Mo	18	<0.8	<2	~100	<1
^{99m} Tc	4	<0.1	<0.5	~100	<0.5
¹⁰³ Ru	73.2	0.07	13.9	80.0	3.3
¹⁰⁶ Ru	82.0	0.05	14.4	82.2	3.4
¹²⁴ Sb	4.3	<0.08	<0.5	3.9	60.5
¹²⁵ Sb	68.5	0.05	0.5	35.9	64.2
¹³¹ I	337	0.09	2.6	96.3	0.8
¹³⁷ Cs	0.028	21	79	-	-
¹⁴⁰ Ba	<0.3	-	-	-	-
¹⁴⁰ La	<0.8	-	-	-	-
¹⁴¹ Ce	<0.05	-	-	-	-
¹⁴⁴ Ce	<0.5	-	-	-	-
¹⁵⁴ Eu	<0.3	-	-	-	-

SPRINGS, JUNE 20, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	<0.07	-	-	-	-
⁵⁹ Fe	<0.2	-	-	-	-
⁵⁸ Co	1.91	<0.1	49.1	43.3	7.3
⁶⁰ Co	152	0.03	45.7	48.3	6.3
⁸⁵ Zn	<0.7	-	-	-	-
⁹⁵ Zr	<0.2	-	-	-	-
⁹⁵ Nb	0.26	-	-	~100	-
⁹⁹ Mo	-	-	-	-	-
^{99m} Tc	-	-	-	-	-
¹⁰³ Ru	132	0.02	10.4	86.0	3.6
¹⁰⁶ Ru	127	0.05	12.6	82.0	4.6
¹²⁴ Sb	5.2	<0.1	<1	19.8	79.8
¹²⁵ Sb	90.4	0.02	<0.4	21.5	78.9
¹³¹ I	32	-	-	~100	-
¹³⁷ Cs	<0.2	-	-	-	-
¹⁴⁰ Ba	<0.3	-	-	-	-
¹⁴⁰ La	55	-	~100	-	-
¹⁴¹ Ce	<0.05	-	-	-	-
¹⁴⁴ Ce	<0.3	-	-	-	-
¹⁵⁴ Eu	<0.05	-	-	-	-

SPRINGS, SEPTEMBER 12, 1983

RADIONUCLIDE	TOTAL pCi/l	% PARTICULATE	% CATIONIC	% ANIONIC	% NONIONIC
⁵⁴ Mn	<0.1	-	-	-	-
⁵⁹ Fe	<0.3	-	-	-	-
⁵⁸ Co	0.44	<0.4	28.2	72.6	<5
⁶⁰ Co	79.3	0.2	48.7	47.6	3.6
⁸⁵ Zn	<0.4	-	-	-	-
⁹⁵ Zr	0.015	~100	-	-	-
⁹⁵ Nb	0.022	~100	-	-	-
⁹⁹ Mo	74.9	<0.2	<2	~100	<1
^{99m} Tc	7.9	<0.1	<1	~100	<2
¹⁰³ Ru	60.3	0.09	9.9	87.5	2.5
¹⁰⁶ Ru	89.9	0.13	12.0	85.0	2.7
¹²⁴ Sb	8.2	<0.1	<1	40.4	59.7
¹²⁵ Sb	140	0.09	0.3	38.6	61.1
¹³¹ I	707	0.03	0.9	99.1	<0.02
¹³⁷ Cs	<0.1	-	-	-	-
¹⁴⁰ Ba	<0.5	-	-	-	-
¹⁴⁰ La	<0.7	-	-	-	-
¹⁴¹ Ce	<0.2	-	-	-	-
¹⁴⁴ Ce	<0.5	-	-	-	-
¹⁵⁴ Eu	<1	-	-	-	-

APPENDIX B

CHEMICAL COMPOSITION OF TRENCH WELL AND
SPRING WATERS FOR 1983

CHEMICAL COMPOSITION OF TRENCH, WELL AND SPRING WATERS,
JANUARY, 1983

CONSTITUENT	TRENCH 1/26/83	WELL #1 1/27/83	WELL #3 1/26/83	SPRING 1/28/83	SPRING 1/31/83
TEMPERATURE °C	21	18	18	20	20
pH	7.30	8.10	8.15	8.10	8.15
Eh (mv)	+405	+430	+355	+435	+385
DISSOLVED O ₂ (mg/l)	6.9	7.2	9.2	4.9	4.5
ALKALINITY (As CaCO ₃ mg/l)	38.0	54.9	52.8	57.5	57.7
DOC (mg/l)	<1	<1	<1	<1	<1
Ca (mg/l)	16.3	25.2	25.3	26.0	20.1
Mg (mg/l)	4.0	4.2	4.2	4.3	4.8
Na (mg/l)	1.68	1.70	1.70	1.99	2.17
K (mg/l)	0.40	1.0	1.0	0.50	1.8
Sr (mg/l)	0.081	0.093	0.093	0.094	0.12
SO ₄ ⁻² (mg/l)	14.3	13.9	12.2	13.5	13.9
S ⁻ (POLAROGRAPHIC) (mg/l)	<0.003	<0.003	<0.003	<0.003	<0.003
SO ₄ ⁻² (TOTAL BY ICP) (mg/l)	5.6	5.1	5.1	5.4	4.6
NO ₃ ⁻ (mg/l)	9.2	13.2	8.2	12.5	13.2
NO ₂ ⁻ (mg/l)	2.5	<0.05	<0.05	<0.05	<0.05
F ⁻ (mg/l)	<0.15	<0.15	<0.15	<0.15	<0.15
PO ₄ ⁻³ (mg/l)	3.8	<0.2	<0.2	<0.2	<0.2
P (TOTAL BY ICP) (mg/l)	1.48	<0.1	<0.1	<0.1	<0.1
Cl ⁻ (mg/l)	0.90	0.87	0.70	0.83	0.87
Si (mg/l)	1.8	4.4	4.4	2.9	8.2
Al (µg/l)	<50	<50	<50	<50	<50
As (µg/l)	<30	<30	<30	<30	<30
B (µg/l)	10	10	10	10	10
Ba (µg/l)	34	28	26	16	14
Co (µg/l)	<10	<10	<10	<10	<10
Cr (µg/l)	<10	<10	<10	<10	<10
Fe (TOTAL BY ICP) (µg/l)	110	<20	<20	<20	<20
Fe ⁺² (COLORIMETRIC) (µg/l)	6.1	<2*	6.1*	3.6	<2
Fe ⁺³ (COLORIMETRIC) (µg/l)	84.9	<2*	5.9*	2.5	<2
Fe ⁺² /Fe ⁺³	0.067	-	1.03	1.44	-
Li (µg/l)	<5	<5	<5	<5	<5
Mn (µg/l)	<10	<10	<10	<10	<10
Mo (µg/l)	<10	<10	<10	<10	<10
Ni (µg/l)	<20	<20	<20	<20	<20
Se (µg/l)	<100	<100	<100	<100	<100
Ti (µg/l)	<10	<10	<10	<10	<10
V (µg/l)	<5	<5	<5	<5	<5
Zn (µg/l)	24	<10	<10	<10	<10

*WATER PUMPED THROUGH STAINLESS STEEL SAMPLING DEVICE - PROBABLE CONTAMINATION WITH IRON

CHEMICAL COMPOSITION OF TRENCH, WELL AND SPRING WATERS,
FEBRUARY, 1983

CONSTITUENT	TRENCH (1st MANWAY) 2/15/83	TRENCH (LAST MANWAY) 2/15/83	WELL #1 2/15/83	WELL #3 2/17/83	SPRINGS 2/17/83
TEMPERATURE °C	17	15	15	19	21
pH	9.20	9.25	8.15	8.40	8.10
Eh (mv)	+155	+150		+350	+340
DISSOLVED O ₂ (mg/l)	6.58	5.91	5.57	6.64	5.14
ALKALINITY (As CaCO ₃ mg/l)	63.3	63.0	65.5	53.7	57.1
DOC (mg/l)	0.4	0.7	0.4	2.3	0.3
Ca (mg/l)	15.5	16	23	21	26
Mg (mg/l)	3.9	4.0	4.0	4.9	4.4
Na (mg/l)	1.63	1.72	2.11	2.23	1.82
K (mg/l)	0.4	0.4	0.4	1.2	0.8
Sr (mg/l)	0.082	0.083	0.089	0.12	0.097
SO ₄ ⁻² (mg/l)	10.0	8.7	14.6	8.8	12.5
S ⁻ (POLAROGRAPHIC) (mg/l)	<1 x 10 ⁻⁵	<1 x 10 ⁻⁵	<1 x 10 ⁻⁵	<1 x 10 ⁻⁵	2 x 10 ⁻⁵
SO ₄ ⁻² (TOTAL BY ICP) (mg/l)	6.2	6.4	7.6	4.7	6.0
NO ₃ ⁻ (mg/l)	1.3	1.3	16.3	6.0	13.5
NO ₂ ⁻ (mg/l)	3.4	1.7	<0.03	<0.03	<0.03
F ⁻ (mg/l)	<0.08	<0.08	<0.08	<0.08	<0.08
PO ₄ ⁻³ (mg/l)	<0.2	<0.2	<0.2	<0.2	<0.2
P (TOTAL BY ICP) (mg/l)	<0.1	<0.1	<0.1	<0.1	0.18
Cl ⁻ (mg/l)	0.59	0.40	0.97	0.55	0.86
Si (mg/l)	1.74	1.80	2.88	8.50	4.50
Al (µg/l)	<30	100	<30	70	100
As (µg/l)	<20	<20	<20	<20	<20
B (µg/l)	20	20	10	20	20
Ba (µg/l)	30	31	14	16	30
Cr (µg/l)	<10	<10	<10	<10	<10
Fe (TOTAL BY ICP) (µg/l)	30	30	<5	20	10
Fe ⁺² (COLORIMETRIC) (µg/l)	<2	<2	<2	<2	<2
Fe ⁺³ (COLORIMETRIC) (µg/l)	19	18	2	15	3
Li (µg/l)	<5	<5	<5	<5	<5
Mn (µg/l)	<3	<3	<3	<3	<3
Mo (µg/l)	<10	<10	<10	<10	<10
Ni (µg/l)	<20	20	<20	20	20
Se (µg/l)	<100	<100	<100	<100	<100
Ti (µg/l)	<5	<5	<5	<5	<5
V (µg/l)	<5	<5	<5	8	<3
Zn (µg/l)	37	11	<5	<5	<5

CHEMICAL COMPOSITION OF TRENCH, WELL AND SPRING WATERS,
MARCH, 1983

CONSTITUENT	TRENCH	TRENCH	WELL #1	SPRINGS
	(1st MANWAY) 3/30/83	(LAST MANWAY) 3/30/83	3/31/83	4/1/83
TEMPERATURE °C	22.0	23.0	20.0	21.5
pH	8.58	8.70	8.25	8.10
Eh (mv)	+245	+265	+355	+355
DISSOLVED O ₂ (mg/l)	5.59	4.71	6.09	4.76
ALKALINITY (As CaCO ₃ mg/l)	41.6	41.0	53.7	55.8
Ca (mg/l)	13.9	12.9	26.3	25.7
Mg (mg/l)	3.2	3.3	4.0	4.0
Na (mg/l)	1.7	1.5	2.1	1.9
K (mg/l)	0.8	0.7	1.0	1.4
Sr (mg/l)	0.072	0.067	0.095	0.096
SO ₄ ⁻² (mg/l)	12.7	11.3	12.1	11.8
S ⁻ (POLAROGRAPHIC) (mg/l)	0.205	0.163	0.215	0.163
SO ₄ ⁻² (TOTAL BY ICP) (mg/l)	12.9	11.7	12.9	12.6
NO ₃ ⁻ (mg/l)	2.1	3.0	16.8	13.6
NO ₂ ⁻ (mg/l)	1.9	2.6	<0.05	<0.05
F ⁻ (mg/l)	<0.1	<0.01	<0.1	<0.1
PO ₄ ⁻³ (mg/l)	<0.2	<0.2	<0.02	<0.2
P (TOTAL BY ICP) (mg/l)	<0.1	<0.1	<0.1	<0.1
Cl ⁻ (mg/l)	1.0	0.95	1.1	0.89
Si (mg/l)	2.3	2.1	3.5	5.5
Al (µg/l)	<30	<30	<30	<30
As (µg/l)	30	<20	<20	<20
B (µg/l)	22	25	<10	<10
Ba (µg/l)	36	35	16	31
Fe (TOTAL BY ICP) (µg/l)	20	61	<10	<10
Fe ⁺² (COLORIMETRIC) (µg/l)	<2	6	<2	<2
Fe ⁺³ (COLORIMETRIC) (µg/l)	18	55	2	4
Li (µg/l)	<10	<10	<10	<10
Mn (µg/l)	<3	<3	<3	<3
Mo (µg/l)	<10	<10	<10	<10
Ni (µg/l)	<20	<20	<20	<20
Se (µg/l)	<100	<100	<100	<100
Ti (µg/l)	<5	<5	<5	<5
V (µg/l)	<5	<5	<5	<5
Zn (µg/l)	10	10	<10	20

CHEMICAL COMPOSITION OF TRENCH, WELL AND SPRING WATERS,
MAY, 1983

CONSTITUENT	TRENCH	TRENCH	WELL #1	SPRINGS
	(2nd MANWAY) 5/5/83	(LAST MANWAY) 5/5/83	5/6/83	5/9/83
TEMPERATURE °C	21.0	19.0	28.5	22.0
pH	9.10	8.70	8.05	7.90
Eh (mv)	+155	+315	+335	+355
DISSOLVED O ₂ (mg/l)	4.64	4.56	1.95	3.91
ALKALINITY (As CaCO ₃ mg/l)	46.0	48.4	64.1	59.9
Ca (mg/l)	13.8	16.9	31.1	25.7
Mg (mg/l)	3.2	3.8	4.7	4.0
Na (mg/l)	1.8	2.2	2.8	1.9
K (mg/l)	0.9	1.1	1.5	1.4
Sr (mg/l)	0.078	0.098	0.12	0.095
SO ₄ ⁻² (mg/l)	14.3	12.6	20.0	11.8
S ⁻ (POLAROGRAPHIC) (mg/l)	0.163	0.197	0.160	0.139
SO ₄ ⁻² (TOTAL BY ICP) (mg/l)	13.5	17.7	18.6	12.6
NO ₃ ⁻ (mg/l)	3.2	4.1	22	14.3
NO ₂ ⁻ (mg/l)	9.3	6.6	<0.05	<0.05
F ⁻ (mg/l)	<0.1	<0.1	<0.1	<0.1
PO ₄ ⁻³ (mg/l)	<0.2	<0.2	<0.2	<0.2
P (TOTAL BY ICP) (mg/l)	0.1	<0.1	<0.1	<0.1
Cl ⁻ (mg/l)	0.77	1.0	1.0	0.89
Si (mg/l)	2.1	2.5	4.9	5.5
Al (µg/l)	<30	<30	<30	<30
As (µg/l)	<20	<20	<20	<20
B (µg/l)	<10	25	<10	<10
Ba (µg/l)	26	19	23	31
Cr (µg/l)	<20	<20	<20	<20
Fe (TOTAL BY ICP) (µg/l)	41	18	71*	<10
Fe ⁺² (COLORIMETRIC) (µg/l)	<2	<2	4*	3
Fe ⁺³ (COLORIMETRIC) (µg/l)	32	12	67*	24
Li (µg/l)	<10	<10	<10	<10
Mn (µg/l)	<3	<3	<3	<3
Mo (µg/l)	<10	<10	<10	<10
Ni (µg/l)	<20	<20	<20	<20
Se (µg/l)	<100	<100	<100	<100
Ti (µg/l)	<5	<5	<5	<5
V (µg/l)	<5	<5	<5	<5
Zn (µg/l)	<5	<5	87	20

* SAMPLED WITH IEA PUMP HAVING STAINLESS STEEL COMPONENTS - POSSIBLE CONTAMINATION

CHEMICAL COMPOSITION OF TRENCH AND SPRING WATERS,
JUNE, 1983

CONSTITUENT	TRENCH	TRENCH	SPRINGS
	(2nd MANWAY) 6/16/83	(LAST MANWAY) 6/16/83	6/20/83
TEMPERATURE °C	18.5	20.5	24.8
pH	7.30	7.20	7.80
Eh (mv)	+195	+180	+275
DISSOLVED O ₂ (mg/l)	6.88	7.06	4.55
ALKALINITY (As CaCO ₃ -mg/l)	36.5	39.7	58.3
Ca (mg/l)	14.7	14.8	27.3
Mg (mg/l)	3.3	3.4	4.2
Na (mg/l)	1.9	1.9	2.2
K (mg/l)	0.8	0.8	1.4
Si (mg/l)	0.079	0.079	0.10
SO ₄ ⁻² (mg/l)	15.2	15.0	14.5
S ⁻ (POLAROGRAPHIC) (mg/l)	0.051	0.078	0.068
SO ₄ ⁻² (TOTAL BY ICP) (mg/l)	15.3	15.3	15.3
NO ₃ ⁻ (mg/l)	0.73	1.2	15.5
NO ₂ ⁻ (mg/l)	0.60	0.80	<0.05
F ⁻ (mg/l)	<0.1	<0.1	<0.1
PO ₄ ⁻³ (mg/l)	0.80	1.1	<0.2
P (TOTAL BY ICP) (mg/l)	0.6	0.7	<0.1
Cl ⁻ (mg/l)	1.0	1.6	1.0
Si (mg/l)	2.4	2.4	5.7
Al (µg/l)	<30	<30	<30
As (µg/l)	<20	<20	<20
B (µg/l)	<10	<10	<10
Ba (µg/l)	29	29	32
Cr (µg/l)	<20	<20	<20
Fe (TOTAL BY ICP) (µg/l)	16	20	<10
Fe ⁺² (COLORIMETRIC) (µg/l)	5.9	7.6	<1
Fe ⁺³ (COLORIMETRIC) (µg/l)	7.6	9.5	3.5
Li (µg/l)	<10	<10	<10
Mn (µg/l)	<3	<3	<3
Mo (µg/l)	<10	<10	<10
Ni (µg/l)	<20	<20	<20
Se (µg/l)	<100	<100	<100
Ti (µg/l)	<5	<5	<5
V (µg/l)	<5	<5	<5
Zn (µg/l)	10	10	<10

CHEMICAL COMPOSITION OF TRENCH, WELL AND SPRING WATERS,
SEPTEMBER, 1983

CONSTITUENT	TRENCH	WELL #1	SPRINGS
	(2nd MANWAY) 9/9/83	9/9/83	9/12/83
TEMPERATURE °C	23.0	32.0	32.0
pH	7.80	8.20	8.0
Eh (mv)	+190	+300	+385
DISSOLVED O ₂ (mg/l)	6.00	2.08	4.17
ALKALINITY (As CaCO ₃ -mg/l)	44.0	61.3	58.2
DOC (mg/l)	0.70	0.82	0.44
Ca (mg/l)	16.8	27.5	24.3
Mg (mg/l)	3.5	4.1	3.7
Na (mg/l)	2.1	3.8	3.3
K (mg/l)	0.84	1.7	1.6
Si (mg/l)	0.093	0.104	0.093
SO ₄ ⁻² (mg/l)	17	16	13
S ⁻ (POLAROGRAPHIC) (mg/l)	0.039	0.458	1.15
SO ₄ ⁻² (TOTAL BY ICP) (mg/l)	16.9	16.5	14.1
NO ₃ ⁻ (mg/l)	2.9	13.1	11.0
NO ₂ ⁻ (mg/l)	1.1	<0.05	<0.05
F ⁻ (mg/l)	<0.25	<0.25	<0.25
PO ₄ ⁻³ (mg/l)	-	-	-
P (TOTAL BY ICP) (mg/l)	<0.1	0.17	<0.1
Cl ⁻ (mg/l)	0.97	0.97	0.86
Si (mg/l)	1.8	5.3	6.9
Al (µg/l)	<30	<30	<30
As (µg/l)	<20	<20	<20
B (µg/l)	20	20	20
Ba (µg/l)	29	15	27
Cr (µg/l)	<20	<20	<20
Fe (TOTAL BY ICP) (µg/l)	35	<10	<10
Fe ⁺² (COLORIMETRIC) (µg/l)	15.3	4.2	2.9
Fe ⁺³ (COLORIMETRIC) (µg/l)	20.8	<2	4.9
Li (µg/l)	<10	<10	<10
Mn (µg/l)	<3	<3	<3
Mo (µg/l)	<10	<10	<10
Ni (µg/l)	<20	<20	<20
Se (µg/l)	<100	<100	<100
Ti (µg/l)	<5	<5	<5
V (µg/l)	<5	<5	<5
Zn (µg/l)	<10	<10	16

APPENDIX C

ANALYTICAL AND CALCULATED CHARGE-FORM
DISTRIBUTIONS AND CALCULATED SATURATION
INDICES FOR SELECTED SOLIDS

Table C.1a. Analytical and Calculated Charge-form Distribution (%).

<u>Sample Date</u>	<u>Sample Location</u>	⁷⁶ As		Cationic		Anionic		Nonionic	
		<u>Anal.</u>	<u>Calc.</u>	<u>Anal.</u>	<u>Calc.</u>	<u>Anal.</u>	<u>Calc.</u>	<u>Anal.</u>	<u>Calc.</u>
Feb. 1974	Influent Spring	0.0				77.8		22.2	
March 1981	Influent Spring								
June 1981	Influent Spring								
Sept. 1981	Influent Spring								
Jan. 1983	Manway 1		0.0		100				0.0
	Well 1		0.0		100				0.0
	Well 3		0.0		100				0.0
	Spring (1)		0.0		100				0.0
	Spring (2)		0.0		100				0.0
Feb. 1983	Manway 1		0.0		99.3				0.0
	Manway 10		0.0		99.3				0.0
	Well 1		0.0		100				0.0
	Well 3		0.0		100				0.0
	Spring		0.0		100				0.0
March 1983	Manway 1		0.0		100				0.0
	Manway 10		0.0		98.9				0.0
	Well 1		0.0		100				0.0
	Spring		0.0		100				0.0
May 1983	Manway 2		0.0		99.2				0.0
	Manway 10		0.0		98.9				0.0
	Well 1		0.0		100				0.0
	Spring		0.0		100				0.0
June 1983	Manway 2		0.0		100				0.0
	Manway 10		0.0		100				0.0
	Spring		0.0		100				0.0
Sept. 1983	Manway 2		0.0		100				0.0
	Well 1		0.0		100				0.0
	Spring		0.0		100				0.0

Table C.1b. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	¹⁴⁰ Ba		Anionic		Nonionic	
		Cationic Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Trench Spring	93.6		0.5		5.9	
March 1981	Trench Spring						
June 1981	Trench Spring	98.8		1.1		0.1	
Sept. 1981	Influent Spring	95.5 10.8		4.5 86.5		0 2.7	
Jan. 1983	Manway 1 Well 1 Well 3 Spring (1) Spring (2)	100 0		0 100		0 0	
Feb. 1983	Manway 1 Manway 10 Well 1 Well 3 Spring	98.6 99.6		1.3 0.2		<0.08 <0.2	
March 1983	Manway 1 Manway 10 Well 1 Spring	99.9 99.8 54.3		<0.02 0.2 <42.7		0.02 0 <3.0	
May 1983	Manway 2 Manway 10 Well 1 Spring	99.7 98.8		0.2 1.1		0.1 0.1	
June 1983	Manway 2 Manway 10 Spring	99.3 99.9		0.6 0.1		0 0	
Sept. 1983	Manway 2 Well 1 Spring	99.9		0.1		0	

Note: Stable element concentrations for Ba not given in any water analysis.

Table C.2a. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	¹⁴¹ Ce					
		Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring	78.1		16.3		5.7	
March 1981	Influent Spring	99	100	1		<0.2	
June 1981	Influent Spring	76.3	100	21.1		2.5	
Sept. 1981	Influent Spring						
Jan. 1983	Manway 1		100				
	Well 1		100				
	Well 3		100				
	Spring (1)		100				
	Spring (2)		100				
Feb. 1983	Manway 1	96.0	100	<3.8		<0.2	
	Manway 10	97.5	100	1.1		1.4	
	Well 1		100				
	Well 3		100				
	Spring		100				
March 1983	Manway 1	95.4	100	0.8		3.8	
	Manway 10	94.8	100	1.8		3.5	
	Well 1		100				
	Spring		100				
May 1983	Manway 2	95.8	100	2.6		1.6	
	Manway 10	59.9	100	37.2		2.9	
	Well 1		100				
	Spring		100				
June 1983	Manway 2	63.6	100	34.4		2.0	
	Manway 10	64.2	100	31.2		4.6	
	Spring		100				
Sept. 1983	Manway 2	95.1	100	0.5		4.4	
	Well 1		100				
	Spring		100				

Note: Ce concentrations not always given.

Table C-2b. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	¹⁴⁴ Ce		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring	94.0		10.1		5.9	
March 1981	Influent Spring	99.5 55	100 100	0.5 <30		0 <15	
June 1981	Influent Spring	78	100 100	19		2.4	
Sept. 1981	Influent Spring	82.7		16.9		0.4	
Jan. 1983	Manway 1 Well 1 Well 3 Spring (1) Spring (2)		100 100 100 100 100				
Feb. 1983	Manway 1 Manway 10 Well 1 Well 3 Spring	87.4	100 100 100 100 100	<12.3		<0.3	
March 1983	Manway 1 Manway 10 Well 1 Spring	89.3 91.2	100 100 100 100	<6.8 <6.6		3.9 2.2	
May 1983	Manway 2 Manway 10 Well 1 Spring	89.1 60.4	100 100 100 100	8.1 29.1		2.8 10.4	
June 1983	Manway 2 Manway 10 Spring	62.4 60.0	100 100 100	35.5 34.8		2.1 5.2	
Sept. 1983	Manway 2 Well 1 Spring	92.8	100 100 100	1.3		5.9	

Table C.3a. Analytical and Calculated Charge-form Distribution (%).

<u>Sample Date</u>	<u>Sample Location</u>	¹³⁴ Cs					
		<u>Cationic</u>		<u>Anionic</u>		<u>Nonionic</u>	
		<u>Anal.</u>	<u>Calc.</u>	<u>Anal.</u>	<u>Calc.</u>	<u>Anal.</u>	<u>Calc.</u>
Feb. 1974	Influent Spring	98.4		<1.6		0	
March 1981	Influent Spring		100 100				
June 1981	Influent Spring	100	100 100				
Sept. 1981	Influent Spring	99.7 <47.6		0.3 42.9		9.5	
Jan. 1983	Manway 1 Well 1 Well 3 Spring (1) Spring (2)						
Feb. 1983	Manway 1 Manway 10 Well 1 Well 3 Spring						
March 1983	Manway 1 Manway 10 Well 1 Spring						
May 1983	Manway 2 Manway 10 Well 1 Spring						
June 1983	Manway 2 Manway 10 Spring						
Sept. 1983	Manway 2 Well 1 Spring						

Table C.3b. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	¹³⁷ Cs					
		Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring	99.5		<0.5		0	
March 1981	Influent Spring	100	100				
June 1981	Influent Spring	100	100				
Sept. 1981	Influent Spring	100 48.4		0 45.2		0 <6.5	
Jan. 1983	Manway 1 Well 1 Well 3 Spring (1) Spring (2)	99.9	100	<0.1		<0.1	
Feb. 1983	Manway 1 Manway 10 Well 1 Well 3 Spring	99.9 100 66.2	100 100 100	<0.1 13.5		 20.3	
March 1983	Manway 1 Manway 10 Well 1 Spring	100 99.8 60.4	100 100 100	0.0 <0.1 <36.0		0.0 <0.1 <3.6	
May 1983	Manway 2 Manway 10 Well 1 Spring	100 100 86.7 100	100 100 100 100	0 0 5.5		0 0 7.8	
June 1983	Manway 2 Manway 10 Spring	100 100	100 100				
Sept. 1983	Manway 2 Well 1 Spring	100	100				

Table C.4a. Analytical and Calculated Charge-form Distribution (%).

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Sample Date	Sample Location	Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring	19.5 0.4		78.8 92.0		1.7 7.6	
March 1981	Influent Spring		0 0		100 100		0 0
June 1981	Influent Spring	46 0.9	0 0	<54 96	100 100	0 3	0 0
Sept. 1981	Influent Spring	4.3 2.3		95.6 97.4		0 0.2	
Jan. 1983	Manway 1 Well 1 Well 3 Spring (1) Spring (2)	3.0 4.9 5.7		96.5 94.9 92.5	100 100 100	0.5 10.2 1.6	
Feb. 1983	Manway 1 Manway 10 Well 1 Well 3 Spring	4.9 3.7 4.8 4.0 <0.7	0 0 0 0 0	88.6 96.2 95.0 94.0 91.0	100 100 100 100 100	0.2 <0.1 0.2 <2.0 8.0	0 0 0 0 0
March 1983	Manway 1 Manway 10 Well 1 Spring	3.1 0.5 2.4 1.5	0 0 0 0	96.8 99.4 97.6 94.7	100 100 100 100	0.1 0.1 0 3.8	0 0 0 0
May 1983	Manway 2 Manway 10 Well 1 Spring	6.2 7.4 5.3 2.6	0 0 0 0	93.7 92.6 94.6 96.6	100 100 100 100	0.1 0.1 0.1 0.8	0 0 0 0
June 1983	Manway 2 Manway 10 Spring	30.3 0 0	0 0 0	69.6 100 100	100 100 100	0 0 0	0 0 0
Sept. 1983	Manway 2 Well 1 Spring	1.2 0 0.9	0 0 0	98.7 100 99.1	100 100 100	0.1 0 0	0 0 0

Table C.4b. Analytical and Calculated Charge-form Distribution (%).

133₁

Sample Date	Sample Location	Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring	20.6 0.3		77.9 95.1		1.5 4.5	
March 1981	Influent Spring						
June 1981	Influent Spring						
Sept. 1981	Influent Spring						
Jan. 1983	Manway 1 Well 1 Well 3 Spring (1) Spring (2)	0.0 <0.3	0 0	99.6 99.4	100 100	0.4 0.3	0 0
Feb. 1983	Manway 1 Manway 10 Well 1 Well 3 Spring	<0.9 0.0 12.5 0.0	0 0 0 0	99.4 99.4 83.8 100	100 100 100 100	0.7 0.6 3.4 0.0	0 0 0 0
March 1983	Manway 1 Manway 10 Well 1 Spring						
May 1983	Manway 2 Manway 10 Well 1 Spring						
June 1983	Manway 2 Manway 10 Spring						
Sept. 1983	Manway 2 Well 1 Spring						

Table C.5a. Analytical and Calculated Charge-form Distribution (%).

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<u>Sample Date</u>	<u>Sample Location</u>	<u>Cationic</u>		<u>Anionic</u>		<u>Nonionic</u>	
		<u>Anal.</u>	<u>Calc.</u>	<u>Anal.</u>	<u>Calc.</u>	<u>Anal.</u>	<u>Calc.</u>
Feb. 1974	Influent Spring	99.8		0.2			
March 1981	Influent Spring		99.9 99.9				
June 1981	Influent Spring		99.9 99.9				
Sept. 1981	Influent Spring						
Jan. 1983	Manway 1 Well 1 Well 3 Spring (1) Spring (2)		99.9 99.9 99.9 99.9 99.9				
Feb. 1983	Manway 1 Manway 10 Well 1 Well 3 Spring		99.8 99.8 99.9 99.9 99.9				
March 1983	Manway 1 Manway 10 Well 1 Spring		99.9 99.8 99.9 99.9				
May 1983	Manway 2 Manway 10 Well 1 Spring		99.8 99.8 99.8 99.9				
June 1983	Manway 2 Manway 10 Spring		99.9 99.9 99.8				
Sept. 1983	Manway 2 Well 1 Spring		99.9 99.8 99.8				

Table C.5b. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	⁵⁴ Mn					
		Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring	99.4		0.5		0.1	
March 1981	Influent Spring	100					
June 1981	Influent Spring	99.9	98.4 98.4	0.1	0.0 0.0	0.0	1.6 1.6
Sept. 1981	Influent Spring	96.6	98.8 98.6	3.3	0.0 0.0	0.0	1.2 1.4
Jan. 1983	Manway 1	99	98.4	1.0	0.0	0.0	1.6
	Well 1	99.5	98.6	0.5	0.0	0.0	1.3
	Well 3		98.7		0.0		1.2
	Spring (1)		98.5		0.0		1.3
	Spring (2)		98.4		0.0		1.4
Feb. 1983	Manway 1		99.0		0.0		1.0
	Manway 10		100		0.0		0.0
	Well 1		98.5		0.0		1.4
	Well 3		100		0.0		0.0
	Spring		98.6		0.0		1.2
March 1983	Manway 1		97.9		0.0		1.4
	Manway 10		97.8		0.0		1.3
	Well 1		98.6		0.0		1.2
	Spring		98.7		0.0		1.2
May 1983	Manway 2		98.5		0.0		1.5
	Manway 10		98.1		0.0		1.3
	Well 1		97.7		0.0		2.0
	Spring		98.7		0.0		1.2
June 1983	Manway 2		98.3		0.0		1.7
	Manway 10		98.3		0.0		1.6
	Spring		98.3		0.0		1.5
Sept. 1983	Manway 2		98.0		0.0		1.9
	Well 1		97.6		0.0		1.7
	Spring		98.2		0.0		1.5

Table C.6a. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	⁹⁹ Mo		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent	3.2		94.3		2.4	
	Spring	0.1		99.4		0.5	
March 1981	Influent						
	Spring						
June 1981	Influent						
	Spring						
Sept. 1981	Influent						
	Spring						
Jan. 1983	Manway 1	1	0.0	99	99.9	0.0	0.0
	Well 1	0.8	0.0	99	100	0.1	0.0
	Well 3		0.0		100		0.0
	Spring (1)		0.0		100		0.0
	Spring (2)		0.0		100		0.0
Feb. 1983	Manway 1	0.0	0.0	100	100	0.0	0.0
	Manway 10	0.9	0.0	99.1	100	0.0	0.0
	Well 1	5.7	0.0	94.3	100	0.0	0.0
	Well 3		0.0		100		0.0
	Spring	0.0	0.0	100	100	0.0	0.0
March 1983	Manway 1	3.5	0.0	96.4	100	0.0	0.0
	Manway 10	0.5	0.0	99.5	100	0.0	0.0
	Well 1	1.8	0.0	98.1	100	0.0	0.0
	Spring		0.0		100		0.0
May 1983	Manway 2	<48.1	0.0	50.6	100	<1.4	0.0
	Manway 10	43.4	0.0	55.5	100	0.8	0.0
	Well 1	<0.2	0.0	99.7	100	<0.1	0.0
	Spring	0	0.0	100	100	0.0	0.0
June 1983	Manway 2		0.0		100		0.0
	Manway 10		0.0		100		0.0
	Spring		0.0		100		0.0
Sept. 1983	Manway 2	0	0.0	100	100	0.0	0.0
	Well 1	2.0	0.0	98.0	100	0.0	0.0
	Spring	0	0.0	100	100	0.0	0.0

Table C.6b. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	^{99m}Tc					
		Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring	2.8 0.1		94.9 99.5		2.3 0.4	
March 1981	Influent Spring						
June 1981	Influent Spring						
Sept. 1981	Influent Spring						
Jan. 1983	Manway 1 Well 1 Well 3 Spring (1) Spring (2)	0.8 <0.8		99.2 99.1		0.0 <0.1	
Feh. 1983	Manway 1 Manway 10 Well 1 Well 3 Spring	0.0 <0.7 5.2 0.0		100 99.2 94.6 100		0.0 0.0 <0.2 0.0	
March 1983	Manway 1 Manway 10 Well 1 Spring	0.0 0.5 0.9 0.0		100 99.5 99.1 100		0.0 0.0 0.0 0.0	
May 1983	Manway 2 Manway 10 Well 1 Spring	0.0 <0.1 0.0		99.6 99.8 100		0.3 <0.1 0.0	
June 1983	Manway 2 Manway 10 Spring						
Sept. 1983	Manway 2 Well 1 Spring	0.0 0.0 0.0		100 100 100		0.0 0.0 0.0	

Note: Rai and Seme 1978 TcO_4^- only stable system.

Table C.7a. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	⁵⁸ Co					
		Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring						
March 1981	Influent Spring	99.8	100	<0.1	0.0	0.0	0.0
June 1981	Influent Spring	<86.5 51.4	100 99.7	12.8 37.5	0.0 0.0	0.6 <11.1	0.0 0.3
Sept. 1981	Influent Spring	99.8		<0.2		0.0	
Jan. 1983	Manway 1	99.3	100	0.7	0.0	0.0	0.0
	Well 1	8.2	99.7	90.2	0.0	1.1	0.3
	Well 3	29.7	99.7	58.0	0.0	12.3	0.3
	Spring (1)	5.1	99.7	93.0	0.0	1.8	0.3
	Spring (2)		99.7		0.0		0.3
Feb. 1983	Manway 1	95.7	77.2	3.3	0.0	1.0	22.7
	Manway 10	99.5	73.7	<0.4	0.0	<0.1	26.4
	Well 1		99.7				0.3
	Well 3	52.8	99.1	40.0	0.0	10.2	0.9
	Spring	10.0	99.7	89.0	0.0	<1.0	0.3
March 1983	Manway 1	99.4	98.0	0.4	0.0	0.2	2.0
	Manway 10	99.6	96.5	0.4	0.0	0.06	3.5
	Well 1	36.1	99.5	61.1	0.0	2.8	0.5
	Spring		97.7		0.0		0.3
May 1983	Manway 2	99.6	83.6	0.4	0.0	0.0	16.4
	Manway 10	99.6	96.5	0.3	0.0	0.0	3.5
	Well 1	68.8	99.8	27.3	0.0	<4.0	0.2
	Spring	34.0	99.9	64.1	0.0	1.9	0.1
June 1983	Manway 2	<89.9	100	10.1	0.0	0.0	0.0
	Manway 10	90.4	100	9.2	0.0	0.3	0.0
	Spring	49.2	99.9	43.4	0.0	7.3	0.1
Sept. 1983	Manway 2	99.7	99.9	<0.2	0.0	<0.1	0.1
	Well 1	100	99.5	0.0	0.0	0.0	0.5
	Spring	26.7	99.8	68.6	0.0	4.7	0.2

Table C.7b. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	⁶⁰ Co					
		Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring	96.4		1.9		1.6	
		17.3		73.6		9.1	
March 1981	Influent Spring	100	100	0.0	0.0	0.0	0.0
		47.1	100	43.7	0.0	5.6	0.0
June 1981	Influent Spring	98.8	100	1.1	0.0	0.06	0.0
		45.5	99.7	45.1	0.0	9.4	0.0
Sept. 1981	Influent Spring	89.4		10.3	0.0	0.3	
		27.5		69.0	0.0	3.5	
Jan. 1983	Manway 1	99.7	100	0.2	0.0	0.1	0.0
	Well 1	65.1	99.7	33.8	0.0	1.1	0.3
	Well 3	64.2	99.7	34.4	0.0	1.3	0.3
	Spring (1)	45.0	99.7	51.0	0.0	4.0	0.3
	Spring (2)		99.7		0.0		0.3
Feb. 1983	Manway 1	86.3	77.2	10.1	0.0	3.6	22.7
	Manway 10	89.6	73.7	9.2	0.0	1.1	26.4
	Well 1	35.4	99.7	63.1	0.0	1.5	0.3
	Well 3	52.0	99.1	44.8	0.0	3.2	0.9
	Spring	26.0	99.7	67.0	0.0	7.0	0.3
March 1983	Manway 1	95.6	98.0	3.7	0.0	0.7	2.0
	Manway 10	82.3	96.5	15.8	0.0	1.9	3.5
	Well 1	36.2	99.5	62.4	0.0	1.4	0.5
	Spring	40.7	99.7	52.1	0.0	7.2	0.3
May 1983	Manway 2	96.3	83.6	3.3	0.0	0.4	16.4
	Manway 10	97.2	96.5	2.6	0.0	0.2	3.5
	Well 1	64.3	99.8	34.6	0.0	1.0	0.2
	Spring	40.7	99.9	55.1	0.0	4.2	0.1
June 1983	Manway 2	99.1	100	0.8	0.0	0.0	0.0
	Manway 10	99.4	100	0.6	0.0	0.0	0.0
	Spring	45.7	99.9	48.3	0.0	6.3	0.1
Sept. 1983	Manway 2	98.7	99.9	0.9	0.0	0.4	0.1
	Well 1	61.5	99.5	36.5	0.0	2.0	0.5
	Spring	48.7	99.8	47.6	0.0	3.6	0.2

Table C.8a. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	⁵¹ Cr					
		Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent	43.9		50.7		5.4	
	Spring	1.1		98.2		0.6	
March 1981	Influent	<93.2		4.3		2.5	
	Spring	<4.2		<6.2		89.6	
June 1981	Influent	<93.1		5.4		1.6	
	Spring	3.2		20.9		75.9	
Sept. 1981	Influent	<57.1		<38.9		4.0	
	Spring	9.5		3.6		54.8	
Jan. 1983	Manway 1		77.4		0.0		22.5
	Well 1		1.8		94.6		3.6
	Well 3		29.2		5.0		65.8
	Spring (1)		0.4		98.6		0.9
	Spring (2)		22.0		28.4		49.6
Feb. 1983	Manway 1		2.2		44.4		53.4
	Manway 10		1.9		47.3		50.8
	Well 1		29.9		4.8		64.5
	Well 3		17.0		14.5		68.5
	Spring		31.9		4.1		64.0
March 1983	Manway 1						
	Manway 10						
	Well 1						
	Spring						
May 1983	Manway 2	85.1	3.0	5.8	38.5	9.1	58.5
	Manway 10	87.2	8.7	9.4	20.6	3.3	70.6
	Well 1		34.1		4.6		61.2
	Spring		43.4		2.3		54.4
June 1983	Manway 2		78.5		0.0		21.2
	Manway 10		82.5		0.0		17.2
	Spring		44.9		1.7		52.9
Sept. 1983	Manway 2		50.9		1.2		47.5
	Well 1		26.4		5.9		67.2
	Spring		3.6		90.7		5.8

Table C.8b. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	¹⁵⁴ Eu		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring						
March 1981	Influent Spring						
June 1981	Influent Spring						
Sept. 1981	Influent Spring						
Jan. 1983	Manway 1 Well 1 Well 3 Spring (1) Spring (2)						
Feb. 1983	Manway 1	71	100	29	0.0	<3	0.0
	Manway 10	13.8	100	78.5	0.0	7.7	0.0
	Well 1	0.0	100	77.4	0.0	22.6	0.0
	Well 3 Spring						
March 1983	Manway 1	>99	100	<20	0.0	3.3	0.0
	Manway 10	<67.1	100	<26.8	0.0	6.0	0.0
	Well 1 Spring						
May 1983	Manway 2						
	Manway 10	61.0	100	34.3	0.0	4.8	0.0
	Well 1 Spring						
June 1983	Manway 2	87.8	100	11.7	0.0	0.5	0.0
	Manway 10 Spring						
Sept. 1983	Manway 2 Well 1 Spring						

Note: Rai and Seme 1978 Only cationic species found.

Table C.9a. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	⁵⁹ Fe					
		Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring	29.8		55.3		14.9	
March 1981	Influent Spring	99.9	98.2	<0.1	0.0	0.0	1.4
			98.2		0.0		1.4
June 1981	Influent Spring	<36.0	98.7	63.2	0.0	0.8	1.1
			98.8		0.0		1.1
Sept. 1981	Influent Spring	29.8		68.9		<1.3	
		69.4		<23.5		7.1	
Jan. 1983	Manway 1	69	92.9	30	0.0	1	5.2
	Well 1	16	98.6	82.9	0.0	<1	1.3
	Well 3		98.7		0.0		1.1
	Spring (1)		98.6		0.0		1.3
	Spring (2)		98.5		0.0		1.3
Feb. 1983	Manway 1	70	100	30	0.0	0.0	0.0
	Manway 10	51.5	100	48.5	0.0	0.0	0.0
	Well 1		98.6		0.0		1.3
	Well 3		100		0.0		0.0
	Spring		98.7		0.0		1.2
March 1983	Manway 1	65.9	98.5	33.0	0.0	1.1	1.3
	Manway 10	26.1	98.6	72.2	0.0	1.7	1.1
	Well 1	12.1	98.7	87.9	0.0	0.0	1.1
	Spring		98.7		0.0		1.1
May 1983	Manway 2	47.2	98.5	51.9	0.0	0.9	1.2
	Manway 10	34.0	98.6	64.8	0.0	1.2	1.2
	Well 1	<33.8	97.8	>59.4	0.0	<6.7	2.0
	Spring	<42.7	98.6	>51.3	0.0	<6.0	1.2
June 1983	Manway 2	52.7	96.9	46.1	0.0	1.2	2.6
	Manway 10	40.2	96.6	57.2	0.0	2.6	2.9
	Spring		98.4		0.0		1.4
Sept. 1983	Manway 2	66.8	97.9	30.6	0.0	2.6	1.8
	Well 1	>76.4	97.3	<23.6	0.0	0.0	1.7
	Spring		98.3		0.0		1.5

Table C.9b. Analytical and Calculated Charge-form Distribution (%).

<u>Sample Date</u>	<u>Sample Location</u>	⁶³ Ni					
		<u>Cationic</u>		<u>Anionic</u>		<u>Nonionic</u>	
		<u>Anal.</u>	<u>Calc.</u>	<u>Anal.</u>	<u>Calc.</u>	<u>Anal.</u>	<u>Calc.</u>
Feb. 1974	Influent Spring						
March 1981	Influent Spring						
June 1981	Influent Spring	77.8 <20.5		11.5 <48.7		10.8 30.8	
Sept. 1981	Influent Spring						
Jan. 1983	Manway 1		15.8		0.0		83.8
	Well 1		2.0		1.8		95.9
	Well 3		1.8		1.9		96.0
	Spring (1)		1.8		1.9		95.9
	Spring (2)		1.6		2.2		95.9
Feb. 1983	Manway 1		0.0		17.8		82.0
	Manway 10		0.0		18.5		81.3
	Well 1		1.6		2.2		95.6
	Well 3		0.0		3.4		95.4
	Spring		1.8		2.0		95.6
March 1983	Manway 1		0.0		4.1		95.0
	Manway 10		0.0		5.3		94.0
	Well 1		1.4		2.5		95.8
	Spring		1.8		1.9		95.9
May 1983	Manway 2		0.0		12.2		87.5
	Manway 10		0.0		5.8		93.6
	Well 1		1.6		2.3		95.7
	Spring		2.6		1.3		95.5
June 1983	Manway 2		16.3		0.0		83.2
	Manway 10		18.3		0.0		81.2
	Spring		2.5		1.4		95.5
Sept. 1983	Manway 2		4.1		0.0		94.4
	Well 1		1.1		3.3		95.3
	Spring		1.8		2.0		95.8

Table c.10a. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	⁶⁵ Zn		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring	97.9		1.6		0.5	
March 1981	Influent Spring	99.9 <18.6	81.7 54.0	0.1 >79.1	0.0 4.7	0.0 <2.3	16.5 41.3
June 1981	Influent Spring	98.1	94.2 41.8	1.5	0.0 8.9	0.4	5.5 48.8
Sept. 1981	Influent Spring	88.6 40.7		10.8 53.8		0.6 <5.5	
Jan. 1983	Manway 1		85.5		0.0		14.5
	Well 1		40.1		9.9		49.4
	Well 3		37.7		10.8		51.0
	Spring (1)		38.4		11.2		49.9
	Spring (2)		34.9		12.8		51.7
Feb. 1983	Manway 1	>22	1.1	<74.6	49.7	< 2.9	48.0
	Manway 10	79.8	1.0	20.0	48.1	0.2	49.8
	Well 1		34.3		13.1		52.1
	Well 3	100	22.5		20.1		57.2
	Spring		38.1		11.3		50.0
March 1983	Manway 1	86.5	17.4	13.3	22.1	0.2	60.2
	Manway 10		12.1		26.2		60.5
	Well 1		30.6		15.0		54.0
	Spring		38.5		11.2		49.9
May 1983	Manway 2	63.5	3.9	<1.4	38.5	35.1	57.1
	Manway 10		11.6		29.0		59.2
	Well 1	>55.5	36.6	<37.3	13.5	<7.1	49.2
	Spring		49.4		6.9		43.1
June 1983	Manway 2	99.7	84.9	0.3	0.0		3.6
	Manway 10	96.1	86.5	3.8	0.0		12.2
	Spring		49.4		7.2		43.1
Sept. 1983	Manway 2	97.5	61.2	2.3	3.2	0.2	35.5
	Well 1		27.8		20.2		51.6
	Spring		40.8		11.2		47.4

Table C.10b. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	⁹⁵ Zr					
		Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring	29.4		57.7		12.9	
March 1981	Influent Spring	99.1	0.0	0.8	93.4	0.1	6.6
			0.0		72		28
June 1981	Influent Spring	48.2	0.0	43.7	98.0	0.2	2.0
			0.0		98.0		2.0
Sept. 1981	Influent Spring	<32.5		54.7		12.9	
Jan. 1983	Manway 1	31.9	0.0	63.8	90.9	4.3	9.1
	Well 1	8.1	0.0	91.9	98.4	0.0	1.6
	Well 3		0.0		98.6		1.4
	Spring (1)		0.0		98.4		1.6
	Spring (2)		0.0		98.6		1.4
Feb. 1983	Manway 1	43.4	0.0	54.6	99.9	2.0	0.1
	Manway 10	35.3	0.0	59.4	100	5.3	0.0
	Well 1	0.0	0.0	49.3	98.6	50.7	1.6
	Well 3		0.0	100	99.2		0.8
	Spring		0.0		98.4		1.6
March 1983	Manway 1	25.8	0.0	73.4	99.5	0.8	0.5
	Manway 10	4.5	0.0	94.7	99.6	0.7	0.4
	Well 1	47.0	0.0	51.8	99.0	1.2	1.0
	Spring		0.0		98.4		1.6
May 1983	Manway 2	23.3	0.0	76.2	99.8	0.5	0.2
	Manway 10	38.4	0.0	61.6	99.6	0.0	0.4
	Well 1	<2.0	0.0	90	98.2	<8	1.8
	Spring	0.0	0.0	100	97.6	0.0	2.4
June 1983	Manway 2	28.2	0.0	70.9	90.9	0.9	9.1
	Manway 10	21.8	0.0	77.1	88.8	1.1	11.2
	Spring		0.0		97.6		2.4
Sept. 1983	Manway 2	51.8	0.0	47.2	96.9	1.0	3.1
	Well 1	22.1	0.0	76.3	98.8	1.6	1.2
	Spring		0.0		98.0		2.0

Table C.11a. Analytical and Calculated Charge-form Distribution (%).

 ^{124}Sb

Sample Date	Sample Location	Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent	49.2		32.2		18.6	
	Spring	0.5		5.4		94.1	
March 1981	Influent	<92.5		3.3	13.7	3.3	86.3
	Spring	0.5		<17.5	100	82	
June 1981	Influent	46		<31.5		22.3	100
	Spring	<1.0		<2.0	100	97.0	
Sept. 1981	Influent	79.3		10.4		10.3	
	Spring	<5.7		17.8		76.4	
Jan. 1983	Manway 1	2.0		89.0	100	9.0	
	Well 1	0.4		90.9	100	8.7	
	Well 3						
	Spring (1)	<1.6		26.1	100	74.3	
	Spring (2)						
Feb. 1983	Manway 1	9.7		79.2	100	11.1	
	Manway 10	6.2		83.2	100	10.6	
	Well 1	2.2		96.3	100	1.9	
	Well 3						
	Spring	0.0		0.0	100	100	
March 1983	Manway 1	<11.4		88.1	100	<0.6	
	Manway 10	5.2		94.6	100	0.1	
	Well 1	<1.0		84.3	100	14.7	
	Spring	2.0		5.3	100	92.7	
May 1983	Manway 2	3.6		96.0	100	0.3	
	Manway 10	4.7		93.4	100	1.9	
	Well 1	<0.1		98.2	100	1.7	
	Spring	<0.8		6.0	100	93.2	
June 1983	Manway 2	25.2		74.0	23.6	0.8	76.4
	Manway 10	<55.9		43.6	4.3	0.4	95.7
	Spring	<1.0		19.7	100	79.3	95.7
Sept. 1983	Manway 2	<23.5		76.1	100	0.4	
	Well 1	0.2		85.9	100	13.9	
	Spring	<1		40.4	100	59.7	

Table C.11b. Analytical and Calculated Charge-form Distribution (%).

 ^{125}Sb

Sample Date	Sample Location	Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent Spring	<59.7		23.9		16.4	
		<0.3		4.2		95.5	
March 1981	Influent Spring	43	0.0	35.6	13.7	21.3	86.3
		0.4	0.0	19.2	100	80.3	0.0
June 1981	Influent Spring	23	0.0	46	0.0	30	100
		0.4	0.0	7.5	100	92.1	0.0
Sept. 1981	Influent Spring	36.2		23.5		46.3	
		0.4		16.0		83.6	
Jan. 1983	Manway 1	0.0	0.0	98.0	100	2.0	0.0
	Well 1	1.0	0.0	83.3	100	15.7	0.0
	Well 3	1.7	0.0	96.5	100	1.8	0.0
	Spring (1)	0.6	0.0	23.3	100	76.1	0.0
	Spring (2)						
Feb. 1983	Manway 1						
	Manway 10	20.4	0.0	67.4	100	12.2	0.0
	Well 1	0.4	0.0	94.6	100	5.0	0.0
	Well 3	<4.8	0.0	72.4	100	22.9	0.0
	Spring	2.0	0.0	0.0	100	98.0	0.0
March 1983	Manway 1	<29.7	0.0	>69.9	100	0.4	0.0
	Manway 10	<23.0	0.0	99.1	100	0.9	0.0
	Well 1	<0.7	0.0	82.2	100	18.1	0.0
	Spring	0.0	0.0	17.0	100	83.0	0.0
May 1983	Manway 2	0.0	0.0	100	100	0.0	0.0
	Manway 10	14.2	0.0	84.8	100	1.0	0.0
	Well 1	<0.8	0.0	97.5	100	1.7	0.0
	Spring	0.5	0.0	35.7	100	63.8	0.0
June 1983	Manway 2	27.3	0.0	72.6	23.6	0.1	76.4
	Manway 10	0.0	0.0	100	4.3	0.0	95.7
	Spring	<0.4	0.0	21.3	100	78.3	0.0
Sept. 1983	Manway 2	0.0	0.0	100	100	0.0	0.0
	Well 1	0.7	0.0	84.6	100	14.7	0.0
	Spring	0.3	0.0	38.6	100	61.1	0.0

Table C.12a. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	^{103}Ru					
		Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent	49.1		37.2		13.7	
	Spring	3.7		91.0		5.3	
March 1981	Influent	96.5	100	3.4	0.0	0.1	0.0
	Spring	4.8	100	91.5	0.0	3.6	0.0
June 1981	Influent	28	100	67.8	0.0	18.3	0.0
	Spring	13.2		81.8		5.0	
Sept. 1981	Influent	70.4		<26.8		2.8	
	Spring	39.7		74.5		5.2	
Jan. 1983	Manway 1	46.9	100	48.5	0.0	4.6	0.0
	Well 1	11.9	100	86.8	0.0	1.3	0.0
	Well 3	8.4	100	91.2	0.0	1.4	0.0
	Spring (1)	13.0	100	83.2	0.0	3.6	0.0
	Spring (2)						
Feb. 1983	Manway 1	50.4	100	46.8	0.0	2.7	0.0
	Manway 10	62.1	100	35.8	0.0	2.0	0.0
	Well 1	21.8	100	75.6	0.0	2.6	0.0
	Well 3	15.0	100	82.0	0.0	3.0	0.0
	Spring	3.0	100	92.0	0.0	5.0	0.0
March 1983	Manway 1	48.8	100	45.9	0.0	5.3	0.0
	Manway 10	21.4	100	72.8	0.0	5.8	0.0
	Well 1	4.0	100	95.6	0.0	0.4	0.0
	Spring	13.0	100	80.0	0.0	7.0	0.0
May 1983	Manway 2	48.7	100	46.9	0.0	4.4	0.0
	Manway 10	18.6	100	23.9	0.0	1.7	0.0
	Well 1	35.6	100	61.1	0.0	3.3	0.0
	Spring	14.3	100	82.3	0.0	3.4	0.0
June 1983	Manway 2	38.4	100	57.3	0.0	4.3	0.0
	Manway 10	31.7	100	63.0	0.0	5.3	0.0
	Spring	10.4	100	86.0	0.0	3.6	0.0
Sept. 1983	Manway 2	40.8	100	51.3	0.0	7.9	0.0
	Well 1	12.3	100	86.3	0.0	1.4	0.0
	Spring	9.9	100	87.5	0.0	2.5	0.0

Table C.12b. Analytical and Calculated Charge-form Distribution (%).

Sample Date	Sample Location	^{106}Ru					
		Cationic		Anionic		Nonionic	
		Anal.	Calc.	Anal.	Calc.	Anal.	Calc.
Feb. 1974	Influent	59.4		34.9		5.6	
	Spring	2.4		92.6		5.0	
March 1981	Influent	98.0	100	2.0	0.0	3.4	0.0
	Spring	9.4	100	87.2	0.0	3.4	0.0
June 1981	Influent	37.4	100	57.2	0.0	5.2	0.0
	Spring	11.3	100	82.0	0.0	6.7	0.0
Sept. 1981	Influent	58.4		29.9		11.7	
	Spring	19.0		75.0		5.1	
Jan. 1983	Manway 1						
	Well 1	10.9	100	87.6		1.3	
	Well 3	9.7	100	89.0		1.3	
	Spring (1)	52.2	100	80.6		4.0	
	Spring (2)						
Feb. 1983	Manway 1						
	Manway 10						
	Well 1	21.2	100	72.5		6.4	
	Well 3	21.3	100	75.7		3.0	
	Spring	4.0	100	88.0		7.0	
March 1983	Manway 1	86.1	100	6.9		6.9	
	Manway 10						
	Well 1	10.0	100	89.3		0.7	
	Spring	14.8	100	79.0		6.2	
May 1983	Manway 2	63.5	100	33.2		3.3	
	Manway 10	64.3	100	32.8		2.9	
	Well 1	36.5	100	60.0		3.5	
	Spring	14.4	100	82.2		3.4	
June 1983	Manway 2	46.1	100	50.4		3.5	
	Manway 10	11.0	100	81.5		6.9	
	Spring	12.6	100	82.0		4.6	
Sept. 1983	Manway 2	47.3	100	47.3		5.5	
	Well 1	17.2	100	81.2		1.6	
	Spring	12.0	100	85.0		2.7	

Table C.13. Calculated Saturation Indices.

<u>Sample Date</u>	<u>Sample Location</u>	<u>Calcite</u>	<u>Quartz</u>	<u>Christobalite</u>
March 1981	Influent	-1.039	0.043	-0.400
	Spring	<u>-0.301</u>	0.312	<u>-0.126</u>
June 1981	Influent	-1.682	0.019	-0.413
	Spring	<u>-0.068</u>	<u>-0.329</u>	<u>-0.099</u>
Jan. 1983	Manway 1	-1.046	-0.126	-0.552
	Well 1	0.043	0.306	-0.126
	Well 3	<u>0.079</u>	0.305	<u>-0.126</u>
	Spring (1)	<u>0.104</u>	0.093	<u>-0.335</u>
	Spring (2)	<u>0.046</u>	<u>0.543</u>	<u>-0.115</u>
Feb. 1983	Manway i	0.882	-0.129	-0.563
	Manway 10	0.911	<u>-0.082</u>	-0.519
	Well 1	0.083	<u>0.171</u>	-0.267
	Well 3	<u>0.265</u>	<u>0.571</u>	0.141
	Spring	<u>0.115</u>	0.267	<u>-0.159</u>
March 1983	Manway 1	0.203	-0.051	-0.476
	Manway 10	<u>0.291</u>	<u>-0.113</u>	-0.536
	Well 1	<u>0.226</u>	<u>0.172</u>	-0.256
	Spring	<u>0.110</u>	<u>0.346</u>	<u>-0.079</u>
May 1983	Manway 2	0.676	-0.110	-0.537
	Manway 10	0.411	<u>0.030</u>	-0.400
	Well 1	<u>0.288</u>	<u>0.188</u>	-0.225
	Spring	<u>-0.047</u>	<u>0.341</u>	-0.084
June 1983	Manway 2	-1.117	0.023	-0.406
	Manway 10	-1.164	<u>0.007</u>	-0.420
	Spring	<u>0.003</u>	<u>0.312</u>	<u>-0.107</u>
Sept. 1983	Manway 2	-0.438	-0.159	-0.582
	Well 1	0.426	0.165	-0.241
	Spring	<u>0.156</u>	0.284	-0.123

* $\frac{1}{2}$ detection limit

Table C.14. Calculated Saturation Indices.

<u>Sample Date</u>	<u>Sample Location</u>	<u>Anorthite</u>	<u>Albite</u>
March 1981	Influent	-4.352	-1.821
	Spring	-2.309	-0.310
June 1981	Influent	-4.820	-2.351
	Spring	-1.996	-0.396
Jan. 1983	Manway 1	-3.994	-2.495
	Well 1	-2.496	-0.928
	Well 3	-2.462	-0.913
	Spring (1)	-2.821	-1.483
	Spring (2)	-2.007	-0.081
Feb. 1983	Manway 1	-3.747	-2.306
	Manway 10	-1.981	-1.284
	Well 1	-3.441	-1.513
	Well 3	-0.975	0.506
	Spring	-1.237	-0.394
March 1983	Manway 1	-3.668	-2.156
	Manway 10	-3.819	-2.406
	Well 1	-3.044	-1.415
	Spring	-2.718	-0.962
May 1983	Manway 2	-3.774	-2.276
	Manway 10	-3.423	-1.757
	Well 1	-2.944	-1.355
	Spring	-2.787	-1.016
June 1983	Manway 2	-4.368	-2.291
	Manway 10	-4.401	-2.354
	Spring	-2.766	-1.048
Sept. 1983	Manway 2	-3.968	-2.486
	Well 1	-3.063	-1.344
	Spring	-2.871	-1.049

Table C.15. Calculated Saturation Indices.

Sample Date	Sample Location	Allophane (A)	Allophane (F)	Kaolinite
March 1981	Influent	0.023*	0.915*	5.167*
	Spring	<u>0.133*</u>	1.105*	5.356*
June 1981	Influent	0.204*	0.568*	5.704*
	Spring	<u>-0.336*</u>	0.660*	5.849*
Jan. 1983	Manway 1	-0.392	0.476	4.955
	Well 1	<u>-0.395</u>	0.601	4.946
	Well 3	<u>-0.419</u>	-0.585	4.876
	Spring (1)	-0.564	0.432	4.429
	Spring (2)	-0.529	0.475	5.252
Feb. 1983	Manway 1	-1.338	-0.166	1.825
	Manway 10	<u>-0.333*</u>	<u>0.847*</u>	3.660*
	Well 1	<u>-0.451</u>	0.553	4.219
	Well 3	-0.218	0.826	5.853
	Spring	<u>-0.100</u>	0.896	5.923
March 1983	Manway 1	-1.290	-0.218	2.703
	Manway 10	-1.498	<u>-0.406</u>	2.257
	Well 1	-0.838	0.182	3.900
	Spring	-0.830	<u>0.166</u>	4.401
May 1983	Manway 2	-1.675	-0.519	1.672
	Manway 10	-1.100	-0.008	2.901
	Well 1	-1.392	<u>-0.404</u>	3.599
	Spring	-0.737	<u>0.227</u>	4.686
June 1983	Manway 2	-0.478	0.390	4.754
	Manway 10	-0.500	0.352	4.830
	Spring	-0.949	0.015	4.441
Sept. 1983	Manway 2	-0.837	0.111	3.793
	Well 1	-1.834	-0.822	2.940
	Spring	-1.623	-0.643	3.576

All used $\frac{1}{2}$ detection limit except *.

Table C.16. Calculated Saturation Indices.

Sample Date	Sample Location	Gibbsite	Mont-CA	Halloysite
March 1981	Influent	0.675	0.187	1.750
	Spring	0.740	1.635	2.462
June 1981	Influent	0.950	0.755	2.357
	Spring	0.461	1.351	2.032
Jan. 1983	Manway 1	0.713	0.027	1.643
	Well 1	0.285	0.781	1.600
	Well 3	0.250	0.715	1.529
	Spring (1)	0.234	-0.028	1.105
	Spring (2)	0.195	1.533	1.928
Feb. 1983	Manway 1	-0.838	-3.145	-1.533
	Manway 10	0.037*	-1.003*	0.279*
	Well 1	0.065	-0.357	0.838
	Well 3	0.470	2.319	2.518
	Spring	0.804	1.985	2.610
March 1983	Manway 1	-0.490	-2.036	-0.598
	Manway 10	-0.654	-2.566	-1.033
	Well 1	-0.110	-0.486	0.576
	Spring	-0.038	0.336	1.094
May 1983	Manway 2	-0.944	-3.182	-1.640
	Manway 10	0.464	-1.758	-0.434
	Well 1	-0.298	-0.554	0.369
	Spring	0.110	0.613	1.385
June 1983	Manway 2	0.468	-0.073	1.425
	Manway 10	0.519	-0.001	1.512
	Spring	0.008	0.348	1.171
Sept. 1983	Manway 2	0.160	-1.124	0.503
	Well 1	-0.614	-1.183	-0.253
	Spring	-0.415	-0.359	0.382

Table C.17. Calculated Saturation Indices.

Sample Date	Sample Location	Ferrihydrite $\text{Fe}(\text{OH})_3$	$\text{Fe}_2(\text{OH})_6$	Greenalite
March 1981	Influent	1.726*	2.554*	-0.522*
	Spring	1.189*	-1.532*	-8.941*
June 1981	Influent	-3.218*	-9.835*	-7.879*
	Spring	-0.220*	-6.313*	-14.516*
Jan. 1983	Manway 1	2.247	0.094	-11.079
	Well 1	1.478*	-3.447*	-16.317*
	Well 3	1.466	-2.232	-12.604
	Spring (1)	1.478	-3.529	-16.925
	Spring (2)	1.466*	-2.753*	-13.628
Feb. 1983	Manway 1	1.190	-0.635	-7.057
	Manway 10	1.143	-0.703	-6.950
	Well 1	0.864*	-3.761*	-13.943*
	Well 3	1.660	-1.817	-11.957
	Spring	1.478	-1.904	-11.670
March 1983	Manway 1	1.545	-0.567	-8.670
	Manway 10	1.939	0.139	-9.006
	Well 1	1.131	-3.349	-14.147
	Spring	1.177	-3.239	-13.696
May 1983	Manway 2	1.419	0.091	-6.091
	Manway 10	1.356	-2.428	-13.051
	Well 1	2.336	0.766	-8.739
	Spring	1.187	-2.840	-12.559
June 1983	Manway 2	0.729	-1.122	-5.368
	Manway 10	0.090	-2.426	-5.443
	Spring	1.183	-1.513	-8.520
Sept. 1983	Manway 2	1.851	2.056	-2.821
	Well 1	1.148	2.390	-11.022
	Spring	1.190	-3.467	-14.261

* $\frac{1}{2}$ detection limit