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GROUNDWATER MONITORING IN THE SAVANNAH RIVER PLANT LOW LEVEL WASTE BURIAL GROUND (U)

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SUMMARY

This document describes chemical mechanisms that may affect trace-level radionuclide migration through acidic sandy clay soils in a humid environment, and summarizes the extensive chemical and radiochemical analyses of the groundwater directly below the SRP Low-Level Waste (LLW) Burial Ground (643-G). Anomalies were identified in the chemistry of individual wells which appear to be related to small amounts of fission product activity that have reached the water table. The chemical properties which were statistically related to trace level transport of Cs-137 and Sr-90 were iron, potassium, sodium and calcium. Concentrations on the order of 100 ppm appear sufficient to affect nuclide migration.

Several complexation mechanisms for plutonium migration were investigated, but most of these were shown to be incapable of mobilizing more than trace quantities of plutonium. The parameters of greatest importance were oxidation - reduction potential, pH, dissolved organic carbon, phosphate and carbonate. Of these, organic and phosphate complexation had the greatest potential for mobilizing plutonium in the SRP groundwater. In the absence of such complexants, plutonium would be essentially immobile in the soil/water system of the SRP burial ground.

A thorough radiochemical analysis was also performed on all wells that had a history of alpha and/or non-volatile beta-gamma contamination, as determined by routine monitoring. Additional wells were selected for radioanalysis on the basis of a survey that included in-situ gamma scans of the monitoring wells.[1] Of the twenty wells that were tested, only two contained alpha or fission product activity above EPA standards for public water supplies. Across the burial ground, gross levels of activity in the groundwater have actually decreased through dilution and dispersion over the past five years.

In addition to decreases in gross activity, there is evidence that the rate of radionuclide migration has decreased over the past few years, especially in the older, eastern section of the SRP burial ground. This observation is attributed to reduced concentration, in the groundwater, of materials from the buried waste that could mobilize radionuclides. Transport velocities will gradually decrease as the trench leachate becomes more dilute. Plutonium-239, which is presently detectable only at trace levels, should follow this general trend, becoming fixed in the soil of the waste trenches.

INTRODUCTION

The LLW Burial Ground

The solid waste disposal area that will be discussed in this report is the 643-G Low Level Waste (LLW) Burial Ground. Located between the two Separations areas at the Savannah River Plant, this area received solid wastes from Plant and Laboratory facilities from 1954 through 1971.[2] Liquid wastes were not buried there, but some contaminated organic solvents were stored in tanks at the site, and large volumes of that solvent were burned in open pans in shallow pits during the first fifteen years of operation.[3]

Major sources of activity include both fission and activation products on discarded process equipment and reactor hardware. Plutonium-contaminated waste from various locations was also received routinely.

Waste materials were placed in categories for disposal with "low-level" (low beta-gamma or suspect alpha) waste going in some trenches, "high activity" (high gamma activity solids) waste in others. Plutonium or "alpha" waste was disposed of in separate trenches. After 1965 most of this category was buried retrievably in concrete containers and since 1974 all Pu waste >10 nCi/g has been stored on pads on the surface.[2]

Additional details of burial ground operations at SRP can be found in the Final Environmental Impact Statement.[4]

Groundwater Monitoring

The groundwater below the (643-G) Low Level Waste Burial Ground has been monitored for radionuclide contamination since 1956. Although that site stopped receiving waste in 1971, SRP continues to monitor the groundwater there for radionuclide transport.[4] There are now 63 accessable groundwater monitoring wells within the burial ground perimeter. These are laid out on a 200-foot grid with 20-foot well screens centered at the water table, approximately 40 feet below grade (Figures 1, 2). Since installation in 1973, only 12 of these wells have contained more than background levels of non-volatile beta-gamma or alpha emitters (Figure 3).[5-8]

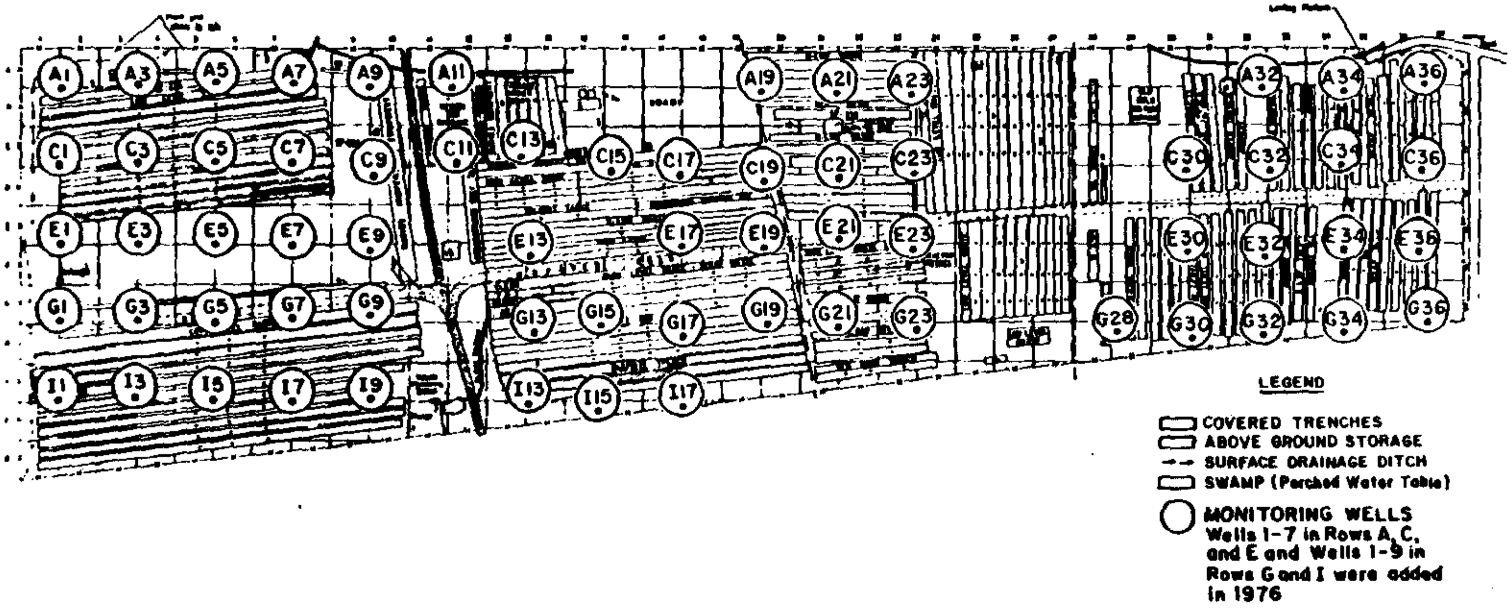
A thorough analysis of the groundwater monitoring wells was performed, from July, 1980 through July, 1982, to establish "baseline" chemical conditions, and to identify some of the mechanisms by which traces of radionuclides are mobilized in the subterranean environment. With that data, reported here, it will be possible to measure changes in water quality caused by the presence of waste material in the burial trenches. Radionuclide migration velocities will also be more predictable now that most of the water quality parameters which affect migration have been measured. Finally, the combination of better transport information and actual long-term radiochemical information will be useful for calibrating the existing radionuclide transport models.

DISCUSSION

Solubility, Ion Exchange and Complex Formation as Factors Affecting Migration

Radionuclide migration and the spread of conventional chemical pollutants are closely linked in a low-level waste burial ground. Many common chemicals, including simple salts and possibly even the degradation products of standard packaging materials, can affect the mobility of radionuclides.[9-15] These effects can be either positive or negative, and their overall impact on the efficiency of radionuclide containment at a low-level waste burial site is determined by the average of the effects in each isolated system (trench location) within the site. Vertical transport through the unsaturated zone to the groundwater is specifically defined for each trench by its contents. Lateral transport in the groundwater is then determined by the soil and water chemistry of the total system.

Figure 1 - Monitoring Wells in 643-G



Wells A-7 and A-9 were temporarily out of service and well I-3 was dry during the chemical analysis survey. Well E-15 (between E-13 and E-17) was drilled in 1977, and went dry in 1980.

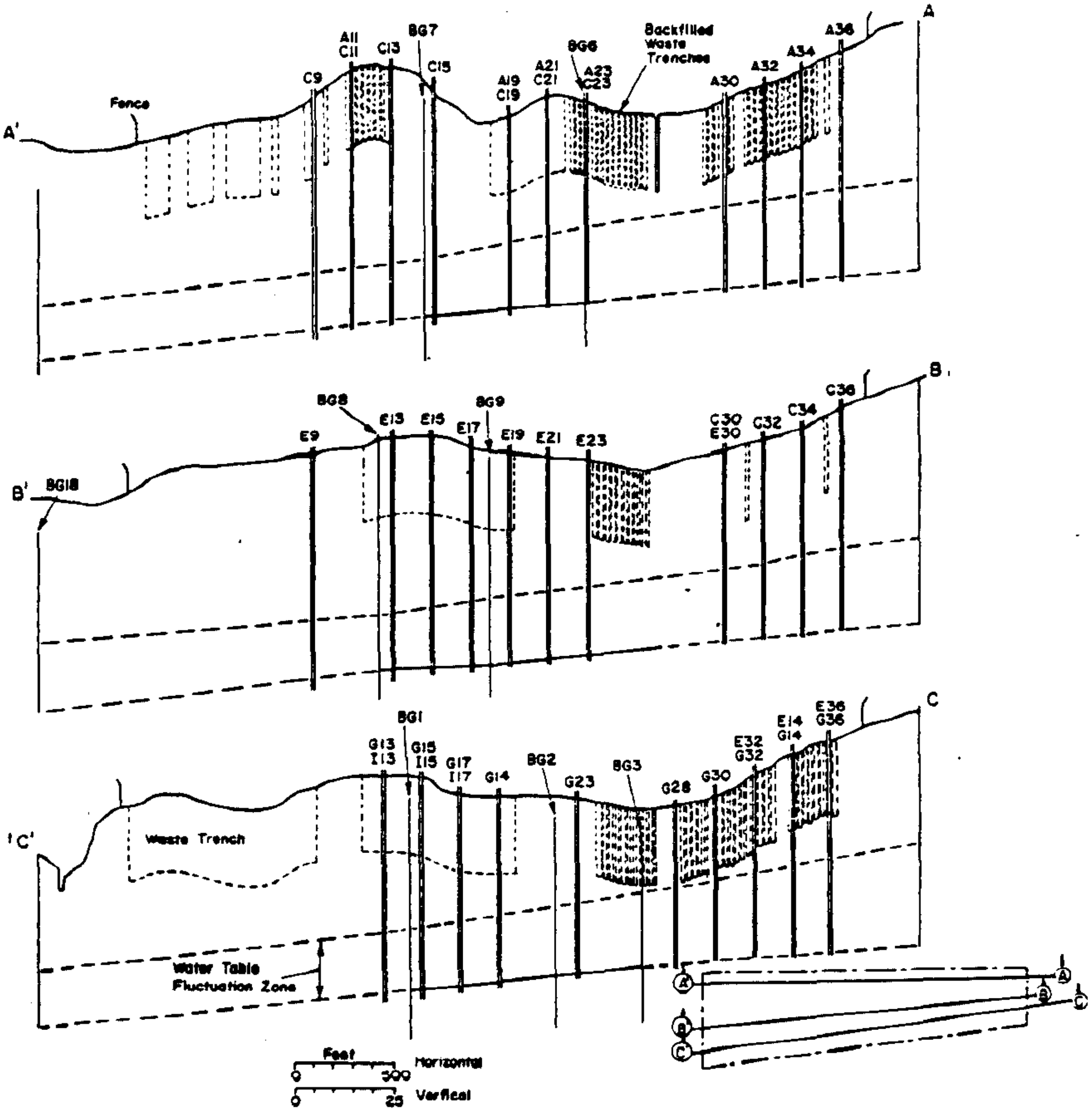


FIGURE 2 . Burial Ground Cross Sections Showing Details of Water Table Well Construction (Ref 8).

643-G Wells That Contain Measurable Radioactivity

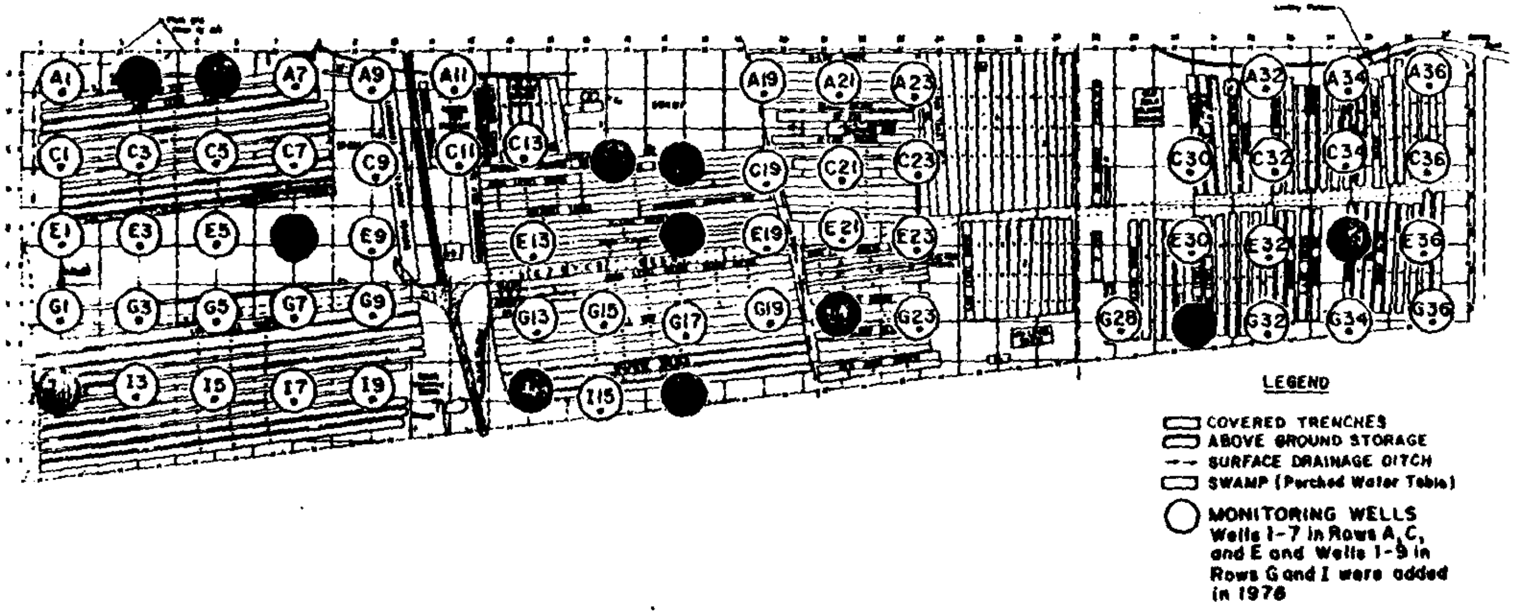


FIGURE 3. Filled in circles are wells where measured radioactivity has exceeded background in past routine monitoring surveys from 1973 to 1976. Tritium data is not included here (see Ref 8).

The solubility of a radionuclide is determined by the chemistry of the solution in which it is dissolved. The presence of certain counterions may tend to limit or enhance that solubility, depending on the nature of their interaction. For example, Sr-90 is virtually insoluble in alkaline carbonate.[15] On the other hand, although most plutonium compounds are normally extremely insoluble in solutions of low ionic strength, one of the most important factors in ultra trace level plutonium migration may be its tendency to form soluble carbonate complexes.[18] Chemical factors that can also affect the solubility of a radionuclide include the oxidation potential, pH, and ionic strength.

Although they are certainly related, the solubility of a radionuclide in the groundwater should not be equated with its mobility in the groundwater system. Many ions that are quite soluble move very little in the natural system because of their interaction with soil (e.g., cesium).[13,17,19] The most important of these interactions is ion exchange adsorption by the soil.

The efficiency of radionuclide adsorption and retention by soil depends on the soil cation exchange capacity, its structure, and on the water chemistry. For example, the same stable complexes that improve solubility also tend not to be adsorbed by ion exchange sites on soils.[9-11] Competitive ion exchange can also occur, where high concentrations of other salts "swamp out" radionuclides in the competition for ion exchange sites on the soil, thereby causing them to stay in solution and move with the groundwater flow. The type of ions in solution is also important, because some are more efficiently retained by soil, and therefore, compete more effectively for exchange sites.

Around a typical low-level waste burial ground, both the solubility and the ion exchange behavior of radionuclides are influenced by complexation. Anionic and soluble neutral complexes are sorbed very poorly by most soils.[11] This includes the strong chelation complexes formed by some organic materials (e. g., EDTA) as well as the more loosely associated complexes that are formed with some common anions (e.g. Cl^- and HPO_4^-) and many common organic species (e. g. organic esters, acids, ketones and aldehydes). Generally, the stronger the complex, the greater the mobility that will be imparted to the migrating radionuclide. Very weak complexes, and those which include multiple monovalent anions, usually affect ion mobility only at high ligand concentrations. This tends to limit the effects of such weak complexes to a region within or very near the trenches, making them relatively unimportant.

Anionic radionuclides (e.g. I-129, Tc-99) are inherently mobile and are not greatly affected by cation exchange or ligand complexation. However, the oxidation-reduction potential of the system can have an important impact on their migration velocity. For instance, the oxidized form of Tc-99 is mobile, but the reduced form is not.[20] Owing to the reactivity of I_2, just the opposite may be true for I-129. Often, the dissolved oxygen content of the water (DO) is the preferred indicator of the oxidation potential of natural water systems, because measured oxidation-reduction potentials tend to be erratic in dilute solutions.[21] However, direct measurement with a platinum electrode may still be the best indicator of an unusually oxidizing or reducing condition.

Possibly the most important parameter for predicting if mobilizing mechanisms will be operative is the pH of the solution. Most of the mechanisms, including complex formation, oxidation-reduction, and hydrolysis, are pH-dependent. Low pH (acidic) systems favor radionuclide mobility by improving the solubility of metals, by competing for ion exchange sites on the soil, and by dissolving potential adsorption sites. More alkaline conditions, in the absence of complexing anions, generally tend to immobilize radionuclides. Clay soils become less permeable, especially in the presence of sodium. In addition, the formation of metal hydroxides and carbonates provides more potential adsorption sites for cationic radionuclides, and the solubilities of many radionuclides are substantially reduced by hydrolysis. However, anionic carbonate and hydroxycarbonate species and other complexes may be formed under alkaline conditions, thus mobilizing radionuclides such as plutonium and Co-60.

Summarizing, the water quality parameters that have the greatest impact upon radionuclide migration include pH, the concentration of iron and other competitive cations, total organic carbon, oxidation-reduction potential, dissolved oxygen, conductivity, and complexing anions. The mechanisms most likely to enhance radionuclide mobility are non-exchangeable complex formation, and competitive cation exchange.

This information will be presented for each of the groundwater (GW) monitoring wells in the chemistry section of this report. The theoretical impact of the normal and extreme values for each parameter will then be discussed and compared to the radiochemical summary of the GW wells. The following section provides a brief account of pertinent burial ground operations history - relating events that are responsible for appearance of radiochemical contamination at the water table in two locations.

OPERATIONS HISTORY

"Normal" Wells Containing Incidental Contamination

In addition to long-term radionuclide transport from the waste disposal trenches, incidental contamination from burial ground operations is also detectable in some of the groundwater monitoring wells. This is why a few of the monitoring wells in the burial ground contained traces of plutonium, even though their chemistry was apparently normal. These can be divided into two categories, the "solvent spill wells" and the wells affected by equipment decontamination operations. These will be discussed separately, and will be referred to throughout this report.

Solvent Spill

In 1971, approximately 200 gallons of contaminated TBP-kerosene was accidentally released to the groundwater, when a dry monitoring well was mistaken for a solvent storage tank header. This accident took place near Well C-17, but when the solvent reached the groundwater it spread over a considerable distance.[22] A series of cores were taken to the depth of the water table to determine the extent of the contamination (see Figure 4).

Approximately 250,000 gallons of water were pumped from Well C-17 to measure the extent of the radioactivity there.[23] The water was discharged to the H-Area seepage basin. Small amounts of activity have remained in the area, and the contamination levels in the surrounding monitoring wells have tended to fluctuate with the height of the water table. This is believed to be due to an extremely disperse film of contaminated solvent that essentially floated on the water table. Some of that solvent probably remained on the soil when periods of drought caused the water table to recede. As the water table returns to higher levels in wet years, the groundwater tends to pick up material that was sorbed on soil when the water level declined.[5-8]

The wells that were originally affected by the solvent release were C-15, C-17, E-13, E-17, E-19, G-13, G-15, G-17, G-19, I-7, I-9, I-13, I-15, and I-17. So far, of the wells analyzed by low level alpha pulse height, C-17, E-13 and E-17 are below 1 pCi/L. Well E-17 still contains the fission and activation products Cs-137 and Co-60; but these are now at levels considerably less than the drinking water standards, which are currently 200 and 100 pCi/L, respectively.[24]

Future low-level analyses should be able to establish whether the plutonium activity in Well G-21 is from the same source (solvent spill) as the rest. However, preliminary isotope ratio

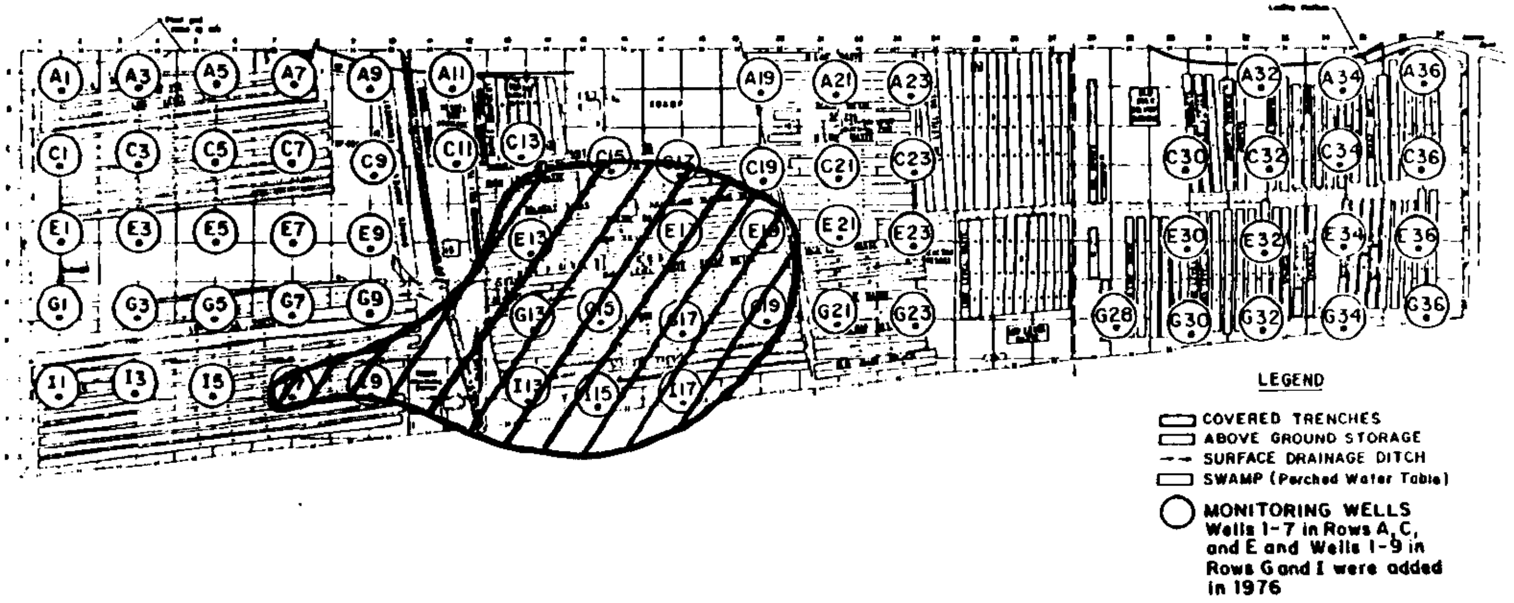


Figure 4. Area of 1971 contaminated solvent spill.

measurements indicate that it is not. The rate of dissipation of plutonium will also be of interest as the less contaminated wells are monitored.

Equipment Decontamination Area

The activity in Wells A-3, C-1 and C-3 may have been present before waste was even buried in that region. Early in the operations history of the 643-G burial ground, the northwest corner was used as a storage area for burial ground equipment.[25] A decontamination station was operated to the east of that storage area, between and just north of the A-3 and A-5 monitoring wells (see Figure 5) which were installed much later.

Complexing agents such as EDTA were apparently used in addition to phosphate detergents to remove contamination from the metal equipment. These agents have apparently mobilized traces of plutonium and Sr-90 to the extent that they are now detectable at the water table, which is forty feet below grade. The predicted contamination area drawn in Figure 5 is based on the water table contour of that area.

Usually it is possible to judge whether a single source is responsible for contamination at several locations by the reproducibility of the isotopic ratios. However, since many sources of activity were present in the form of different pieces of burial ground equipment, each could have a different isotopic ratio. The monitoring wells are not all the same distance from the decontamination station. They would, therefore, not be expected to have the same isotopic ratios of plutonium, because the source ratio would have varied with time. Furthermore, the decontamination area is probably too large compared to the migration path length to be considered a point source, so the material that came from one side would not necessarily be the same as material from the other.

A major point in support of this explanation is that there is little or no other plutonium in that area of the burial ground, regardless of the transport mechanisms involved. There are two trench wells along the A-Line that have been known to contain water. These (TW-7 and TW-9) are sampled at regular intervals whenever water is present, and gross alpha/non-volatile beta-gamma measurements are made. A low-level pulse height analysis was also performed on the closest trench well to A-3 (TW-9). Pu-238 was detectable at only 7 pCi/L, which is approximately the highest alpha concentration ever observed in TW-7 and 50% of the maximum observed in nine years of monitoring TW-9. If such low concentrations are found in the nearest trenches, it isn't likely that even higher levels could be migrating from them to the groundwater. (Furthermore, the burial ground records do not show any likely source of plutonium in that general area.)

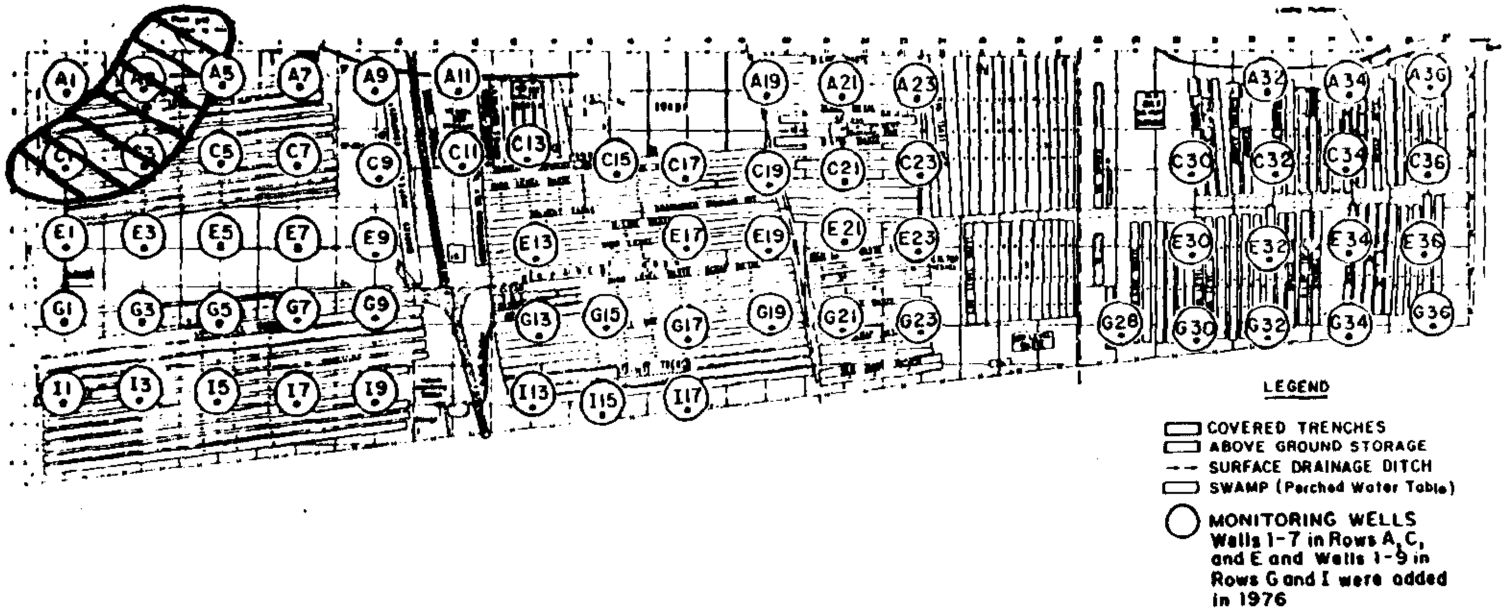


Figure 5. Groundwater affected by equipment decontamination solvents (postulated).

If the conclusions concerning the decontamination solvents are true, it should be possible to locate the old decontamination station and obtain a soil core in that area. To determine whether the contamination level in Wells A-3, C-1 and C-3 will go down or up in the near term, one would have to know more about the source. But since the complexants are probably all but gone now, the migration behavior of the radionuclides should be similar to what will occur elsewhere in the burial ground many years from now. The activity levels in the groundwater will decline.

RADIOCHEMICAL ANALYSES

Gross Alpha, Non-Volatile Beta-Gamma

Gross alpha and non-volatile beta-gamma measurements have been made of the 643-G grid wells on a bimonthly basis since they were first installed.[5-8] The 43 wells in the east and central sections have been in place since 1973. The wells in the western section (A1-9, C1-7, E1-7, G1-9, and I1-9) were added in 1976. Nominal background levels for these measurements with the equipment and methods used are 3 pCi/L gross alpha, and 50 pCi/L non-volatile beta-gamma. Wells that contained more than these levels of activity as of 1980 were considered contaminated, and samples from each were taken for Sr-90 determinations and low-level counting by gamma spectrometry and alpha pulse height. With the exception of Well E-15, which was temporarily inaccessable for sampling, the low-level analyses of these wells are listed in Table B1 (Appendix B) along with those of the wells chosen for analysis based on in-situ gamma spectrometric scans,[26] as described below.

Natural activities were low in the gross alpha - nonvolatile beta-gamma measurements since the samples were allowed to settle for at least 48 hours before aliquots were taken for analysis. This procedure tends to minimize the activity associated with particles suspended in solution in much the same manner as coarse filtration, but with less handling. However, there is normally an additional reduction of activity in some wells via coprecipitation of certain radioisotopes, as Fe(II) slowly oxidizes to Fe(III) and precipitates from many samples. There is evidence that this loss of activity occurs in samples from Well G-21, which contains roughly 110 ppm of iron. Routine non-volatile beta-gamma measurements detected 219 pCi/L of activity in 1980 (6 samples, bimonthly). However, 1600 pCi/L of strontium was detected in that well in the same year by more exact methods.[27]

In Situ Gamma Scan

In 1980-81, W. W. Bowman performed low-level, high resolution gamma scans on each of the monitoring wells in the 643-G burial ground.[1] This was accomplished by lowering an intrinsic lithium-germanium crystal detector into each well and recording the output as a function of depth. Early in the study, several wells were found with very small areas of Cs-137 and/or Co-60 contamination located below the depth of the trench bottoms.[26] After re-scanning these wells it was determined that these locations probably represented contamination that was transferred when the monitoring wells were drilled. The possible exception was Well A-3, which seemed to contain small amounts of Cs-137.

This study was used as additional input for the selection of wells that could be contaminated at very low levels. All of the wells that gave any indication of contamination at depth via the in-situ scans[26] were sampled and analyzed by the low-level methods described below. Low-level counting results are recorded in Table B1, Appendix B.

Low-Level High Resolution Counting

Methods and Sample Preparation

In spite of the apparent sensitivity of the gross alpha and non-volatile beta-gamma methods, low-level high-resolution counting is indispensable to the investigation of a low-level waste burial site. The counting and sample preparation methods that were applied to routine monitoring (gross alpha and non-volatile beta-gamma) at SRP were designed to handle large sample loads and to detect gross changes in activity. Variations in sensitivity caused by sample chemistry [e.g. coprecipitation of radionuclides with Fe(III)] were not considered, and all but the major differences in the natural activity background were necessarily ignored. The resolution of decay energy was also extremely poor. As a result, significant qualitative information was lacking.

High-resolution low-level counting is the only good way to discern between natural and anthropogenic radioactivity. It is also the only way to obtain information about specific radionuclides and gain insight into the hazard potential of the system. And happily, it is usually the only way that radionuclides from SRP low-level waste can be detected, even in the groundwater directly beneath the disposal site 10-25 years after disposal operations have ceased.

As mentioned previously, low-level counting was performed on all of the wells that gave any indication of radioactive contamination in the normal monitoring and high-resolution gamma spectrometric surveys. The methods used included high-resolution gamma spectrometry at SRL's Low-Level Counting Facility. Alpha pulse-height was also performed on each sample following plutonium extraction and electroplating. The detectability of Cs-137 and

Co-60 was roughly 8 pCi/L, while the detection limit for Pu-238 and Pu-239 was less than 1 pCi/L.

Sr-90 analysis was performed on 2-liter samples by concentrating the water and using the appropriate extraction and counting procedures to measure Y-90 ingrowth.[27] The detection limit of the method was 6 pCi/L of Sr-90, and recovery was high (90%).

Sample preparation procedures for low-level gamma spectrometry and alpha pulse height analysis were designed to obtain the maximum sensitivity for activity in the original water sample. With this in mind, samples were acidified to pH 2 prior to filtration with a glass membrane filter, thus preventing adsorption of radionuclides on the sedimentary particles or the filtration apparatus*. This procedure had the ancillary effect of greatly increasing the natural radioactivity that was detected in these samples.

Natural Radioactivity

The coastal plain sediments that make up the lithology of the burial ground are naturally high in U-238 and Th-232 in the form of monazite. The daughters of these isotopes make up most of the radioactivity that can be detected in the groundwater monitoring wells. Acidification of water samples prior to filtering causes many of the daughters (e. g. Bi-214, Pb-212, Ra-226) to leach from the solids that are suspended in solution during sampling, thus increasing the apparent natural activity.

In some wells the routine nonvolatile beta-gamma measurements of the monitoring wells are low compared to the natural activity that is released from just a small amount of suspended clay following acidification. For example, in a sample from Well A-3, approximately 7 mg of suspended solids contained over 300 pCi/L of Pb-212 that was leached at pH 2. Because the nonvolatile beta-gamma method selectively removes the activity entrained on suspended solids, it is not detected, and the beta-gamma activity measured by that method in Well A-3 was only 66 pCi/L. Several of the monitoring wells in the burial ground contained Bi-214 and Pb-212. Filtered and acidified water samples ran as high as 629 ± 148 pCi/L Bi-214 (Well I-13) and 346 ± 100 pCi/L Pb-212 (Well A-1).

Five control wells that were nearby, but completely outside of the burial ground and away from its influence, averaged over 200

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* Please note that, as stated in Appendix A, all other samples that required filtration and acidification prior to analysis (e. g., ICP and AA) were first filtered with a Gelman 0.45 micron membrane filter and then acidified with ultrapure (Ultrex) nitric acid.

pCi/L of Bi-214. Samples were acidified and then filtered. Samples that were neither filtered nor acidified averaged slightly higher, but with less consistent results. Pb-212 was also detectable, and two of the five control wells contained over 300 pCi/L of this Th-232 daughter.

Results

The resulting data (Table B1) confirm Bowman's final conclusion[1] that none of the groundwater monitoring wells are contaminated by gamma emitters at significant concentrations. In fact, Cs-137 was identified in only three of the wells tested, and Co-60 in only one. In all cases, the gamma activities were less than 10% of the proposed drinking water standards, which are based on a calculated dose of 4 mrem per person-year.[24]

The data also show that the alpha emitters Pu-239 and Pu-238 are present at extremely low concentrations in some of the monitoring wells. Only one sample exceeded the drinking water standard of 5 pCi/L for Pu-239. That was an unfiltered sample obtained from Well G-21, which yielded an activity of 10 pCi/L. A duplicate sample that was filtered and then acidified was 4.0 ± 1.0 pCi/L. Two wells, A-3 and G-21 were above the 5 pCi/L drinking water standard for Pu-238, with 11.0 ± 3.0 and 17.0 ± 3.0 pCi/L respectively.

Many of the wells that once contained plutonium, as the result of a TBP-kerosene solvent spill, are now virtually clean. The slope of the gross alpha activity trends over the past five years are decidedly negative (Table B1, Appendix B). Three wells were identified as containing Sr-90 above the 8 pCi/L (4 mrem/person-year) drinking water standard. These were A-3, G-21 and I-13. Well G-21 contained 1600 pCi/L of Sr-90. This is over two orders of magnitude more than any other monitoring well in the burial ground.

CHEMICAL ANALYSIS OF 643-G GROUNDWATER

Based on the statistical treatment described below, and backed up by visual inspection of computer-generated histograms of the data, a group of wells was identified which appeared to have been chemically contaminated by the waste trenches (Table E1, Appendix E). The potential influence of the contaminants on the migration behavior of specific radionuclides was evaluated first. The list of wells with high potential radionuclide mobility was then compared to the radioactive contaminant profiles obtained by low-level counting to identify locations where waste chemistry has enhanced radionuclide migration. (Table E1 and E2, Appendix E.)

Monitoring Well Data Treatment

The groundwater monitoring wells were analyzed for seven different cations, five anions, silica, dissolved oxygen, conductivity, pH, total inorganic carbon (TIC) and total organic carbon (TOC). While some of the early data was obtained in 1980, and some as recently as July, 1982, the bulk of the analyses were performed on samples obtained from March to July of 1981. The values recorded in Table C1 (Appendix C) represent the average of all data taken for each well.

Over 1100 water quality parameters have been measured and recorded so far. Of these, roughly 350 (32%) were performed at least two times on samples taken from the wells 3 or more months apart. Some of the wells were analyzed for the same parameter three or more times, making it possible to calculate coefficients of variation for samples taken and analyzed at different times of the year from the same well (Table C2). The relative importance a change in any of the parameters can be estimated with some degree of confidence on the basis of this statistical information.

Although statistical treatment of the well analysis data is required to evaluate the apparent trends, it should be remembered that the burial ground is not a random system. Each location in the burial ground is classified according to the kind of material placed there, and the migration mechanisms are likely to be different for each radionuclide in each type of waste. Therefore, the correlation of statistical outliers (the "abnormal" values for each parameter) with radionuclide contamination in the groundwater is more important than the statistical correlation of a given parameter to a level of contamination. In fact, since radionuclide contamination was generally too low to be measured, the latter approach was not even attempted.

The water quality parameters for each well were averaged and descriptive statistics were generated for each variable (Tables D1-D22, Appendix D). Most of the parameters of the burial ground well samples were not normally distributed. The Kolomogrov-Smirnov test of normality was applied to all of the values for each variable. The null hypothesis in this test is that the data is distributed normally. The test statistic, D, represents the greatest difference between the cumulative distribution of the data and the calculated cumulative distribution of an ideal set (standardized normal sample) with the same mean and standard deviation. With the exceptions of conductivity, pH, nitrate, barium, and strontium, the probability of D exceeding the calculated value was less than 1%, a clear indication (P=0.99) that the data were not normally distributed.

The data were also generally skewed toward values indicating the influence of the waste trenches, as evidenced by the sign and magnitude of their coefficients of skewness and kurtosis (Tables D4-D22). This is consistent with the observation that the groundwater chemistry in the 643-G burial ground is moderately influenced by the waste in the trenches above it.

Wells that were definitely influenced by the waste in the burial ground trenches were identified as follows. The distribution of the data was calculated separately for each variable (Table D1(a), or chemical parameter. Each of the wells were then evaluated separately for each variable. If a measured parameter for a given well was more than three standard deviations from the mean for all wells, it was considered anomalous; such wells were assumed to have been contaminated by waste leachate from the trenches above (or upstream of) them. After the anomalous wells were identified, a new distribution was calculated excluding the anomalous values (Table D1(b)). The remaining data was then examined a second time, and any additional outliers were identified in the same way.

The anomalous wells with enhanced potential for radionuclide mobility were identified as follows. Values ranging above the 95th percentile for ionic concentrations, TOC, and conductivity were chosen from the list of extreme values listed for each parameter. Values outside the 5-95th percentile pH range were selected, and values below the 5th percentile were selected from the DO and mV data. These wells are listed in Tables E1 and are discussed in Appendix E.

General Parameters

pH

The hydronium ion activity, as represented by the negative logarithm of that quantity (pH), is one of the most important factors in radionuclide migration. The solubility of many salts, the exchange capacity of the soil, and the prevalence of complexes are all dependent upon pH. The pH of the groundwater below 643-G is slightly acidic (averaging 5.3). This favors radionuclide solubility, while decreasing the potential for anionic complex formation. The effect of pH on fluoride, phosphate and carbonate complex formation is shown graphically in Figures 6-8, where the fraction of each of those potentially complexing anions that is available as the free ligand is plotted as a function of pH.

With the possible exception of plutonium, only I-129 in the form of silver iodide is immobilized primarily by its lack of solubility. In some cases, it may actually be advantageous that the SRP Low-Level Waste Burial Ground is in an acidic environment,

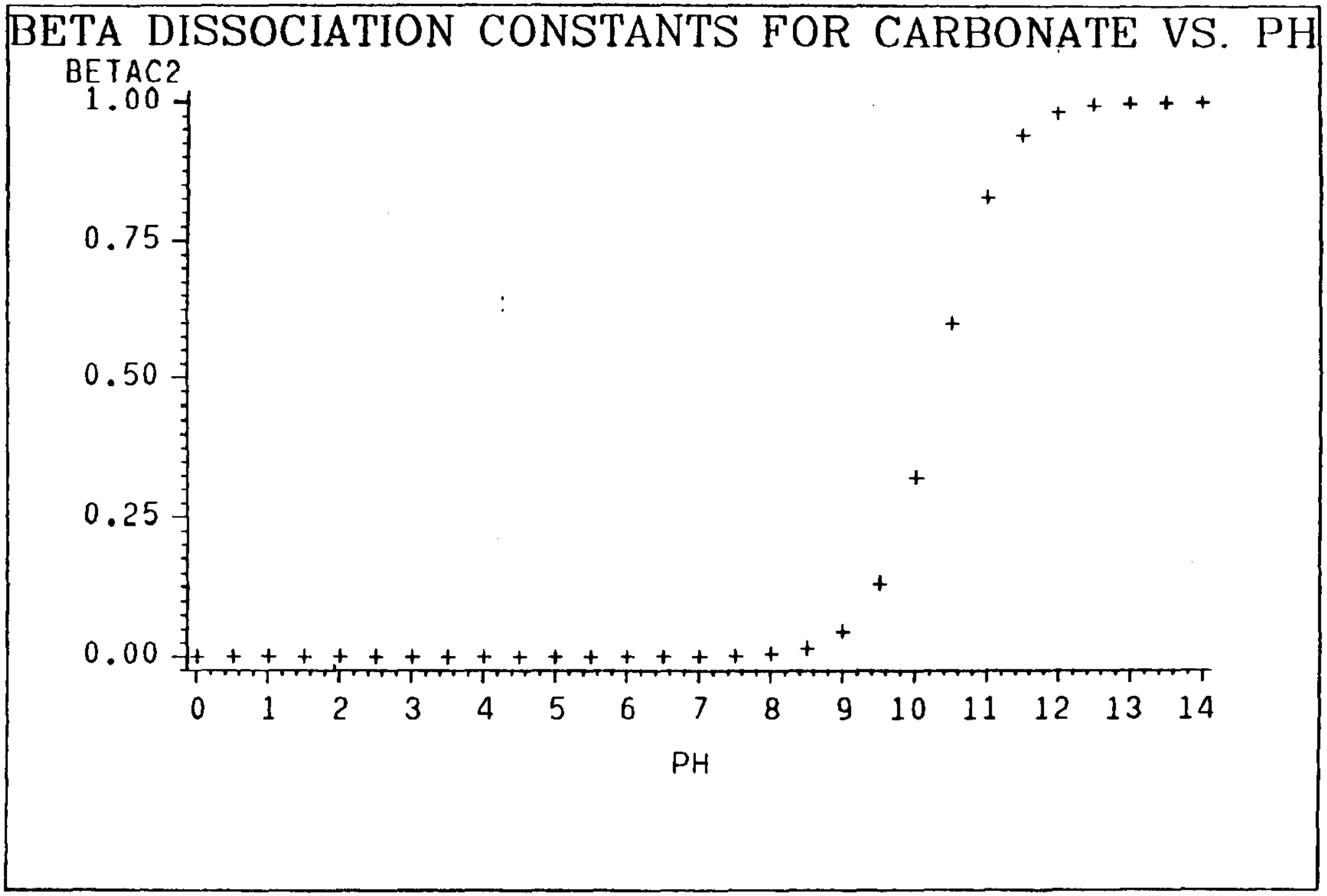

Figure 6. BETAC2 is the fraction of free carbonate available for precipitation - complexation reactions in a solution containing bicarbonate.

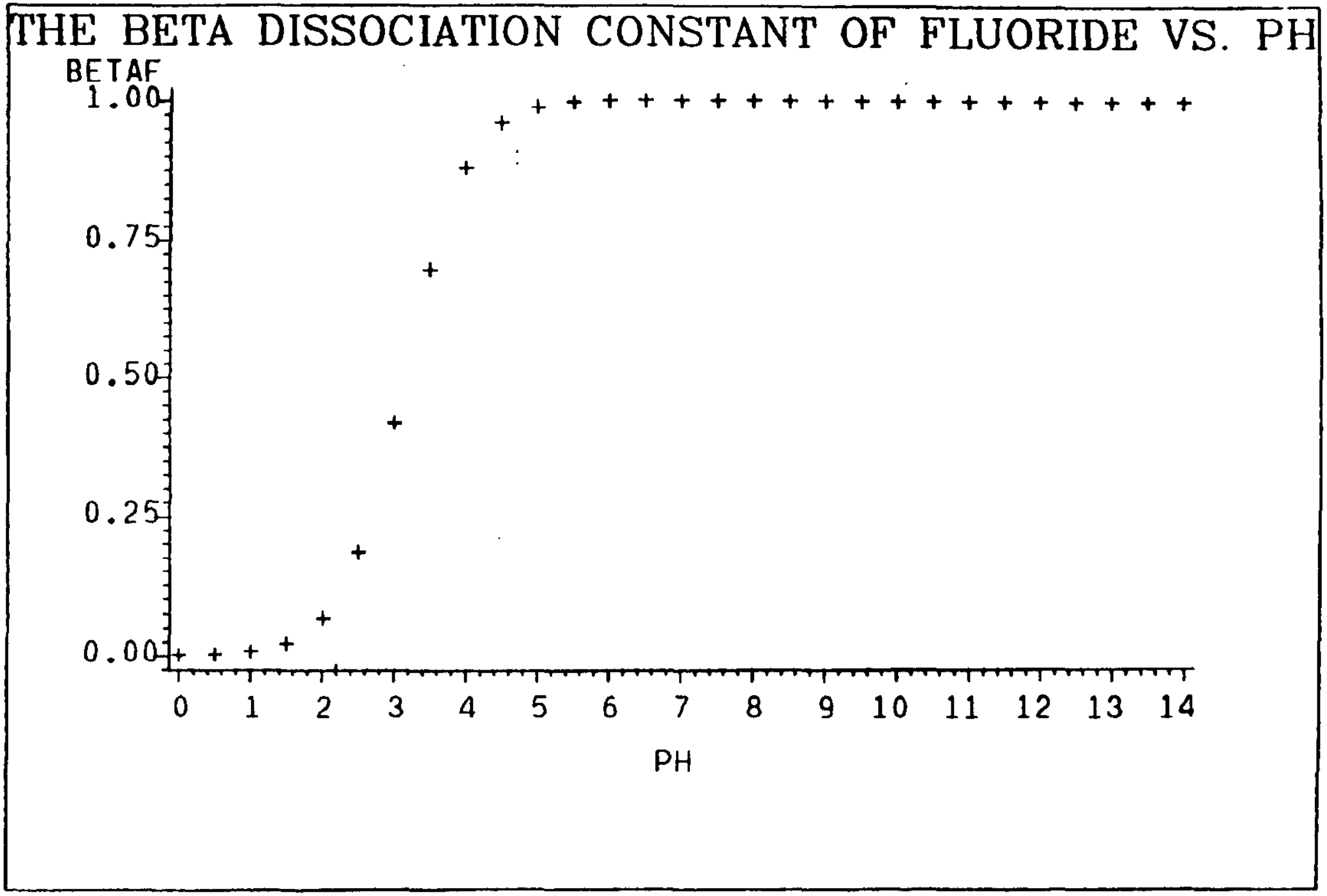

Figure 7. BETAF is the fraction of fluoride available for metal complexation in a solution containing hydrogen fluoride.

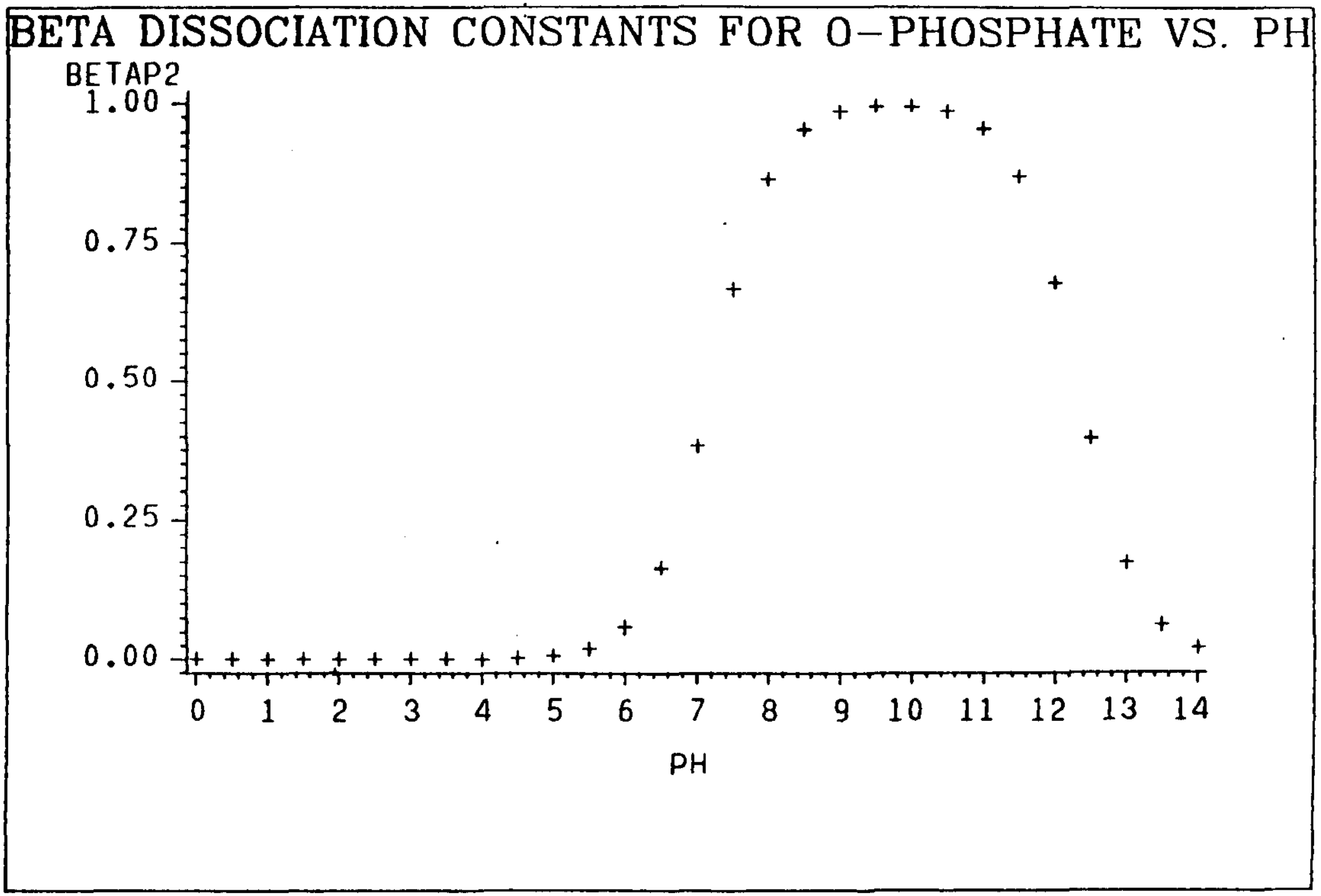

Figure 8. BETAP2 is the fraction of phosphate present as $HPO_4^=$ in a solution containing phosphoric acid or phosphate.

because the acid reduces the formation of mobile radioactive complexes. Acidity also favors the reduction of plutonium to the Pu(III) and Pu(IV) states,[28] which have a greater affinity for soil than oxidized plutonium.[13]

One adverse affect of an acidic environment is a diminished soil (ion) exchange capacity.[29] The pH of the groundwater was normally above the isoelectric point (IEP) expected for typical SRP soil. The soil surface potential (I_o) is determined by the equation:

$$I_o = \frac{RT}{F} \ln \frac{[H^+]_o}{[H^+]}$$

where

R = gas constant
F = Faraday's constant
$[H^+]_o$ = Hydronium ion concentration at the Isoelectric Point (IEP).
$[H^+]$ = Hydronium ion concentration
T = temperature in °K.

and at 25°C

$$I_o = 0.059 \text{ (pH - IEP)}$$

As expressed here, positive values of I_o favor cation exchange. Therefore, the cation exchange capacity of the soil tends to increase with pH. This has been proven experimentally at SRP where the cation exchange capacity may increase by a factor of 100 from pH 4 to pH 10.[13,30] Kaolinite clay, the dominant clay mineral in SRP soil, has an IEP of 3.0-4.6.[29,31]

Only two wells were more alkaline than pH 7.0. One well, E-17, was at pH 10, indicating that a major source of chemical contamination must be leaching from the trenches nearby.* There are several potential effects of such contamination. However, the undesirable effects are essentially only two, the mobilization of radionuclides due to competitive ion exchange, and the formation of mobile radioactive complexes. The first effect, competition for ion exchange sites, is more closely related to ionic strength and the effects of specific cations. The formation of mobile inorganic

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* This well was analyzed one year later, (June 1982). The pH had dropped to 7.00 and phosphorus had decreased to less than 0.1 ppm.

complexes is discussed in the sections on phosphate, fluoride and carbonate.

Oxidation - Reduction Potential (E_H)

The measurement of oxidation-reduction potential in the groundwater monitoring wells is designed to identify excessively reducing environments. Although highly oxidizing environments are possible, the presence of reduced carbon (eg. cardboard, wood, cloth, rubber, etc.) in the trenches gives most of them relatively low oxidation potentials instead, expecially in the presence of biological (bacterial) activity. The oxidation-reduction potential measurement is most meaningful for reduced, non-oxygenated systems, because once at equilibrium, oxygenated systems would not contain enough of the lower oxidation state species of the oxidation-reduction couples to measure with this simple technique.

The oxidation-reduction potential is determined by the potential of one or more couples in solution, but it is not usually measurable with any degree of certainty about the nature of the couple actually being measured. This can have serious drawbacks in the interpretation of E_H data for dilute solutions, or in solutions where equilibrium has not been reached among the major sample constituents. Both conditions are prevalent in groundwater systems. The contaminants in the monitoring wells are generally very dilute, and the chromatographic effects of soil adsorption virtually preclude equilibrium in waste-affected sampling locations. The meaning of the measured potential is therefore subject to question.

Possibly the only measureable redox couple of consequence in the groundwater is that of Fe(II)-Fe(III). Approximately 10^{-5} M (ie; 0.5 ppm) concentrations of Fe(II) and Fe(III) are required for an exchange current of a microampere at the platinium electrode.[21] This is approximately the point at which the electrode can be presumed to be following only the iron couple, successfully rejecting mixed equilibrium potentials. Mn(III)-Mn(IV) could also be considered if the concentration of manganese in the groundwater were sufficiently high.

Although it is generally inferred that a relationship exists between the dissolved oxygen concentration (DO) and the oxidation-reduction potential (E_H), the correlation can be very poor. Under the dynamic conditions present in the burial ground, the oxygen in the groundwater has insufficient time to react completely with reduced components of the waste. Waste components also lack the time to react with each other, so equilibrium is not complete. However, the poor correlation can probably best be explained by the fact that most of the wells have no dominant oxidation-reduction couple.

The three wells that contained more than 1 ppm of iron all had low dissolved oxygen concentrations and low redox potentials, but the Eh of the two wells that contained iron at 0.5-0.8 ppm appeared to be more closely related to the pH and nitrate couple than to dissolved oxygen. This is plausible, since the reduction of nitrate is dependent on pH, but not necessarily on oxygen concentration (for a system not at equilibrium). However, the kinetics of nitrate reduction indicate that it is still the iron couple that is actually being measured.[32]

The oxidation reduction potential of the groundwater was measured as high as 0.683 volts versus the standard (or normal) hydrogen electrode (Well C-13). But even this was too low to allow a significant amount of plutonium to exist in the Pu(VI) or Pu(IV) states. Except in strong complexes, trace levels of plutonium should be either as Pu(III) or Pu(V), depending on the oxidation potential and pH of the system.[28,33-35] The potentials required to convert these to Pu(IV) and Pu(VI) are given below.[28]

	E° (volts)
$PuO_2^{2+} + e^- = PuO_2^+$	+0.93
$Pu^{4+} + e^- = Pu^{3+}$	+0.97

Unlike the equilibria described above, the conversion of Pu(V) to Pu(III) and Pu(VI) to Pu(IV) are highly pH-dependent.

	E° (volts)
$PuO_2^+ + 4H^+ + 2e^- = Pu^{3+} + 2H_2O$	1.06
$PuO_2^{2+} + 4H + 2e^- = Pu^{4+} + 2H_2O$	1.03

The next equation can be used to establish the equilibrium constant at various solution potentials (E_H) of the groundwater. At equilibrium $E = E_H$.

$$E = 1.06 + \frac{0.059}{2} \log \frac{[PuO_2^+]}{[Pu^{3+}]} + 0.12 \text{ pH}$$

Inspection of this equation reveals that a one pH unit change in acidity affects the ratio of Pu(V) to Pu(III) by a factor of 10^4, with greater acidity favoring the lower oxidation state. A change in the solution potential of 100 millivolts has slightly less effect, altering the ratio by a factor of $10^{3.3}$.

The relationship between Pu(VI) and Pu(IV) would be the same way, but for the dominance of the Pu(V) and Pu(III) forms that exist at the oxidation potential of most natural systems.

As a result of these equilibria, the most favorable conditions for retaining plutonium on the soil in a complex system is to maintain a moderate oxidation potential under slightly acidic conditions. This will stabilize Pu(III), which is the least mobile of all the plutonium species.

Dissolved Oxygen (DO)

Dissolved oxygen is measured in parts per million at the temperature (TEMP) listed for each well. This parameter is related to radionuclide mobility in two ways. It is a measure of the reducing character of the water, in that low dissolved oxygen concentrations result from oxygen consumption in a reducing environment at or upstream of the sampling location. It is also an indicator of the relative isolation of the sampling location from the flow of oxygenated water, giving some indication of the relative dominance of the waste chemistry.

High dissolved oxygen concentrations in the waste trenches and in the groundwater around such trenches are generally good in terms of radionuclide isolation, but technicium, ruthenium, and plutonium are much more mobile in their most oxidized states. The major drawback to highly reducing environments is the fact that Fe(II) often becomes dominant in such systems, eliminating or attenuating the affinity of the soil (metal-oxide layer) for radionuclides such as Sr-90. The higher ionic strength of such aqueous systems also uses up the ion exchange capacity of the surrounding soil, further mobilizing the cationic radionuclides.

Other contamination problems are encountered in highly oxygenated systems. Ruthenium VI and VII are anionic, but they are also unstable at the slightly acidic pH of the burial ground system, even in oxygen-saturated systems.[28] Nitrosyl complexes of Ruthenium II and III are probably the main component of mobile Ru-106 at SRP. Technetium is mobile only as the pertechnetate (TcO_4-) anion,[20] which is its normal oxidation state (VII) in groundwater. Reducing conditions in the trenches could immobilize Tc-99 temporarily, but its long half-life makes its eventual escape to the groundwater certain. Fortunately, low specific acivity, low decay energy, and short biological half-life combine to make Tc-99 relatively harmless. Plutonium is most stable as Pu(III) (See Fluoride) in aqueous environmental systems, although Pu IV, V, and possibly even VI are also present. Plutonium speciation is not directly affected by variations the dissolved oxygen concentration, but low dissolved oxygen concentrations may indicate the proximity of a reducing system that could also react with Pu(VI) and Pu(IV).

Conductivity (COND)

Ionic strength is approximately linearly related to conductivity. Both parameters are thus related to the activity coefficients of the radionuclides in a given system. For the most part the ionic strength of the well water samples was so low that the activity coefficient could be considered unity. Activity coefficients were calculated (Debye-Huckel) for Cs^+ and Sr^{2+} in the normal and higher ionic strength groundwater, but the differences over the observed range of ionic strengths were negligible.

The most important aspect of conductivity (and ionic strength), then, is as a measure of the ion exchangeable material in solution that could compete with radionuclides for adsorption (exchange) sites on the soil. More specific information about the wells will be given under the heading "Competing Cations." However, electrically conductive groundwater generally has the greatest potential for transporting exchangeable, cationic radionuclides (see Sodium). Therefore, the most conductive wells (G-7, G-21, I-13 and possibly A-5) would be suspected of containing dissolved radionuclides.

Complexing Anions

Total Inorganic Carbon (TIC)

Inorganic carbon (carbonate) has a strong impact upon the properties of groundwater. As dissolved carbon dioxide (carbonic acid) it tends to reduce the pH of natural groundwaters. In the form of carbonate and bicarbonate salts, it contributes to the alkalinity of the groundwater and makes up much of the buffering capacity of natural systems. The carbonate anion can form an insoluble salt with strontium, but under certain conditions it can also form mobile, anionic complexes with plutonium and uranium. At SRP, the latter effect is more important.

The fraction of the total carbonate in a given solution that is actually present as the free, divalent anion is referred to in this paper as BETAC2. The fraction is constant at a given pH. The dependence of BETA upon pH is illustrated in Figure 6. It can be seen from this figure that only a small fraction of the carbonate remains freely available at the pH of the normal groundwater at SRP.

The equations that apply to the dissociation of carbonic acid are:[28,36]

Equation	Dissociation Constant
[1] $H_2CO_3 = H^+ + HCO_3^-$	4.45×10^{-7}
[2] $HCO_3^- = H^+ + CO_3^=$	4.7×10^{-11}

The free carbonate fraction is BETAC2

$$[3] \quad \text{BETAC2} = \frac{[CO_3^=]}{[H_2CO_3] + [HCO_3^-] + [CO_3^=]}$$

$$[4] \quad \text{BETAC2} = \frac{1}{[H^+]^2/k_1k_2 + [H^+]/k_2 + 1}$$

where k_1 and k_2 are the dissociation constants of equations 1 and 2 respectively.

The formation of insoluble strontium carbonate is described by the solubility product equation,

$$[5] \quad SrCO_3 = Sr^{2+} + CO_3^{2-} \qquad ksp = 5.62 \times 10^{-10}$$

To calculate the solubility of strontium, the molar concentration of carbonate is first calculated from TIC (in ppm of carbon) as follows:

$$[6] \quad [CO_3^{2-}] = \frac{\text{TIC} \times \text{BETAC2}}{12{,}000}$$

The solubility of strontium is then:

$$[7] \quad [Sr^{2+}] = \frac{5.62 \times 10^{-10}}{[CO_3^{2-}]}$$

It can be shown through the application of these basic equations that none of the monitoring wells are close to being saturated with strontium. The application of similar equations for calcium[28]

[8] $$[Ca^{2+}] = \frac{7.08 \times 10^{-4}}{[CO_3^{2-}]}$$ (ksp of amorphous $CaCO_3 = 7.08 \times 10^{-4}$)

results in the same conclusion, that subsaturation conditions exist in all of the wells. Neither calcium nor strontium is present as the carbonate salt, so Sr-90 cannot be immobilized as an inclusion. At SRP, the (relatively low concentrations of) calcium and strontium in the groundwater merely serve to compete with ion exchange sites on the soil, while the carbonate serves as a counterion for their dissolution. This equilibrium is shifted somewhat for calcite, which has a ksp of 4.5×10^{-9}. Only well C-5 contains calcium at saturation levels with respect to this mineral (100 ppm Ca in 1.8×10^{-6}M free carbonate). Well E-19 is the only other well above 20% saturation in $CaCO_3$, being 92.6% saturated.

Plutonium (VI) and uranium (VI) both form anionic complexes with carbonate. The related equations are[28]:

		log k
[9]	$Pu_{(S)} = Pu^{4+} + 4e^-$	83.38
[10]	$PuO_2^{2+} + 4H^+ + 2e^- = Pu^{4+} + H_2O$	34.86
[11]	$PuO_2(CO_3)^o + 4H^+ + 2e^- = Pu^{4+} + CO_3^{2-} + 2H_2O$	22.86
[12]	$PuO_2(CO_3)_2^{2-} + 4H^+ + 2e^- = Pu^{4+} + 2CO_3^{2-} + 2H_2O$	19.94
[13]	$PuO_2(CO_3)OH^- + 5H^+ + 2e^- = Pu^{4+} + CO_3^{2-} + 3H_2O$	25.03
[14]	$PuO_2CO_3(OH)_2^{2-} + 6H^+ + 2e^- = Pu^{4+} + CO_3^{2-} + 4H_2O$	39.83

Combining these equations gives the following relationships for Pu(VI).

		log k
[15]	$PuO_2^{2+} + CO_3^{2-} = PuO_2CO_3$	12.00
[16]	$PuO_2^{2+} + 2CO_3^{2-} = PuO_2(CO_3)_2^{2-}$	14.92
[17]	$PuO_2^{2+} + CO_3^{2-} + H_2O = PuO_2CO_3OH^- + H^+$	9.83

[18] $PuO_2^{2+} + CO_3^{2-} + 2H_2O = PuO_2CO_3(OH)_2^{2-} + 2H^+$ -4.93

The ratio of the concentrations of the major anionic carbonate species with respect to the concentration of free (cationic) Pu(VI) is then given by:

[19]
$$\frac{[PuO_2(CO_3)_2^{2-}]}{[PuO_2^{2+}]} = [CO_3^{2-}]^2 \times 10^{14.92}$$

[20]
$$\frac{[PuO_2(CO_3)OH^-]}{[PuO_2^{2+}]} = \frac{[CO_3^{2-}]}{[H^+]} \times 10^{9.83}$$

Where each of the quantities in brackets represents the molar concentration of that species, and the carbonate concentration is calculated as before (i.e., $[CO_3^{2-}] = TIC/12{,}000 \times BETAC2$).

These equations were applied to each of the water compositions found in the monitoring wells. The results of these calculations showed that the anionic plutonium hydroxycarbonate species was often favored over both the cationic Pu(VI) and the neutral PuO_2CO_3 species by several orders of magnitude. The $PuO_2(CO_3)_2^{2-}$complex was not favored with respect to neutral PuO_2CO_3 (see Table I).

The thermodynamic dominance of the anionic hydroxycarbonate species may be an additional factor in explaining the trace level mobility of plutonium in the northwest corner of the 643-G burial ground (wells A-3, C-1, and C-3). However, there is still a question concerning the stability of Pu(VI) in the environment[37, 38, 39] (see also Oxidation-Reduction Potential). Its reduction potential is so high that it is certainly reduced to Pu(V), (IV), and (III) in most trench burial regimes. These do not form anionic carbonate (or fluoride) complexes in the environment and are, therefore, adsorbable by the soil. Still, the existence of a mobile complexed Pu(VI) species could help to explain the trace levels of plutonium that were detected in the groundwater. If so, some enrichment of Pu(VI) would have occurred through selective adsorption and retention of the reduced plutonium species along the path of migration.

Probably the best way to estimate the potential of a system for the formation of plutonium hydroxycarbonate is to consider an equilibrium between that species and the anionic hydroxide complex of Pu(IV).

TABLE 1

ANION/CATION SPECIATION RATIOS FOR PU (VI) WITH CARBONATE

CASE	pH	$[CO_3]_T$ (moles/liter)	$BETACO_3$	$\frac{PuO_2(CO_3)OH^-}{PuO_2^{2+}}$	$\frac{PuO_2(CO_3)_2^{2-}}{PuO_2^{2+}}$
MEDIAN	5.3	$1.1x10^{-3}$	$9.1x10^{-6}$	$1.4x10^{7}$	$8.3x10^{-2}$
WORST REAL	6.86	$5.3x10^{-3}$	$3.4x10^{-4}$	$8.8x10^{10}$	$2.7x10^{3}$
HIGH CARBONATE	5.3	$1.0x10^{-2}$	$9.1x10^{-6}$	$1.2x10^{8}$	6.9
HIGH	10.0	$1.1x10^{-3}$	0.32	$2.4x10^{16}$	$1.0x10^{8}$

Written as a reduction:

	log k
[21] $PuO_2(CO_3)OH^- + 2H_2O + 2e^- = Pu(OH)_5^- + CO_3^{2-}$	10.08

(Derived using equations 22 and 23
from Benson and Teague[28])

[22] $PuO_2(CO_3)OH^- + 5H^+ + 2e^- = Pu^{4+} + CO_3^{2-} + 3H_2O$	25.03
[23] $Pu(OH)_5^- + 5H^+ = Pu^{4+} + 5H_2O$	14.95

The reduction potential for this cell (E°) is derived from the Gibbs Free Energy relationship

[24] $G° = nFE° = - RT \ln K_1$

[25] $E° = \frac{0.059}{2} \log K_1$

Substituting 10.08 for log K_2

$E° = 0.296$ V

the equilibrium is then defined by the Nernst equation

[26] $$E = 0.296 + \frac{0.59}{2} \log \frac{[PuO_2(CO_3)OH^-]}{[Pu(OH)_5^-]\ [CO_3^{2-}]}$$

A worst case can then be calculated for the source, assuming that the free carbonate and Pu(IV) concentrations can be calculated. These are estimated to be 10^{-7} and 10^{-8} molar, respectively as an approximation to the worst case.

The amphoteric behavior of $Pu(OH)_4$ as described in equation [23], and the hydrolysis of Pu^{4+} to insoluble $Pu(OH)_4$, can be related to $PuO_2(CO_3)OH^-$. At pH 6, $Pu(OH)_5^-$ is roughly 10^{15} times the Pu^{4+} concentration. Pu^{4+}, as limited by hydrolysis is roughly 10^{-23} M. This agrees well with empirical evidence that plutonium exists at these pH's at concentrations of roughly 10^{-8} M (ionic strength = 1.0).[40] The highest free carbonate concentration in the groundwater is about 10^{-7} M.

After substituting the appropriate values into equation [6] the resulting calculation gives 10^{-5} M plutonium at a redox potential of 600 mv (SHE). This is roughly 250 nCi/L of plutonium as Pu-239. This estimate gives a base figure representing only the maximum concentration of Pu(VI) hydroxycarbonate that could occur in the diffusion layer around Plutonium(IV) hydroxide in its standard state. This "source concentration" would diminish exponentially with the distance travelled through the soil, and away from the source, because of the conversion of Plutonium(VI) to (V) and (IV) and their subsequent adsorption by the soil.[41]

Away from a plutonium source (saturated solubility conditions) the dominant factor in Pu(VI) transport is the equilibrium with Pu(V), which does not form a stable anionic complex with carbonate. Pu(V) was more than 10^4 times more thermodynamically stable than Pu(VI) in water from every groundwater monitoring well in the 643-G burial ground. Therefore, Plutonium(VI) is rapidly converted to Plutonium(V) in the presence of any redox couple in the groundwater (even at $E_H = + 0.6$ volts). Since the hydroxycarbonate species must be in equilibrium with Pu(VI), it is gradually consumed through the adsorption of the reduced plutonium in soil.

In terms of operating low-level burial sites in a humid environment, it would seem to be advantageous to avoid disposing of plutonium in a highly oxidizing environment.[38] This constraint would not normally affect disposal operations, but it would assure that plutonium mobility would not be enhanced by conversion to the oxidized Pu(V) and Pu(VI) species, the latter of which is mobile in the presence of basic or neutral carbonate.

Total Organic Carbon (TOC)

The groundwater in the vicinity of the burial ground is naturally oxic, and contains very little natural organic carbon. Aside from small amounts of humic material from areas that might have been low or marshy prior to burial ground operation (none have been identified) virtually all of the organic carbon dissolved in the groundwater must have come from the trenches. Some of this material is most likely in the form of acidic and potentially complexing compounds.

The nature of organic contaminants in the groundwater and burial ground trenches is critical to the mobility of plutonium. Organics that form strong complexes tend to mobilize Pu(IV) very efficiently. The most critical compounds in this respect are EDTA and its analogs (eg., EDTA and DTPA), but others such as TBP, DEHP, and even citrate could carry plutonium to the water table at trace levels. Plutonium(IV) forms such strong complexes that it competes favorably with naturally occurring Fe(III) and Ca in the groundwater for sub-stoichiometric concentrations of these anions.[34,36,42]

Several of the monitoring wells contain measurable amounts of dissolved organic carbon. Some of these, including C-17, E-21 and G-21, are located near the old solvent burning trenches. Others, like G-7, contain high concentrations of dissolved carbon for no apparent reason. Until an analysis of the organic material in the monitoring wells is completed, the source and potential impact of the organic carbon will be uncertain.

Fluoride

Fluoride is usually found at such low concentrations in groundwater that it is not important in radionuclide migration studies. Normally, it does not affect the mobility of fission products, although at high concentration, fluoride can precipitate Cm, Cr, Ni, and Sr. The importance of fluoride here is its ability to form anionic, and therefore mobile, complexes with plutonium and uranium in their most oxidized valence states. $PuO_2F_4^{2-}$ and $PuO_2F_3^-$ are the most thermodynamically stable species of plutonium(VI) in aqueous media at concentrations of about 5 x 10^{-6} M fluoride and above.[28] Uranium(VI) is stable as $UO_2F_4^{2-}$ at concentrations greater than about 1 x 10^{-3} M fluoride.

The equilibrium concentration of anionic fluoride complexes relative to PuO_2^{2+} and UO_2^{2+} concentrations were calculated from basic thermodynamic data[28,33] as follows.

For Uranium:

Equation		log k
[1]	$UO_2^{2+} + 4H^+ + 2e^- = U^{4+} + 2H_2O$	+9.20
[2]	$UO_2F_3^- + 4H^+ + 2e^- = U^{4+} + 3F^- + 2H_2O$	-2.59
[3]	$UO_2F_4^{2-} + 4H^+ + 2e^- = U^{4+} + 4F^- + 2H_2O$	-3.78
[1-3]	$UO_2^{2+} + 4F^- = UO_2F_4^{2-}$	+12.98
[1-2]	$UO_2^{2+} + 3F^- = UO_2F_3^-$	+11.79

Therefore:

$$\frac{[UO_2F_4^{2-}]}{[UO_2^{2+}]} = [F^-]^4 \times 10^{12.98}$$

$$\frac{[UO_2F_3^-]}{[UO_2^{2+}]} = [F^-]^3 \times 10^{11.79}$$

$$[F^-] = (BETAF) \times ([F]_{measured})$$

BETAF, the fraction of fluorine present as the fluoride anion, is:

$$BETAF = \frac{[F^-]}{[F^-] + [HF]} = \frac{1}{[H^{(+)}]/DS + 1}$$

Where:

$$DS = \frac{[H^+]\,[F^-]}{[HF]} = 7.2 \times 10^{-4}$$

And for Plutonium:

Equation		log k
[1]	$PuO_2^{2+} + 4H^+ + 2e^- = Pu^{4+} + 2H_2O$	+34.86
[2]	$PuO_2F_3^- + 4H^+ + 2e^- = Pu^{4+} + 3F^- + 2H_2O$	+18.60
[3]	$PuO_2F_4^{2-} + 4H^+ + 2e^- = Pu^{4+} + 4F^- + 2H_2O$	+15.57
[1-3]	$PuO_2^{2+} + 4F^- = PuO_2F_4^{2-}$	+19.29
[1-2]	$PuO_2^{2+} + 3F^- = PuO_2F_3^-$	+16.26

Therefore:

$$\frac{[PuO_2F_3^-]}{[Pu_2^{2+}]} = [F^-]^3 \times 10^{16.26}$$

$$\frac{[PuO_2F_4^{2-}]}{[PuO_2^{2+}]} = [F^-]^4 \times 10^{19.29}$$

$$[F^-] = (BETAF) \times ([F]_{measured})$$

The fluorine concentration statistics for the groundwater wells are summarized in Table 2. The average fluoride concentration in all wells was 0.1 ppm (5×10^{-6} M). The highest concentration measured was 0.8 ppm (4×10^{-5} M). The fraction of the total fluorine actually present as free fluoride (BETAF) is a function of pH. This function is plotted in Figure 7.

Using the equations above, the ratios of anionic Pu(VI) and U(VI) concentrations to the concentrations of the corresponding anionic fluoride complexes were calculated for the average and the highest fluoride concentrations that were measured in the groundwater wells (Table 2). The potential impact of the elevated fluoride concentrations is well illustrated by the effect of just 1×10^{-3} M fluoride on the equilibrium ratio of anionic to cationic species of both uranium and plutonium. And, as seen in the table, even the low concentrations of fluoride that were measured in the groundwater wells could have a significant impact upon the speciation of Pu(VI).

Pu(VI) may represent a significant fraction of the soluble, mobile plutonium in the 643-G groundwater, and its mobility is almost certainly enhanced by the formation of anionic and neutral complexes. A study of plutonium speciaton in well C-17 concluded that over 40% of the dissolved, unfilterable plutonium was Pu(VI).[39]*

The same study found that some of the plutonium in well C-17 may have been adsorbable by anion exchange resin (in the chloride form), but the results were statistically uncertain - the absolute quantity of such material being represented by the difference between 32.0 ± 3.0 pCi/L and 28.4± 4.2 pCi/L. Additional tests showed that almost 96% of the soluble plutonium could be adsorbed on cation exchange resin (in the acid form), but only 75% was adsorbed on soil in batch K_d tests. Therefore, although very little anionic plutonium was detected by direct extraction with ion exchange resins, some anionic plutonium probably does exist.

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* Some or all of the Pu(VI) that was detected may have been Pu(V), a species that does not form stable complexes.

TABLE 2

ANION/CATION SPECIATION RATIOS* FOR U (VI) AND PU (VI) WITH FLUORIDE

CASE	pH	[F]	BETAF	$\frac{[UO_2F_3^-]}{[UO_2^{2+}]}$	$\frac{[UO_2F_4{}^{2-}]}{[UO_2^{2+}]}$	$\frac{[PuO_2F_3^-]}{[PuO_2^{2+}]}$	$\frac{[PuO_2F_4^{2-}]}{[PuO_2^{2+}]}$
AVERAGE	5.34	$5x10^{-6}$	0.99	$7.3x20^{-5}$	$5.5x10^{-9}$	2.2	$1.1x10^{-2}$
WORST REAL	5.24	$4x10^{-5}$	0.99	$3.8x10^{-2}$	$2.3x10^{-5}$	$1.1x10^{3}$	$4.8x10^{1}$
HIGH F	5.34	10^{-3}	0.99	$6.2x10^{2}$	9.5	$1.8x10^{7}$	$1.9x10^{7}$

* At thermodynamic equilibrium

More than half of the soluble oxidized plutonium in the experimental water sample (from well C-17) may have been in the form of anionic Pu(VI) complexes. The equilibrium ratio for Pu(VI) in an aqueous system with 0.1 ppm total fluorine at a pH of 5.15 is roughly 69% $PuO_2F_3^-$ and 31% PuO_2^{2+}. This is approximately equivalent to the ratio that might be predicted based solely on the results of the soil adsorption (K_d) tests, in which 25% of the soluble plutonium in all oxidation states was not adsorbed. If all of the non-adsorbed plutonium in the soil adsorption experiment is assumed to have been Pu(VI), then roughly 63% (25%/40% of the total plutonium in the original water sample) of the Pu(VI) could have been in the form of anionic fluoride complexes.

The apparent discrepancy between the results of the soil adsorption tests and the ion exchange tests may be accounted for by examining the experimental method. As the equations for the formation constants show, the fraction of Pu(VI) in a solution as the anionic fluoride complex exhibits a third and even a fourth order dependence on the free fluoride concentration. The addition of acidic cation exchange resin to a sample of natural well water would have lowered the pH of the solution to less than 2.5, reducing BETAF by a factor of almost 20 (see Figure 7). If fluoride were present, consumption of free fluoride would easily explain why 96% of the total plutonium was adsorbed by the cation exchange resin instead of the 75% that was adsorbed by the less acidic soil. Additional tests with cation exchange resin that had been pre-equilibrated with an aliquot of the same well water sample would test that hypothesis.

The anion exchange resin adsorption test would also be expected to yield a poor recovery of a plutonium fluoride complex ($PuO_2F_3^-$) because the complexing anion, F^-, would be consumed by the resin. Any of the complex that was initially adsorbed by the anion exchange resin would have exchanged, reversibly, with chloride. However, as the resin equilibrated with the water sample, the fluoride activity of the solution would have decreased to the point that the plutonium fluoride complex would dissociate rapidly in the process of exchange with the free chloride in solution. Even with an adsorption selectivity coefficient of 20 for chloride/fluoride,[43] the reduction of fluoride activity in the solution would be greater than 99% under the reported experimental conditions. Pre-equilibration of the resin with an aliquot of the sample using a column arrangement is probably the easiest way to assure that the resin does not affect the sample.

In making these calculations, it was assumed that the well water composition (well C-17 was used) has not changed substantially with respect to fluoride, chloride, and pH. In attempting to analyze the data on well C-17, as reported here and in reference 39, it should also be mentioned that a significant amount of organic carbon has been detected in that well, and there is reason to believe that it could be a complexant/diluent combination, specifically TBP/ultrasene (see Operations History Section). This would be expected to influence the overall behavior of plutonium in the local environment, but it would not directly affect the behavior of Pu(VI). However, if a significant fraction of the non-adsorbed plutonium was in lower oxidation states, as was probably the case, the dissolved amount of Pu(VI) in the form of an anionic fluoride complex would be lower. The conclusions about Pu(VI) that are based on thermodynamic calculations would remain unchanged, but the mechanisms responsible for mobilizing the less oxidized plutonium (e. g., organic complexation) would assume relatively greater importance.

In spite of the complexes just described, plutonium distribution coefficients (K_d's) on soil are commonly on the order of 10,000, because the lower oxidation states of plutonium tend to dominate most systems.[13] These species are extremely insoluble at normal groundwater pH's and are also quite readily adsorbed on virtually all soils. Pu(VI), the only plutonium species that reacts to form anionic fluoride complexes, is relatively unstable in the environment. It only exists in strongly oxdizing solutions, in concentrated plutonium solutions (as a result of the disproportionation of Pu(IV)) or in extremely dilute quantities. Otherwise, it is dominated by Pu(V) in oxidized systems[37] and Pu(III) and (IV) in reducing or "anoxic" stystems (see Oxidation-Reduction Potential).

At the highest fluoride concentrations that have been observed in the groundwater wells (0.8 ppm) Pu(VI) would move with the groundwater until it was reduced to a lower oxidation state or until the fluoride concentration dropped back to normal. However, no plutonium has been detected recently in any of the monitoring wells that contain F^-. Also, well C-17 no longer contains measurable plutonium activity. At present, there is not more than 4 pCi/L of Pu-239 (see Radiochemical Summary) in any one well. Even the rapid migration of this amount of activity would merely serve to disperse plutonium concentrations to below the limits of detectability, as they are already below the EPA drinking water standard.[24]

Chloride

Except for its function as a counterion for cations in solution, chloride does not specifically affect the migration of any of the major radionuclides. None of the elements that form strong (anionic) chloride complexes (e.g. Hg, Zr, Pb, Sn, Bi) has a radioisotope that is of significance in Low-Level Waste Management.

However, high levels of chloride in the groundwater are a clear indication that salts from the waste trenches are affecting the chemistry of the groundwater.

The mean chloride concentration was approximately 8.1 ppm, the median value, 3.8 ppm. The five wells with the highest chloride concentrations are listed in Table E1. At least one of the wells, I-13, is significantly out of line, indicating the influence of chemicals from the low-level waste trenches above. High ionic strength, as inferred by a high chloride concentration, tends to enhance the mobility of strontium and cesium and any other radionuclides that are normally immobilized by ion exchange with the soil.

Nitrate

Nitrate is generally a weaker complexing ligand than chloride. It is relatively easy to reduce in acid solutions and is consumed rapidly by biological activity in the environment. At high concentrations, nitrate forms insoluble salts with the alkaline earths Np (V) and others, but the dissociation constants of these salts are all too great to be of significance in environmental modeling.

As was the case for high chloride concentrations, high concentrations of nitrate in the groundwater are indicative of a source of chemical contamination nearby. However, since some of the nitrate sources, (e.g. anion exchange resin, neutralized acid nitrate waste) contain plutonium, it is important to identify the migration paths of this anion. The mean nitrate concentration in all wells was 10.0 ppm; the median was 10.5 ppm. The highest nitrate concentration measured was 28 ppm (well I-15), which was just within three standard deviations of the mean.

Sulfate

In many cases sulfate tends to be a stronger complexing ligand than nitrate. It also forms moderately soluble salts (Ksp $SrSO_4 = 10^{-2.55}$) with the alkaline earths and other ions. But again, the solubility of these compounds does not limit the mobility of radionuclides in a low-level waste environment. Anionic sulfate complexes can be formed with nickel and neptunium, but the sulfate concentration in the groundwater was far too low for these to be favorable. The calculated ratio of $Ni(SO_4)_2^{2-}/ Ni^{2+}$ was 1.4×10^{-5} at 111 ppm, the highest sulfate concentration measured. The ratio for $NpO_2(SO_4)_2^{2-}/NpO_2^{2+}$ was only slightly more favorable at 7.8×10^{-4}.

The most important effect of the sulfate anion at these concentrations may be its tendency to reduce the hydrolysis of Pu(IV) in the environment, making it slightly more soluble.[44]

Phosphate

The radionuclides most affected by phosphate are uranium and plutonium.[28,31] Uranium is not widely prevalent in the low-level burial ground, although low concentrations occur naturally there, so it will not be considered in this discussion. A few wells were found to contain phosphate at measurable concentrations but none of these were located where plutonium contamination was likely to be present. Plutonium reacts extensively with phosphate and probably forms mobile, anionic complexes of Pu (IV) in the environment. The limiting factors in the formation of these complexes are the concentration of free HPO_4^{2-}, and the amount of free plutonium available. The thermodynamic data quoted by Benson and Teague[28] and the stability constants quoted by Cleveland[34] indicate that the formation of anionic phosphate complexes is highly favored, even under environmental conditions.

Relevant equations were calculated, and the results are given below:

Complex Dissociation Constants	log K
$Pu(HPO_4)^{2+} = Pu^{4+} + H^+ + PO_4^{3-}$	-25.33
$Pu(HPO_4)_2(s) = Pu^{4+} + 2H^+ + 2PO_4^{3-}$	-52.62
$Pu(HPO_4)_3^{2-} = Pu^{4+} + 3H^+ + 3PO_4^{3-}$	-70.35
$Pu(HPO_4)_4^{4-} = Pu^{4+} + 4H^+ + 4PO_4^{3-}$	-92.16

Acid Dissociation Constants	log K
$HPO_4^{2-} = H^+ + PO_4^{3-}$	-12.3
$H_2PO_4^- = H^+ + HPO_4^{2-}$	- 7.2
$H_3PO_4 = H^+ + H_2PO_4^-$	- 2.1

$$[HPO_4^{2-}] = B_2[PO_4]_{measured}$$

$$B_2 = \frac{[HPO_4^{2-}]}{[H_3PO_4] + [H_2PO_4^{-}] + [HPO_4^{2-}] + [PO_4^{3-}]}$$

$$\frac{1}{B_2} = \frac{[H^+]^2}{K_1K_2} + \frac{[H^+]}{K_2} + 1 + K_3\,[H^+]$$

See Figure 8 for a plot of B_2 vs. pH

	log K
$Pu(HPO_4)_3{}^{2-} = Pu^{4+} + 3HPO_4{}^{2-}$	-33.4
$Pu(HPO_4)_4{}^{4-} = Pu^{4+} + 4HPO_4{}^{2-}$	-43.0

$$\frac{[Pu(HPO_4)_3{}^{2-}]}{[Pu(IV)]} = \frac{[HPO_4{}^{2-}]^3}{10^{-33.4}} = 2.5\ x10^{33}(B_2[PO_4]_{measured})^3$$

At pH 5.5 ($B_2 = 1.9 \times 10^{-2}$) and the highest phosphate concentration measured ($5.9x10^{-5}$ M), the divalent phosphate complex is favored by 3.5×10^{15}. At the mean phosphate concentration (0.5 ppm as P) this ratio is reduced to 8.1×10^{13}, but the divalent anionic complex is still highly favored. The tetraphosphate complex is favored even more (see Table 3).

$$\frac{[PU(HPO_4)_4^{4-}]}{[Pu^{4+}]} = 10^{43.0}\ (B_2\ [PO_4]\ _{measured})^4$$

$$= 1.6 \times 10^{19}$$ at pH 5.5 and 1.8 ppm P (5.8×10^{-5}M phosphate)

and

$$= 9.5 \times 10^{16}$$ at pH 5.5 and 0.5 ppm P

TABLE 3

ANION/CATION SPECIATION RATIOS* FOR PU (IV) WITH PHOSPHATE

CASE	pH	$[PO_4]_T$	BETAP	$\frac{[Pu(HPO_4)_3^{2-}]}{[Pu^{4+}]}$	$\frac{[Pu(HPO_4)_4^{4-}]}{[Pu^{4+}]}$
AVERAGE	5.32	2.2×10^{-6}	1.30×10^{-2}	5.8×10^{10}	6.7×10^{12}
WORST REAL	5.50	5.8×10^{-5}	1.96×10^{-2}	3.5×10^{15}	1.6×10^{19}
HIGH PHOSPHATE	5.50	5.0×10^{-4}	1.96×10^{-2}	2.4×10^{18}	9.2×10^{22}

* At Thermodynamic Equilibrium

$[PO_4]_T$ Represents total phosphate in solution in moles/liter

These calculations indicate that the anionic phosphate complexes of Pu(IV) are highly favored, even at very low phosphate concentrations. These relationships are described in terms of Pu(IV) concentration, which is normally soluble only to 7 x 10^{-22} M at pH 5.5 without complexation. However, even the average phosphate concentration (0.5 ppm measured as phosphorus) would result in an equilibrium concentration of anionic plutonium of 5.7 x 10^{-8}M tying up only 1.4% of the total phosphate. This would represent a limit of 848 nCi/L of Pu-239 (based purely on solubility considerations).

Away from any solid plutonium source, and in the presence of normal burial ground soil, the steady state would be shifted by a factor of 10^3 (a conservative value of Kd for Pu^{4+}) decreasing the activity of the anionic plutonium to a maximum of about 1000 pCi/L of Pu(IV). This shows that while the complexing effects of phosphate are very important in terms of radionuclide transport, the amount of activity that could be mobilized by normal amounts of phosphate is fairly small. This statement is further supported by the phosphate analyses which show that phosphate is virtually non-existent in most of the groundwater beneath the low-level waste trenches.

Two wells, E-5 and E-17, were found to be contaminated with a phosphorus containing compound in 1981. The preliminary analysis was done by Inductively Coupled Plasma Emission Spectroscopy, and the concentrations were 13-15 ppm phosphorus. However, no phosphate was detectable by anion chromatography, indicating that these wells were probably contaminated by a TBP solvent spill that occurred around that area in 1971. Subsequent analyses performed in June 1982, were unable to detect phosphorus in either well. This illustrates the importance of obtaining a profile of the organic compounds in the groundwater also.

Summarizing, just the appearance of phosphate in any of the monitoring wells is an anomolous event. The median phosphate concentration was below the detection limit, and 90% of the wells contained less than 0.07 ppm of phosphorus. The effect of phosphate on radionuclide migration around the SRP burial ground should be minimal as long as these low concentrations do not increase by more than an order of magnitude.

Silicate

Dissolved silicates appear to have very little affect on radionuclide mobility at SRP. At the normal groundwater pH, dissolved silicates are largely in the form of hydrated silica $SiO_2 \cdot 2H_2O$, or in common notation, H_4SiO_4. Polymeric forms of silica, especially metasilicates, are also prevalent, but silica is considered hydrolyzed for the purpose of simple equilibrium calculations. In systems that are more acidic than about pH 10, dissolved silica has virtually no complexing strength.[45] At high pH, silicates tend to hydrolyze, and react weakly with most cations, including sodium, aluminium, potassium, strontium, and possibly cesium. These complexes are weak and generally unimportant, especially since none of the 643-G wells are alkaline.

Competing Cations

Sodium

Chemically, sodium affects radionuclide migration by simply raising the ionic strength of the aqueous system. This tends to make some salts less soluble, but only a few radionuclides are affected by solubility considerations, and these tend to be influenced far more strongly by the complexation strength of the counterions (anions). The major effect of excessive sodium is to enhance the mobility of cationic radionuclides by competing with them for ion exchange sites on the soil.

The highest sodium concentrations were on the order of 0.5 to 5 percent of the total ion exchange capacity that would normally be in the surrounding soil (dry bulk density 2.0, porosity 0.25, ion exchange capacity 0.5 meq/100g). This is not significantly large with respect to the normal calcium concentration because of the disparity in their relative soil affinities. Spalding[16] found the calcium selectivity with respect to sodium to be roughly 10^3 for soils derived from Conasauga shale (montmorillinite and kaolinite) at pH 5. Prout's data[13,46] can be analyzed to obtain a strontium/sodium selectivity of roughly 180 for Savannah River Plant soil (20% clay) at pH 5. Therefore, calcium concentration tends to be more important than sodium concentration in its effect on radionuclide retention where simple ion exchange with the soil is involved (eg., Sr-90, Co-60, etc.).

High concentrations of sodium have long been known to cause clay soils such as those at 643-G, to swell, thus decreasing their permeability to water. At moderately low concentrations, however, sodium may compete for adsorption sites on the soil, thus enhancing radionuclide migration. Two important radionuclides that would be affected are Sr-90 and Cs-137, both of which are normally adsorbed by an ion exchange mechanism.[19,47] Cesium is generally adsorbed irreversibly on clays, but sodium or potassium could compete somewhat for adsorption sites[19]. High concentrations of cations also tend to shift the pH at which the soil is electrically neutral (zero point of charge) to more alkaline (higher) values. This reduces or even reverses the electrostatic driving force for cation adsorption by the soil. This may have the effect of making the kinetics of adsorption slower, and permitting greater transport distances, especially for site-specific cations such as cesium. Cesium is adsorbed irreversibly on kaolinite once it contacts the proper adsorption site, but it can be easily dislodged from less specific exchange sites by other cations.

The statistics of the sodium data are summarized in Table D9. The median concentration (5.6 ppm) is slightly below the mean (8.3 ppm), and all but one of the wells (I-13) are within three standard deviations of the mean. Well I-13 had about 40 ppm of sodium, and four other wells contained 30 ppm or more. Excessive sodium would be expected to enhance the mobility of Cs-137, Sr-90, and Ru-106 as well as any other radionuclides that are cationic and not strongly complexed. There is no definite threshold for these effects, but they tend to be nearly additive. That is, solutions of high ionic strength are generally better at mobilizing cations, even when there are several different species involved.

Potassium

Potassium, like sodium, can only affect the migration of radionuclides by the mechanism of competing for ion exchange sites on the soil. As with sodium, elevated potassium concentrations in the groundwater are an indication that chemical - and possibly radioactive - contamination has migrated from the trenches. Potassium, with its smaller (unhydrated) volume might be expected to compete somewhat better with cesium for adsorption sites on the soil than does sodium. More recent work with SRP trench water and soil samples tends to support this theory,[49] but it is not necessarily confirmed in the literature.[19]

One extreme outlier (I-13) was identified among the groundwater wells. This well contained more than 50 times the median potassium concentration. A re-calculation of the overall data distribution without this well reduced the mean from 3.97 to 2.21 ppm. With the possible exception of Cs-137, which may be more affected, high potassium concentrations are roughly equivalent to high sodium with respect to radionuclide mobilization.

Calcium

Calcium is important to radionuclide migration, especially with respect to Sr-90. It competes with strontium for ion exchange sites on the soil to the extent that there is virtually no selectivity between them.[16] This means that where calcium exists in solution, strontium will also, and their exchange ratios with the soil (Kd's) will be about the same.

Calcium also affects the formation of complexes by competing for complexing ligands. This tends to be beneficial, especially in cases where polydentate ligands like EDTA may be present. For example, even though EDTA could form a strong, mobile complex with Sr-90, calcium in the environment competes so strongly that an excess of EDTA-to-calcium would be required to affect the mobility of the strontium at all. The calcium-EDTA complex is more than one hundred times stronger than the strontium-EDTA complex.[42] Unfortunately, this competition for EDTA doesn't really help in the case of plutonium, which complexes with EDTA considerably more strongly than does calcium.[34]

There was a wide range of calcium concentrations in the monitoring wells, but calcium was universally detectable. At least two values (C-5 and A-5) were more than three standard deviations from the mean, but this is not necessarily an indication of groundwater contamination. The mineralogy of the region around the burial ground includes calcareous deposits, which often cause calcium concentrations in nearby groundwater to reach 100 ppm or more. A sampling of wells in the area around the burial ground confirmed the existence of some locally high concentrations of natural calcium.

Barium, Strontium

Because they are usually present at such low concentrations, barium and strontium do not normally contribute significantly to the groundwater chemistry of the burial ground region. At subsaturated concentrations they simply provide more competition in cation exchange processes. However, the observation of 21 ppm of barium in Well I-13 proves that at least one of the nearby trenches is contaminating the groundwater.

In spite of the status of its isotope (Sr-90), strontium is no more important to radionuclide migration than barium. It is just naturally swamped out by calcium, and their chemical similarity makes consideration of the far less concentrated strontium unncessary. Except for the purpose of modelling strontium in the environment, or for tracing unusual sources of contamination, the concentration of strontium in these wells is of small importance.

Iron

Iron plays an extremely important part in radionuclide migration. Its oxides and, to a lesser extent, those of manganese, are considered to be two of the major adsorption substrates for the immobilization of radionuclides on sandy soils.[48] The presence of iron in the groundwater is also strong evidence of the influence of a nearby waste disposal site, or some other source of chemically reducing or strongly complexing contamination.

At the pH of the groundwater monitoring wells, iron is soluble only as Fe(II), Fe(III) being soluble to only about 80 ppb at pH 4. Therefore, groundwaters that contain iron concentrations in excess of about 0.1 ppm and are not overly acidic must be either strongly reducing or strongly complexing. Any of these properties can enhance the mobility of several radionuclides.[15] Recent work with SRP soils and water from the burial trenches indicates there is an inverse correlation between iron in the trenches and the distribution coefficient (K_d) of strontium.

Wells that are suspected of being contaminated because of their high iron concentrations include G-21, G-7, and C-19. Wells I-1, G-9, G-30 and C-5 are also considered high, even though they are all less than 1 ppm in iron.

Magnesium

Magnesium is another cationic species that contributes to the total ionic strength of the natural groundwater. Like calcium, it is a chemical analog of strontium, and they can be considered together in predicting their effect on strontium migration.

The average magnesium concentration in the groundwater wells was 1.6 ppm, but excluding well G-7, which was 39.9 ppm in magnesium, all of the other wells were within three standard deviations of the mean.

CONCLUSIONS

Burial Ground Performance

The Low Level Waste Burial Ground at the Savannah River Plant (SRP) is operating well within the guidelines set by the Department of Energy for such facilities. Furthermore, the results of this extensive monitoring survey prove that the waste ~~water~~ buried there does not pose a threat to the environment through groundwater contamination. With the exception of tritium,* the radioactivity levels in 63 out of the 66 groundwater monitoring wells are below drinking water standards (EPA standards based on 4 mrem exposure- Ref 24).

The anthropogenic activity on the sediments of the monitoring wells was also much less than the natural background from uranium and thorium decay products in the soil.

In addition to the radiochemical determinations, the groundwater survey detected seven wells that exceeded EPA drinking water standards for hazardous but non-radioactive chemicals.[24] These include two wells that exceeded the 1.0 ppm drinking water standard for barium and five that exceeded the 0.3 ppm standard for iron. No other chemical pollutants were detected at significant levels.

Long-Term Radionuclide Migration from Waste Trenches

A study was also performed on wells screened at the base of the waste trenches.[49] Of the twenty-four trench wells in the SRP burial ground, only seven contained perched water. These were analyzed for the same chemical parameters as in this study. Batch distribution coefficients were also determined for Sr-85 and Cs-137 with well-characterized soil taken from the burial ground.

The results of this study prove that, as anticipated, "perched" water in the trenches has greater ionic strength and is more reducing than the water in most of the groundwater monitoring wells. Furthermore, the distribution coefficients of Sr-85 and Cs-137 on burial ground soil were generally lower with trench water than they were with groundwater. As a result, these isotopes would move more slowly as they reached the groundwater.

The opposite is probably true for long-lived plutonium isotopes, at least in the absence of strong organic complexants (EDTA, DTPA, etc.). The reducing environment of the trenches favors plutonium (IV), and plutonium (III), which are highly insoluble at near neutral pH and very strongly retained by soil ($K_d = 10^3 - 10^4$). Therefore, the predominant tendency of plutonium is to remain stationary. Any traces of plutonium that eventually reach the groundwater environment would gradually disperse, because the higher oxidation potential of the groundwater would permit some conversion to Pu (V), with extremely small fractions (7.2×10^{-6} at $E_H = 600$ mV) of Pu (VI). The traces of Pu (VI) would then be transported as the hydroxycarbonate

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* The behavior of tritium in the groundwater at SRP is known extremely well and is described by J. W. Fenimore in Reference 8.

in concentrations that would probably be too small to measure without preconcentration.

The process described above should operate as a means of dispersing long-lived plutonium isotopes that reach the water table. However, the amount of plutonium that actually reaches the water table has been, and will continue to be, limited by the quantity of available complexant as well as the amount of plutonium in the waste. Therefore, the concentration of plutonium at the water table will gradually diminish over the long term, as the soluble complexants are leached away and the less soluble plutonium is left behind in the trenches.

A secondary increase in the rate of plutonium transport might be postulated to occur later, after the (organic) waste degradation products leach away and the trenches become more oxidizing. However, even relatively oxidizing rainwater would not favor migration unless a complexant anion was available to mobilize Pu (IV). Acidic rainfall would keep the plutonium in the Pu (III) and (IV) states, and any plutonium that became oxidized would most likely remain as Pu (V). Only very small fractions of the total would be ever free to move as $PuO_2(CO_3)OH^-$.

The most important mechanisms by which the waste in the SRP burial ground could contribute to the population dose would be through fairly direct pathways, such as plant uptake, intrusion, and erosion.[50] The radionuclides with moderate half-lives (less than 100 years) will have decayed away long before they reach the perimeter of the burial ground, let alone the nearest groundwater outcrop, a third of a mile away. Radionuclides in this class include Cs-137, Sr-90, Co-60, Ni-63, Pu-238, and others. Others, like I-129 and Tc-99, will be around for extremely long periods of time but in very dilute concentrations.

Pu-239, with its poor mobility and long half-life, will be gradually dispersed by the mechanisms described in this paper. However, this dispersion will probably not occur rapidly enough to dominate other mechanisms of transport (e.g. plant uptake, erosion, human intrusion). All of these factors are considered in the models that SRL is using to estimate the potential consequences (dose to any member of the population) of scenarios that may occur as a result of decommissioning the burial ground.[50]

RECOMMENDATIONS

Low-level counting should be performed on all groundwater wells in order to complete the evaluation of the overall performance of the burial ground and to see if low-level activity can be detected in the "suspect" wells as predicted.

The volume of the gross alpha and non-volatile beta-gamma sampling program could be diminished substantially. Yearly or semi-yearly surveys should be sufficient, especially if these samples are tested for a few basic chemical parameters (pH, conductivity, iron and D.O.).

Finally, the wells should be analyzed for organic materials that might be capable of complexing plutonium.

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M. B. Wood	Sr-90 Determination

APPENDIX A

ANALYTICAL METHODS

(INCLUDING TABLE A1)

APPENDIX A

Analyses

Anions (F^-, Cl^-, NO_3^-, $SO_4^=$, $HPO_4^=$)

Anions were determined by ion chromatography (IC) with conductivity detection (Dionex, Sunnyvale, CA). The mobile phase was a carbonate buffer, so bicarbonate and carbonate could not be measured by this technique. Samples were filtered with 0.45 micron membrane filters prior to analysis.

Cations (Na^+, K^+, Ca^{++}, Ba^{++}, Sr^{++}, Fe^{++}, Mg^{++})

Cations were determined by inductively coupled plasma (ICP) and/or atomic absorption spectroscopy (AAS). The methods were in good agreement for the ions (Ca^{++}, K^+, Na^+, and Mg^{++}) that were measured with both instruments. Samples were taken in polyethlene, filtered, and then acidified with ultrapure nitric acid prior to analysis.

Silica

Silica was determined by ICP as silicon in filtered and acidified samples. However, the acid spike typically contained more silica than the sample, so this analysis was not precise (± 50%).

Phosphate

Phosphate concentrations below 1 ppm were measured by ICP as phosphorus in filtered and acidified samples.

Dissolved Oxygen (DO)

Oxygen concentrations were determined with a polarographic type (YSI of Model #57) DO meter, standardized with air. Samples were obtained in glass DO bottles equipped with siphon-type caps. These allowed samples to be taken from below the water surface with minimum turbulence. These samples were also used in oxidation-reduction potential measurements and dissolved carbon measurements. Conductivity was measured with the YSI Model #33 (probe Model #3310) standardized with KCl in distilled water.

pH

pH was measured with a Markson Model #4403 and Sensorex S200C glass electrode.

APPENDIX A cont'd

Oxidation-Reduction Potential

The oxidation-reduction potential of the well water samples was measured with an Orion platinum electrode (Model #96-78) versus the saturated Calomel Electrode (SCE) that is built into the electrode. Results are reported with respect to the normal hydrogen electrode (NHE).

Total Carbon (Organic, Inorganic)

The organic carbon dissolved in the groundwater was measured as the difference between the total carbon (TC) and total inorganic carbon (TIC) in each sample. Samples were obtained from the dissolved oxygen sample bottles, and were filtered (Gelman AE, glass) prior to analysis. The instrument was a Total Carbon Analyzer (Coulometrics Incorporated, Wheat Ridge, CO).

TABLE A1 - ANALYSIS SENSITIVITY AND PRECISION

	Reported As	Detection Limit (ppm)	Precision (σ) (1-5 ppm)
Ion Chromatography			
F^-	(ppm)	0.05	5%
Cl^-	(ppm)	0.05	5%
NO_3^-	(ppm)	0.5	3%
$SO_4^=$	(ppm)	0.5	3%
$HPO_4^=$	(ppm)	0.5	3%
ICP			
P	(ppm of PO_4)	0.060	<5%
Si	(ppm)	0.030	<5%
Ca	(ppm)	0.025	<5%
Ba	(ppm)	0.002	<5%
Sr	(ppm)	0.001	<5%
Fe	(ppm)	0.005	<5%
Mg	(ppm)	0.002	<5%
AA			
Na	(ppm)	0.020	<5%
K	(ppm)	0.030	<5%
Ca	(ppm)	0.050	<5%
Fe	(ppm)	0.050	<5%

APPENDIX B

RADIOCHEMICAL DATA

(INCLUDING TABLES B1 AND B2)

RADIOCHEMICAL GROUNDWATER DATA

Table B1 describes the results of routine radiochemical surveys performed on the groundwater monitoring wells from 1973 through 1980. Units are pCi/L. Dots represent analyses that have not been performed (no data).

The first five headings are low level analyses performed in 1980: Pu-239, Pu-238 (with chemical separation), Cs-137, Sr-90, and Co-60. Zero values were below the routine detection limit for the low-level counting facility at SRL (<1 pCi/L of Pu-238/239; <8 pCi/l of Cs-137/Co-60; <6 pci/L Sr-90).

The second five headings, YEAR 6 through YEAR 0 represent the gross alpha measurements from 1976 through 1980. YEAR 36M represents the mean of all gross alpha measurements from 1973 through 1976, and Slope 1 is the slope (in pCi/L/year) of the data over that time. YEAR M is the average of all gross alpha measurements made on a particular well, from 1973 through 1980. Slope 2 is the slope of the gross alpha measurements from 1976 through 1980.

The variables YEAR 6 BG through YEAR 0 BG are the non-volatile beta-gamma measurements of the wells from 1976 through 1980. YEAR MBG is the average of all measurements of beta-gamma (non-volatile) for each well, and Slope 2BG is the slope of the beta-gamma analyses (pCi/L/year) from 1976 through 1980.

TABLE B-1

WELL	PU239	PU238	CS137	SR90	CO60	YEAR6	YEAR7	YEAR8	YEAR9	YEAR0	YEAR36M	SLOPE1	YEARM	SLOPE2	YEAR6BG	YEAR7BG	YEAR8BG	YEAR9BG	YEAR0BG	YEARMBG	SLOPE2BG
A-01	.	.	0	.	0	1	1	1	0	0	0.6	-0.4	0.6	-0.25	20	40	30	5	10	21.0	-2.50
A-03	0	11	0	30	0	1	1	2	1	0	0.6	0.5	1.0	-0.25	620	200	100	82	66	213.6	-138.50
A-05	.	.	.	.	.	3	5	3	5	4	3.6	0.9	4.0	0.25	30	60	20	18	7	27.0	-5.75
A-07	.	.	.	.	.	1	0	0	1	3	0.5	-0.4	1.0	0.50	10	60	10	20	3	20.6	-1.75
A-09	0	0	0	0	0	2	4	1	4	1	1.6	-0.3	2.4	-0.25	20	40	30	20	9	23.8	-2.75
A-11	.	.	.	.	.	2	1	4	1	0	0.8	0.1	1.6	-0.50	40	30	30	8	3	22.2	-9.25
A-19	.	.	.	.	.	1	2	2	0	1	0.6	0.0	1.2	0.00	20	50	40	14	4	25.6	-4.00
A-21	.	.	.	.	.	1	1	1	1	1	0.7	0.0	1.0	0.00	30	30	50	19	5	26.8	-6.25
A-23	.	.	.	.	.	1	2	1	2	0	1.3	0.0	1.2	-0.25	30	50	20	13	11	24.8	-4.75
A-32	.	.	.	.	.	3	0	2	1	2	1.5	0.1	1.6	-0.25	30	60	30	16	2	27.6	-7.00
A-34	.	.	.	.	.	1	1	1	1	1	1.0	0.0	1.0	0.00	50	20	30	19	9	25.6	-10.25
A-36	.	.	.	.	.	2	1	1	1	1	0.8	-0.2	1.2	-0.25	15	7	1	1	1	5.0	-3.50
C-01	2	4	0	18	0	2	1	1	0	0	1.5	-0.6	0.8	-0.50	40	20	20	34	28	28.4	-3.00
C-03	0	2	0	0	0	1	3	2	1	1	0.8	-0.3	1.6	0.00	30	30	20	10	17	21.4	-3.25
C-05	.	.	.	.	.	1	0	3	1	1	0.5	-0.2	1.2	0.00	30	30	50	10	4	24.8	-6.50
C-07	.	.	.	.	.	0	1	1	1	1	0.8	-0.3	0.8	0.25	10	40	30	2	11	18.6	0.25
C-09	.	.	.	.	.	1	2	1	0	1	1.2	-0.3	1.0	0.00	6	10	2	2	3	4.6	-0.75
C-11	.	.	.	.	.	1	1	1	0	1	1.2	-0.2	0.8	0.00	1	5	1	0	1	1.6	0.00
C-13	.	.	0	.	0	5	4	3	3	3	2.8	0.1	3.6	-0.50	60	51	120	18	7	51.2	-13.25
C-15	2	2	0	7	0	15	38	161	54	8	8.5	0.3	55.2	-1.75	40	70	3100	167	17	678.8	-5.75
C-17	0	0	0	0	0	390	47	41	2	12	104.0	28.3	98.4	-94.50	110	130	30	15	2	57.4	-27.00
C-19	.	.	.	.	.	1	0	1	1	1	0.6	-0.2	0.8	0.00	3	7	0	1	5	3.2	0.50
C-21	.	.	.	.	.	1	3	3	3	2	1.3	0.0	2.4	0.25	40	50	40	5	4	27.8	-9.00
C-23	.	.	.	.	.	1	0	1	1	1	0.6	0.0	0.8	0.00	1	2	0	0	0	0.6	-0.25
C-30	.	.	.	.	.	3	1	2	1	2	1.1	-0.1	1.8	-0.25	30	50	40	16	5	28.2	-6.25
C-32	.	.	.	.	.	2	2	1	1	2	1.8	0.0	1.6	0.00	7	8	3	1	8	5.4	0.25
C-34	.	.	.	.	.	2	2	3	1	1	1.5	0.0	1.8	-0.25	10	60	50	72	42	46.8	8.00
C-36	.	.	.	.	.	0	0	1	1	1	0.6	0.0	0.6	0.25	8	3	7	1	1	4.0	-1.75
E-01	.	.	.	.	.	1	2	3	1	2	1.8	0.5	1.8	0.25	5	40	30	17	17	21.8	3.00
E-03	.	.	.	.	.	3	2	3	1	1	2.2	-0.4	2.0	-0.50	10	40	20	12	21	20.6	2.75
E-05	.	.	.	.	.	1	2	0	3	2	2.6	-0.6	1.6	0.25	30	30	20	20	7	21.4	-5.75
E-07	.	.	.	.	.	4	3	3	1	1	3.5	-0.9	2.4	-0.75	20	3	10	19	5	11.4	-3.75
E-09	.	.	.	.	.	1	0	1	0	1	1.0	-0.1	0.6	0.00	3	5	4	1	2	3.0	-0.25
E-13	0	0	0	0	0	1	4	2	1	1	0.6	0.1	1.8	0.00	30	10	40	0	5	17.0	-6.25
E-15	.	.	.	.	.	0	0	2	2	2	1.2	1.2	1.2	0.50	40	40	40	104	74	59.6	8.50
E-17	0	0	10	0	13	61	38	1	1	0	14.7	6.2	20.2	-15.25	110	120	40	13	22	61.0	-22.00
E-19	.	.	0	.	0	1	0	1	0	0	0.6	0.1	0.4	-0.25	20	30	20	14	9	18.6	-2.75
E-21	.	.	.	.	.	1	1	1	1	1	0.4	-0.1	1.0	0.00	2	5	2	0	1	2.0	-0.25
E-23	.	.	.	.	.	1	1	1	1	2	0.7	0.1	1.2	0.25	20	30	10	23	0	16.6	-5.00
E-30	.	.	.	.	.	1	1	1	1	1	1.0	-0.1	1.0	0.00	30	10	5	2	8	11.0	-5.50
E-32	.	.	.	.	.	3	2	3	1	4	2.1	0.2	2.6	0.25	35	30	20	9	37	26.2	0.50
E-34	.	.	.	.	.	1	1	1	1	1	1.0	0.8	1.0	0.00	50	30	10	20	12	24.4	-9.50
E-36	.	.	.	.	.	2	2	2	1	2	1.7	0.0	1.8	0.00	20	40	50	14	5	25.8	-3.75
G-01	.	.	.	.	.	1	4	1	1	0	0.8	-0.1	1.4	-0.25	30	30	10	19	0	17.8	-7.50
G-03	.	.	.	.	.	1	1	1	1	2	1.5	0.6	1.2	0.25	30	30	20	35	0	23.0	-7.50
G-05	.	.	.	.	.	2	1	1	1	1	1.8	-0.5	1.2	-0.25	50	30	10	23	1	22.8	-12.25
G-07	.	.	.	.	.	1	2	0	2	1	1.2	0.2	1.2	0.00	20	50	30	45	15	32.0	-1.25
G-09	.	.	.	.	.	2	1	4	2	2	1.4	-0.2	2.2	0.00	30	40	[illegible]	10	7	21.4	-5.75
G-13	.	.	.	.	.	3	3	3	2	4	2.6	0.0	3.0	0.25	60	30	[illegible]	6	14	[illegible]	[illegible]

TABLE-B1 (Cont'd)

WELL	PU239	PU238	CS137	SR90	CO60	YEAR6	YEAR7	YEAR8	YEAR9	YEAR0	YEAR36M	SLOPE1	YEARM	SLOPE2	YEAR6BG	YEAR7BG	YEAR8BG	YEAR9BG	YEAR0BG	YEARMBG	SLOPE2BG
G-15	.	.	.	.	.	1	1	1	1	1	0.7	-0.2	1.0	0.00	2	10	3	0	0	3.0	-0.50
G-17	.	.	0	.	0	1	1	2	1	1	0.7	5.2	1.2	0.00	30	30	70	11	13	30.8	-4.25
G-19	.	.	.	.	.	1	1	0	1	0	0.5	-0.1	0.6	-0.25	20	10	6	1	1	7.6	-4.75
G-21	4	17	16	1600	0	14	52	54	42	33	87.6	-13.5	39.0	4.75	140	120	90	294	452	219.2	78.00
G-23	.	.	.	.	.	1	1	1	0	0	1.0	-0.1	0.6	-0.25	14	4	0	2	4	4.8	-2.50
G-28	.	.	.	.	.	2	1	1	1	1	0.8	-1.5	1.2	-0.25	10	7	1	1	3	4.4	-1.75
G-30	.	.	.	.	.	0	1	2	2	1	0.7	-0.1	1.2	0.25	25	40	20	3	4	18.4	-5.25
G-32	.	.	.	.	.	2	1	0	2	2	1.1	0.2	1.4	0.00	20	30	20	2	28	20.0	2.00
G-34	.	.	0	.	0	2	1	1	1	1	1.1	0.1	1.2	-0.25	20	30	30	14	19	22.6	-0.25
G-36	.	.	.	.	.	1	0	1	0	0	1.0	-0.3	0.4	-0.25	1	6	0	0	0	1.4	-0.25
I-01	.	.	0	.	0	4	2	3	3	6	3.4	0.4	3.6	0.50	10	30	40	10	36	25.2	6.50
I-03	.	.	.	.	.	3	2	6	4	4	2.2	0.1	3.8	0.25	30	30	50	14	14	27.6	-4.00
I-05	.	.	0	.	0	1	1	1	0	1	1.0	0.0	0.8	0.00	30	90	20	9	18	33.4	-3.00
I-07	.	.	0	.	0	1	1	0	2	0	1.2	-0.6	0.8	-0.25	10	30	30	18	2	18.0	-2.00
I-09	0	2	0	0	0	2	1	1	1	0	1.6	-0.9	1.0	-0.50	30	30	10	12	2	16.8	-7.00
I-13	0	2	10	19	0	22	16	16	2	18	26.5	-0.2	14.8	-1.00	340	320	180	46	130	203.2	-52.50
I-15	.	.	.	.	.	4	3	2	1	3	2.1	0.2	2.6	-0.25	50	30	30	24	26	32.0	-6.00
I-17	0	2	0	0	0	5	5	5	1	3	4.0	0.1	3.8	-0.50	30	80	30	27	33	40.0	0.75

Table B2 is a statistical summary of the radiochemical data. Variables are as explained in Table 1.

TABLE B-2

VARIABLE	N	MEAN	STANDARD DEVIATION	MINIMUM VALUE	MAXIMUM VALUE	STD ERROR OF MEAN	SUM
PU239	12	0.66666667	1.30267789	0.00000000	4.0000000	0.37605072	8.0000000
PU238	12	3.50000000	5.21361853	0.00000000	17.0000000	1.50504203	42.0000000
CS137	20	1.80000000	4.53756253	0.00000000	16.0000000	1.01462983	36.0000000
SR90	12	139.50000000	460.05049130	0.00000000	1600.0000000	132.80513750	1674.0000000
CO60	20	0.65000000	2.90688837	0.00000000	13.0000000	0.65000000	13.0000000
YEAR6	67	9.01492537	47.91516889	0.00000000	390.0000000	5.85376956	604.0000000
YEAR7	67	4.26865672	10.35371707	0.00000000	52.0000000	1.26490786	286.0000000
YEAR8	67	5.62686567	20.93267405	0.00000000	161.0000000	2.55733316	377.0000000
YEAR9	67	2.67164179	8.13966729	0.00000000	54.0000000	0.99441863	179.0000000
YEAR0	67	2.35820896	4.70557855	0.00000000	33.0000000	0.57487792	158.0000000
YEAR36M	67	4.83582090	16.54477581	0.40000000	104.0000000	2.02126607	324.0000000
SLOPE1	67	0.33731343	3.99191743	-13.50000000	28.3000000	0.48769033	22.6000000
YEARM	67	4.78805970	14.31747737	0.40000000	98.4000000	1.74915826	320.8000000
SLOPE2	67	-1.66417910	11.68304199	-94.50000000	4.7500000	1.42731075	-111.5000000
YEAR6BG	67	41.76119403	84.91762310	1.00000000	620.0000000	10.37433883	2798.0000000
YEAR7BG	67	41.98507463	48.28215447	2.00000000	320.0000000	5.89860398	2813.0000000
YEAR8BG	67	73.95522388	376.48507688	0.00000000	3100.0000000	45.99497265	4955.0000000
YEAR9BG	67	22.13432836	42.75118128	0.00000000	294.0000000	5.22288806	1483.0000000
YEAR0BG	67	19.88059701	57.33012387	0.00000000	452.0000000	7.00398938	1332.0000000
YEARMBG	67	39.94328358	89.62285833	0.60000000	678.8000000	10.94917480	2676.2000000
SLOPE2BG	67	-5.47014925	20.99037128	-138.50000000	78.0000000	2.56438200	-366.5000000

APPENDIX C

CHEMICAL DATA

(INCLUDING TABLES C1 AND C2)

<u>Table C1</u> gives the average value of each chemical parameter for each well. Units are given. See Appendix A for analytical details.

TABLE C1 - BURIAL GROUND MONITORING WELL ANALYSES (1980-1982)

WELL	F (ppm	CL (ppm)	NO3 (ppm)	SO4 (ppm)	PO4 (ppm)	SI (ppm)	NA (ppm)	K (ppm)	CA (ppm)
A1	0.00	2.40	6.0	3.70	0.040	5.55	5.750	1.30	19.20
A3	0.00	3.10	5.8	5.20	0.020	4.60	2.800	0.92	19.60
A5	0.20	1.60	3.4	15.80	0.070	15.70	21.600	6.60	92.30
A11	0.00	0.90	3.3	0.70	0.040	3.70	1.700	18.40	0.87
A19	0.50	10.40	0.8	0.80	0.010	2.70	4.800	0.45	3.10
A21	0.10	1.70	4.1	0.70	0.020	3.00	2.300	0.30	2.30
A23	0.00	2.70	11.1	0.20	0.020	2.90	5.600	0.33	0.09
A32	0.00	2.90	15.6	0.30	0.020	2.60	6.900	0.18	0.12
A34	0.00	2.60	14.4	0.07	0.030	2.80	7.400	0.34	3.10
A36	0.08	7.30	16.1	0.80	1.800	3.39	5.320	1.57	0.89
C1	0.00	3.50	12.9	2.20	0.000	4.00	2.900	0.50	9.30
C3	0.05	7.50	2.6	16.40	0.030	15.70	31.900	2.99	44.60
C5	0.06	4.40	10.2	24.00	0.070	10.50	9.700	4.21	102.30
C7	0.03	5.80	8.3	1.30	0.025	4.90	4.200	1.22	7.50
C9	0.42	7.00	0.0	2.60	0.000	0.08	38.800	5.18	17.20
C11	0.00	1.00	9.4	2.00	0.000	3.28	1.600	1.48	1.80
C13	0.09	3.80	14.3	0.20	0.010	3.60	2.550	2.24	0.85
C15	0.40	3.10	4.9	3.50	0.000	3.10	4.000	1.23	7.50
C17	0.10	2.70	7.1	3.80	0.000	2.16	6.400	2.84	2.15
C19	0.00	4.20	0.0	0.00	0.000	1.59	2.270	0.77	3.88
C21	0.09	2.10	4.0	0.20	0.000	3.86	1.718	0.89	0.40
C23	0.05	1.95	7.2	0.70	1.600	4.28	4.010	1.07	7.82
C30	0.02	5.60	13.5	0.30	0.000	2.86	4.300	1.07	0.18
C32	0.33	5.80	13.2	3.20	0.000	3.38	5.550	1.34	1.91
C34	0.02	5.60	13.4	0.20	0.010	2.92	6.060	0.95	0.32
C36	0.11	2.40	8.7	0.60	0.00	3.34	3.610	0.75	0.34

TABLE C1 - BURIAL GROUND MONITORING WELL ANALYSES (1980-1982)

WELL	BA (ppm)	SR (ppm)	FE (ppm)	MG (ppm)	DO (ppm O_2)	TEMP (°C)	COND (µmho/cm)	pH	mV (Vs. NHE)	TC (ppmc)	TOC (ppmc)	TIC (ppmc)
A1	0.030	0.120	0.020	0.80	6.7	18.0	105	6.11	601	27.6	3.7	23.0
A3	0.020	0.100	0.020	0.60	5.6	15.0	138	6.95	547	14.1	0.0	14.1
A5	0.090	0.600	0.010	3.40	5.7	18.5	360	6.81	507	71.1	0.0	71.1
A11	0.010	0.005	0.020	0.03	8.3	18.5	55	4.90	553	14.9	0.0	14.9
A19	0.010	0.020	0.010	0.57	8.6	20.0	31	5.65	552	29.0	0.0	29.0
A21	0.000	0.040	0.000	0.18	8.0	20.0	20	5.22	558	9.8	0.0	9.8
A23	0.010	0.001	0.010	0.25	8.2	20.0	30	4.79	565	10.5	5.6	4.9
A32	0.010	0.001	0.000	0.27	8.6	20.0	35	4.72	571	10.4	5.2	5.2
A34	0.020	0.008	0.020	0.36	4.6	19.0	60	6.02	544	17.5	0.4	17.1
A36	0.170	0.120	0.000	0.15	5.6	21.0	50	4.24	541	4.0	4.0	0.0
C1	0.010	0.030	0.040	2.90	4.8	18.0	55	5.77	533	13.0	0.0	13.0
C3	0.090	0.290	0.040	2.37	7.9	19.0	130	6.14	394	56.5	0.0	56.5
C5	0.030	0.610	0.610	2.37	4.4	19.0	170	6.86	379	63.5	0.0	63.5
C7	0.020	0.370	0.070	0.95	6.6	19.0	25	5.61	392	63.5	0.0	9.0
C9	0.170	0.330	0.150	3.15	2.4	21.0	250	6.64	471	45.0	6.0	39.0
C11	0.240	0.130	0.046	0.74	6.3	21.0	20	5.04	582	11.0	11.0	0.0
C13	0.020	0.010	0.010	2.20	7.4	17.0	38	5.00	683	8.7	0.0	8.7
C15	0.012	0.017	0.020	0.75	6.0	19.0	20	5.80	448	10.0	0.0	10.0
C17	0.040	0.060	0.020	0.34	8.6	19.0	35	5.23	492	28.0	20.9	7.1
C19	0.240	0.000	13.320	0.00	2.7	22.0	100	6.04	345	8.0	0.0	8.0
C21	0.010	0.003	0.040	0.14	8.4	19.0	15	4.52	447	29.5	18.0	11.5
C23	0.240	0.010	0.000	0.38	6.1	21.0	30	5.40	541	7.0	0.0	7.0
C30	0.014	0.002	0.020	0.35	8.4	19.0	35	4.22	456	31.5	12.1	19.4
C32	0.000	0.010	0.000	0.51	6.5	21.0	40	4.67	581	1.0	0.0	1.0
C34	0.010	0.190	0.010	0.22	8.6	20.0	30	4.61	452	15.0	8.0	7.0
C36	1.200	0.500	0.000	0.17	8.3	19.0	25	4.63	563	4.0	3.0	1.0

TABLE C1 - BURIAL GROUND MONITORING WELL ANALYSES (1980-1982)

WELL	F (ppm)	CL (ppm)	NO3 (ppm)	SO4 (ppm)	PO4 (ppm)	SI (ppm)	NA (ppm)	K (ppm)	CA (ppm)
E1	0.20	2.50	2.9	3.80	0.00	3.63	4.27	1.72	32.80
E3	0.40	2.40	8.3	1.90	0.00	3.81	4.29	2.50	42.50
E5	0.02	9.00	14.6	1.80	15.10	6.61	7.90	3.74	8.50
E7	0.01	4.10	16.0	0.06	0.01	3.00	2.90	1.67	1.98
E9	0.00	4.00	16.2	0.00	0.00	5.93	4.47	1.96	19.70
E13	0.00	1.20	7.8	0.70	0.00	4.00	2.20	0.90	1.30
E17	0.40	3.80	10.5	111.00	13.70	5.52	30.00	10.40	56.00
E19	0.05	5.90	13.9	5.20	0.00	6.00	10.00	1.33	29.30
E21	0.00	2.00	10.6	1.80	0.00	3.09	4.69	1.13	0.20
E23	0.00	3.10	8.1	1.00	0.00	2.48	4.00	1.19	0.70
E30	0.00	3.80	16.5	1.20	0.00	3.74	6.52	0.77	1.49
E32	0.00	9.20	25.2	0.00	0.00	4.02	7.97	1.90	2.25
E34	0.00	1.70	12.6	0.00	0.00	3.42	5.89	0.98	3.30
E36	0.00	5.40	11.7	0.00	0.00	2.85	5.89	0.84	2.04
G1	0.00	2.60	9.6	3.50	0.00	4.47	3.61	1.05	5.84
G3	0.09	7.10	13.6	2.00	0.00	4.37	4.24	3.17	10.30
G5	0.14	4.20	7.1	2.90	0.00	4.88	4.57	1.27	7.94
G7	0.00	45.00	0.1	0.50	0.00	5.18	32.20	2.80	38.60
G9	0.10	5.20	1.2	21.00	0.00	5.71	11.00	8.50	16.00
G13	0.20	4.30	13.8	1.30	0.00	4.12	2.91	4.19	0.50
G15	0.00	2.00	16.8	1.60	0.00	4.52	5.15	0.97	1.13
G17	0.20	4.40	11.2	1.00	0.00	2.50	14.75	0.85	0.35
G19	0.00	3.70	5.4	3.30	0.00	3.15	4.80	2.65	4.81
G21	0.00	33.00	1.5	5.40	0.00	1.64	6.00	0.97	3.30
G23	0.10	4.40	0.5	3.70	0.00	6.38	4.30	2.82	105.00
G28	0.15	4.10	12.0	0.70	0.00	3.18	5.77	1.71	1.93

TABLE C1 - BURIAL GROUND MONITORING WELL ANALYSES (1980-1982) (Cont'd)

WELL	F (ppm)	Cl (ppm)	NO3 (ppm)	SO4 (ppm)	PO4 (ppm)	SI (ppm)	NA (ppm)	K (ppm)	CA (ppm)
G30	0.30	3.40	8.6	4.80	0.00	2.57	6.03	1.91	23.70
G32	0.29	1.85	14.8	0.20	0.00	3.45	6.39	0.19	0.40
G34	0.00	1.30	18.2	0.30	0.00	3.07	6.50	0.26	0.65
G36	0.22	3.80	14.1	2.00	0.00	3.00	6.30	0.44	1.05
I1	0.10	9.70	22.0	3.10	0.20	4.72	10.90	1.44	21.80
I5	0.00	2.40	5.5	2.50	0.04	5.57	4.60	0.80	6.50
I7	0.00	7.00	0.0	2.70	0.00	4.52	7.30	1.20	9.30
I9	0.00	3.20	14.0	7.20	0.00	5.92	5.70	1.20	17.70
I13	0.30	152.00	14.8	0.40	0.00	2.49	40.10	110.00	6.40
I15	0.10	35.00	28.0	0.00	0.00	1.04	19.20	3.70	4.30
I17	0.10	16.30	12.4	0.50	0.00	2.55	5.10	4.70	3.10

TABLE C1 - BURIAL GROUND MONITORING WELL ANALYSES (1980-1982)

WELL	BA (ppm)	SR (ppm)	FE (ppm)	MG (ppm)	DO (ppm O_2)	TEMP °C	COND μmho/cm	PH	MV vs. NHE	TC (ppmc)	TOC (ppmc)	TIC (ppmc)
E1	0.050	0.070	0.020	1.10	7.0	17	120	6.60	499	40.0	10.1	29.9
E3	0.080	0.120	0.010	1.16	6.7	20	138	6.25	402	60.5	11.9	48.6
E5	0.050	0.050	0.010	1.86	7.1	20	45	5.24	423	42.0	3.9	38.1
E7	0.010	0.020	0.020	1.70	7.9	20	30	2.38	478	9.0	5.0	4.0
E9	0.000	0.070	0.016	1.04	6.5	21	40	3.88	588	10.0	10.0	0.0
E13	0.001	0.008	0.000	0.43	8.4	20	20	2.91	491	9.5	4.3	5.2
E17	0.006	0.210	0.000	0.59	6.0	21	310	6.54	598	4.5	0.4	4.1
E19	0.020	0.140	0.000	0.62	5.6	17	76	7.17	491	14.4	0.2	14.2
E21	0.500	0.000	0.023	0.26	6.0	21	30	4.18	495	11.0	10.0	1.0
E23	0.060	0.005	0.000	0.42	5.3	21	25	4.15	448	8.5	3.3	5.2
E30	0.830	0.140	0.000	0.43	2.0	21	40	4.07	546	5.0	3.0	2.0
E32	0.830	0.010	0.000	1.12	2.6	21	47	4.31	541	0.0	0.0	0.0
E34	0.830	0.170	0.000	0.34	4.9	21	35	4.95	567	3.0	0.0	3.0
E36	0.830	0.010	0.000	0.34	7.5	21	40	5.39	497	0.0	0.0	0.0
G1	0.920	0.030	0.000	0.45	7.2	21	55	5.70	536	5.0	0.0	5.0
G3	0.500	0.040	0.012	0.79	4.6	21	70	5.69	430	11.0	0.0	11.0
G5	0.420	0.060	0.008	0.28	5.5	21	67	5.39	492	7.0	0.0	7.0
G7	0.670	0.420	60.600	39.90	1.5	21	530	6.48	172	93.0	45.0	48.0
G9	0.670	0.200	0.220	3.53	4.4	21	140	5.83	264	27.0	9.0	18.0
G13	0.500	0.000	0.015	1.51	4.5	20	55	4.27	543	0.0	0.0	0.0
G15	0.190	0.019	0.013	0.68	5.5	21	46	4.69	538	5.0	0.0	5.0
G17	0.020	0.010	0.018	0.66	7.4	18	60	5.44	668	0.0	0.0	0.0
G19	0.038	0.019	0.027	0.77	6.2	21	65	6.26	556	1105	4.0	7.0

TABLE C1 - BURIAL GROUND MONITORING WELL ANALYSES (1980-1982)

WELL	BA (ppm)	SR (ppm)	FE (ppm)	MG (ppm)	DO (ppm O_2)	TEMP °C	COND μmho/cm	PH	MV vs. NHE	TC (ppmc)	TOC (ppmc)	TIC (ppmc)
G21	0.380	0.020	112.800	4.30	2.8	21	500	5.03	369	245.0	225.0	20.0
G23	0.010	0.510	0.000	1.10	1.7	21	270	6.26	497	37.0	0.0	37.0
G28	0.380	0.010	0.000	0.14	4.4	21	35	5.15	534	3.0	0.0	3.0
G30	0.380	0.190	0.670	0.41	5.5	21	92	6.45	451	21.0	4.0	17.0
G32	0.000	0.000	0.030	0.37	4.7	18	35	4.62	546	3.0	0.0	3.0
G34	0.150	0.010	0.000	0.40	8.7	18	28	5.19	624	3.0	0.0	3.0
G36	0.000	0.090	0.000	0.41	7.2	21	30	3.45	558	4.0	4.0	0.0
I1	0.000	0.040	0.840	1.50	3.8	17	85	5.40	662	23.0	16.0	7.0
I5	0.000	0.030	0.030	0.71	2.5	15	45	5.40	653	12.0	6.0	6.0
I7	0.240	0.030	0.060	0.92	3.4	17	45	5.60	642	18.0	8.0	10.0
I9	0.000	0.060	0.015	0.89	2.8	16	90	5.50	634	9.0	3.0	6.0
I13	21.100	0.480	0.034	2.70	7.5	17	450	5.36	589	2.0	1.0	1.0
I15	0.700	0.280	0.010	7.60	4.2	17	70	6.41	411	5.0	0.0	5.0
I17	2.400	0.030	0.010	2.40	9.0	17	105	5.87	609	4.0	1.6	2.4

<u>Table C2</u>	compares the coefficient of variation of the overall groundwater chemistry (C.V. all wells) with that of the individual wells as a function of time (C.V. each well).
	<u>C.V. all wells</u> is equal to the standard deviation/mean of all 63 well measurements taken together (see also Appendix D).
	<u>C.V. each well</u> is the average of the coefficients of variation calculated for multiple analyses of individual wells, consisting of 3 or more measurements. Measurements were made on separate samples taken at intervals of 1-10 months, within one 18-month period.

TABLE C2 COEFFICIENTS OF VARIATION

PARAMETER	C.V. ALL WELLS	C. V. EACH WELL (18 MONTHS)
F	135	20
Cl	244	17
NO3	62	14
SO4	315	81
PO4	490	*
Si	62	23
Na	107	15
K	354	31
Ca	173	37
Ba	457	51
Sr	140	24
Fe	534	*
Mg	295	29
DO	35	12
TEMP	9	*
COND	122	30
PH	18	11
MV	18	27
TIC	124	*
TOC	371	*

* Insufficient data to estimate C.V. for this parameter.

APPENDIX D

STATISTICAL SUMMARY OF 643-G GROUNDWATER CHEMISTRY

(INCLUDING TABLES D1-D22)

Statistical Summary*

Table D1 gives the standard statistics for the 63 groundwater monitoring wells. The top part of the table includes all wells, while the bottom part is a modified version derived by excluding observations that more than three standard deviations from the mean of the original data and recalculating the distribution.

N = the number of observations (wells) used in calculating the distribution; other statistical parameters have their usual meanings (e.g., C.V. = coefficient of variance)

Table D2 gives the distribution of all of the groundwater wells, eliminating the zero values for statistical reasons. (This table was provided simply for comparison, since this statistical approach was not used.)

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* Please note that the units of each variable are as described in Appendix C, generally mg/L (ppm). The significant figures quoted in these tables are artifacts of the mathematics of the data analysis and are not meant to imply anything about the precision, which was typically 2 to 3 significant figures, depending on the technique.

TABLE D-1

ALL GW DATA

VARIABLE	N	MEAN	STANDARD DEVIATION	MINIMUM VALUE	MAXIMUM VALUE	STD ERROR OF MEAN	SUM	VARIANCE	C.V.
F	63	0.09714286	0.13084624	0.00000000	0.50000000	0.01648508	6.120000	0.017121	134.695
CL	63	8.19206349	19.96857654	0.90000000	152.00000000	2.51580417	516.100000	393.744049	243.755
NO3	63	10.00634921	6.18869607	0.00000000	28.00000000	0.77970242	630.400000	38.299959	61.848
SO4	63	4.57984127	14.41048392	0.00000000	111.00000000	1.81555032	288.530000	207.662047	314.650
PO4	63	0.52166667	2.55397997	0.00000000	15.10000000	0.32177123	32.865000	6.522814	489.581
SI	63	4.18396825	2.61033834	0.08000000	15.70000000	0.32887172	263.590000	6.813866	62.389
NA	63	8.13092063	8.68898579	1.60000000	40.10000000	1.09470931	512.248000	75.498474	106.863
K	63	3.91968254	13.86807729	0.18000000	110.00000000	1.74721351	246.940000	192.323568	353.806
CA	63	13.36952381	23.07215837	0.09000000	105.00000000	2.90681873	842.280000	532.324492	172.573
BA	63	0.58147619	2.65754968	0.00000000	21.10000000	0.33481979	36.633000	7.062570	457.035
SR	63	0.11393651	0.15951882	0.00000000	0.61000000	0.02009748	7.178000	0.025446	140.007
FE	63	3.00788889	16.06684641	0.00000000	112.80000000	2.02423238	189.497000	258.143553	534.157
MG	63	1.71396825	5.05264336	0.00000000	39.90000000	0.63657323	107.980000	25.529205	294.792
DO	63	5.90158730	2.04722409	1.50000000	9.00000000	0.25792599	371.800000	4.191126	34.689
TEMP	63	19.47619048	1.71903145	15.00000000	22.00000000	0.21657761	1227.000000	2.955069	8.826
COND	63	93.11111111	113.91608557	15.00000000	530.00000000	14.35207775	5866.000000	12976.874552	122.344
PH	63	5.31825397	0.97455124	2.38000000	7.17000000	0.12278192	335.050000	0.949750	18.325
MV	63	513.01587302	94.47878957	172.00000000	683.00000000	11.90320864	32320.000000	8926.241679	18.416
TC	63	20.85873016	34.43652709	0.00000000	245.00000000	4.33859461	1314.100000	1185.874398	165.094
TOC	63	7.72380952	28.71917261	0.00000000	225.00000000	3.61827565	486.600000	824.790876	371.827
TIC	63	13.13492063	16.34085676	0.00000000	71.10000000	2.05875444	827.500000	267.023600	124.408

GW DATA WITH 3-SIGMA VALUES MISSING

VARIABLE	N	MEAN	STANDARD DEVIATION	MINIMUM VALUE	MAXIMUM VALUE	STD ERROR OF MEAN	SUM	VARIANCE	C.V.
F	62	0.09064516	0.12123505	0.00000000	0.42000000	0.01539687	5.620000	0.0146979	133.747
CL	62	5.87258065	7.79643507	0.90000000	45.00000000	0.99014824	364.100000	60.7843998	132.760
NO3	63	10.00634921	6.18869607	0.00000000	28.00000000	0.77970242	630.400000	38.2999590	61.848
SO4	62	2.86338710	4.73413033	0.00000000	24.00000000	0.60123515	177.530000	22.4119900	165.333
PO4	61	0.06663934	0.30518126	0.00000000	1.80000000	0.03907446	4.065000	0.0931356	457.960
SI	61	3.80639344	1.57334785	0.08000000	10.50000000	0.20144655	232.190000	2.4754234	41.334
NA	61	7.10406557	6.65077767	1.60000000	32.30000000	0.85154482	433.348000	44.2328436	93.619
K	62	2.20870968	2.83284858	0.18000000	18.40000000	0.35977213	136.940000	8.0250311	128.258
CA	60	9.04466667	12.58669010	0.09000000	56.00000000	1.62493470	542.680000	158.4247677	139.161
BA	62	0.25053226	0.40648553	0.00000000	2.40000000	0.05162371	15.533000	0.1652305	162.249
SR	61	0.09783607	0.13413416	0.00000000	0.51000000	0.01717412	5.968000	0.0179920	137.101
FE	61	0.26388525	1.70508297	0.00000000	13.32000000	0.21831350	16.097000	2.9073079	646.146
MG	63	1.71396825	5.05264336	0.00000000	39.90000000	0.63657323	107.980000	25.5292050	294.792
DO	63	5.90158730	2.04722409	1.50000000	9.00000000	0.25792599	371.800000	4.1911265	34.689
TEMP	63	19.47619048	1.71903145	15.00000000	22.00000000	0.21657761	1227.000000	2.9550691	8.826
COND	60	73.10000000	70.91883640	15.00000000	360.00000000	9.15558241	4386.000000	5029.4813559	97.016
PH	62	5.36564516	0.90637021	2.91000000	7.17000000	0.11510913	332.670000	0.8215070	16.892
MV	62	518.51612903	84.47130504	264.00000000	683.00000000	10.72786647	32148.000000	7135.4013749	16.291
TC	63	20.85873016	34.43652709	0.00000000	245.00000000	4.33859461	1314.100000	1185.8743984	165.094
TOC	62	4.21935484	7.20502645	0.00000000	45.00000000	0.91503927	261.600000	51.9124061	170.761
TIC	61	11.35901639	13.20762882	0.00000000	56.50000000	1.69106359	692.900000	174.4414590	116.276

TABLE D-2

```
GW DATA WITH ALL ZERO VALUES SET TO MISSING
VARIABLE          N        MEAN       STANDARD       MINIMUM       MAXIMUM     STD ERROR          SUM       VARIANCE          C.V.
                                     DEVIATION         VALUE         VALUE       OF MEAN

F                37    0.16540541    0.13372016    0.01000000    0.50000000    0.02198346       6.120000       0.017881        80.844
CL               63    8.19206349   19.96857654    0.90000000  152.00000000    2.51580417     516.100000     398.744049       243.755
NO3              60   10.50666667    5.90776182    0.10000000   28.00000000    0.76268877     630.400000      34.901650        56.229
SO4              57    5.06192982   15.08070544    0.06000000  111.00000000    1.99748823     288.530000     227.427677       297.924
PO4              21    1.56500000    4.30189086    0.01000000   15.10000000    0.93874955      32.865000      18.506265       274.881
SI               63    4.18396825    2.61033834    0.08000000   15.70000000    0.32887172     263.590000       6.813866        62.389
NA               63    8.13092063    8.68898579    1.60000000   40.10000000    1.09470931     512.248000      75.498474       106.863
K                63    3.91968254   13.86807729    0.18000000  110.00000000    1.74721351     246.940000     192.323568       353.806
CA               63   13.36952381   23.07215837    0.09000000  105.00000000    2.90681873     842.280000     532.324492       172.573
BA               55    0.66605455    2.83751652    0.00100000   21.10000000    0.38261065      36.633000       8.051500       426.019
SR               59    0.12166102    0.16208368    0.00100000    0.61000000    0.02109108       7.178000       0.026245       133.160
FE               44    4.30675000   19.14374634    0.00800000  112.80000000    2.88602835     189.497000     366.483024       444.506
MG               62    1.74161290    5.08908449    0.03000000   39.90000000    0.64631438     107.980000      25.898781       292.205
DO               63    5.90158730    2.04722409    1.50000000    9.00000000    0.25792599     371.800000       4.191126        34.689
TEMP             63   19.47619048    1.71903145   15.00000000   22.00000000    0.21657761    1227.000000       2.955069         8.826
COND             63   93.11111111  113.91608557   15.00000000  530.00000000   14.35207775    5866.000000   12976.874552       122.344
PH               63    5.31825397    0.97455124    2.38000000    7.17000000    0.12278192     335.050000       0.949750        18.325
MV               63  513.01587302   94.47878957  172.00000000  683.00000000   11.90320864   32320.000000    8926.241679        18.416
TC               63   20.85873016   34.43652709    0.00000000  245.00000000    4.33859461    1314.100000    1185.874398       165.094
TOC              35   13.90285714   37.62442484    0.20000000  225.00000000    6.35968855     486.600000    1415.597345       270.624
TIC              55   15.04545455   16.65249195    1.00000000   71.10000000    2.24542156     827.500000     277.305488       110.681
```

Tables D3-D20 provide a reasonably complete summary of the statistical information for each variable in the groundwater system. Calculations were performed using SAS (Statistical Analysis System, SAS Institute Inc; Cary, North Carolina) procedures on the accumulated data.

Variables reported are:

N	number of wells analyzed
STD DEV	Standard Deviation
USS	uncorrected sum of squares
CV	coefficient of variation
T:MEAN=0	Students T value for hypothesis testing MEAN=0 and the probability of a greater magnitude for this T value
NUM=0	Number of observations numerically greater than zero
D:NORMAL	is the Kolomogrov - Smirnov D-statistic for this distribution
STD MEAN	standard error of the mean

Skewness $a_3 = \frac{m_3}{(m_2)^{3/2}}$ where m_2, m_3, m_4, are second, third and fourth moments about the sample mean

Kurtosis $a_4 = \frac{m_4}{(m_2)^2}$

Other statistics, including quantities, extreme values, range, mode, etc., have their usual meaning.

Modified distributions, with all data points more than three standard deviations from the mean set missing, are given under the appropriate heading (GW Data with 3-Sigma Values Missing). The number of values eliminated in this way is reported at the bottom of each table under the heading "Missing Value Count." Variables with no "Anomalous" (>3 sigma) values were nitrate, magnesium, dissolved oxygen (DO), and total carbon. No modified distributions were calculated for these variables.

TABLE D-3

```
ALL GW DATA

UNIVARIATE

VARIABLE=F

                  MOMENTS                                         QUANTILES(DEF=4)                                        EXTREMES

N                 63   SUM WGTS          63       100% MAX        0.5      99%          0.5       LOWEST    ID           HIGHEST    ID
MEAN       0.0971429   SUM             6.12        75% Q3        0.14      95%          0.4            0(I9         )       0.4(C15       )
STD DEV     0.130846   VARIANCE   0.0171207        50% MED       0.05      90%        0.318            0(I7         )       0.4(E3        )
SKEWNESS     1.47792   KURTOSIS     1.29595        25% Q1           0      10%            0            0(I5         )       0.4(E17       )
USS            1.656   CSS          1.06149         0% MIN          0       5%            0            0(G34        )      0.42(C9        )
CV           134.695   STD MEAN   0.0164851                                 1%            0            0(G21        )       0.5(A19       )
T:MEAN=0     5.89278   PROB>|T|      0.0001       RANGE           0.5
SGN RANK       351.5   PROB>|S|      0.0001       Q3-Q1          0.14
NUM ¬= 0          37                              MODE              0
D:NORMAL    0.228916   PROB>D         <0.01
```

```
GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=F

                  MOMENTS                                         QUANTILES(DEF=4)                                        EXTREMES

N                 62   SUM WGTS          62       100% MAX       0.42      99%         0.42       LOWEST    ID           HIGHEST    ID
MEAN       0.0906452   SUM             5.62        75% Q3      0.1175      95%          0.4            0(I9         )      0.33(C32       )
STD DEV     0.121235   VARIANCE   0.0146979        50% MED       0.04      90%          0.3            0(I7         )       0.4(C15       )
SKEWNESS     1.43423   KURTOSIS     1.09275        25% Q1           0      10%            0            0(I5         )       0.4(E3        )
USS            1.406   CSS         0.896574         0% MIN          0       5%            0            0(G34        )       0.4(E17       )
CV           133.747   STD MEAN   0.0153969                                 1%            0            0(G21        )      0.42(C9        )
T:MEAN=0     5.88725   PROB>|T|      0.0001       RANGE          0.42
SGN RANK         333   PROB>|S|      0.0001       Q3-Q1        0.1175
NUM ¬= 0          36                              MODE              0
D:NORMAL    0.227326   PROB>D         <0.01

 MISSING VALUE         .
         COUNT         1
  % COUNT/NOBS      1.59
```

TABLE D-4

```
ALL GW DATA

UNIVARIATE

VARIABLE=CL

                MOMENTS                                     QUANTILES(DEF=4)                                      EXTREMES

N                 63   SUM WGTS          63     100% MAX       152       99%         152         LOWEST    ID             HIGHEST    ID
MEAN         8.19206   SUM            516.1      75% Q3        5.8       95%        34.6            0.9(A11       )         16.3(I17       )
STD DEV      19.9686   VARIANCE     398.744      50% MED       3.8       90%       10.12              1(C11       )           33(G21       )
SKEWNESS      6.3848   KURTOSIS     44.9554      25% Q1        2.4       10%         1.7            1.2(E13       )           35(I15       )
USS          28950.1   CSS          24722.1       0% MIN       0.9        5%        1.22            1.3(G34       )           45(G7        )
CV           243.755   STD MEAN      2.5158                               1%         0.9            1.6(A5        )          152(I13       )
T:MEAN=0     3.25624   PROB>|T| 0.00183245     RANGE         151.1
SGN RANK        1008   PROB>|S|      0.0001     Q3-Q1           3.4
NUM ¬= 0          63                            MODE            2.4
D:NORMAL    0.376613   PROB>D         <0.01



GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=CL

                MOMENTS                                     QUANTILES(DEF=4)                                      EXTREMES

N                 62   SUM WGTS          62     100% MAX        45       99%          45         LOWEST    ID             HIGHEST    ID
MEAN         5.87258   SUM            364.1      75% Q3        5.8       95%     30.4948            0.9(A11       )         10.4(A19       )
STD DEV      7.79644   VARIANCE     60.7844      50% MED       3.8       90%        9.55              1(C11       )         16.3(I17       )
SKEWNESS     3.73255   KURTOSIS     14.5872      25% Q1        2.4       10%         1.7            1.2(E13       )           33(G21       )
USS          5846.05   CSS          3707.85       0% MIN       0.9        5%       1.215            1.3(G34       )           35(I15       )
CV            132.76   STD MEAN    0.990148                               1%         0.9            1.6(A5        )           45(G7        )
T:MEAN=0     5.93101   PROB>|T|      0.0001     RANGE          44.1
SGN RANK       976.5   PROB>|S|      0.0001     Q3-Q1           3.4
NUM ¬= 0          62                            MODE            2.4
D:NORMAL    0.288919   PROB>D         <0.01

 MISSING VALUE       .
         COUNT       1
  % COUNT/NOBS    1.59
```

TABLE D-5

```
ALL GW DATA

UNIVARIATE

VARIABLE=NO3

                MOMENTS                                  QUANTILES(DEF=4)                                        EXTREMES

N                 63  SUM WGTS        63    100% MAX        28      99%          28        LOWEST    ID           HIGHEST    ID
MEAN         10.0063  SUM          630.4     75% Q3       14.1      95%       21.24             0(I7       )        16.8(G15      )
STD DEV       6.1887  VARIANCE      38.3     50% MED      10.5      90%       16.38             0(C19      )        18.2(G34      )
SKEWNESS    0.333874  KURTOSIS   0.20389     25% Q1        5.4      10%    0.959999             0(C9       )          22(I1       )
USS           8682.6  CSS         2374.6      0% MIN         0       5%        0.02           0.1(G7       )        25.2(E32      )
CV           61.8477  STD MEAN  0.779702                             1%           0           0.5(G23      )          28(I15      )
T:MEAN=0     12.8335  PROB>|T|    0.0001     RANGE          28
SGN RANK         915  PROB>|S|    0.0001     Q3-Q1         8.7
NUM ¬= 0          60                         MODE            0
D:NORMAL   0.0726649  PROB>D       >0.15
```

TABLE D-6

```
ALL GW DATA

UNIVARIATE

VARIABLE=SO4

                MOMENTS                                           QUANTILES(DEF=4)                                     EXTREMES

N                  63   SUM WGTS          63      100% MAX        111       99%         111        LOWEST    ID           HIGHEST    ID
MEAN          4.57984   SUM           288.53       75% Q3         3.5       95%     20.0799             0(I15      )          15.8(A5       )
STD DEV       14.4105   VARIANCE     207.662       50% MED        1.3       90%     6.47998             0(E36      )          16.4(C3       )
SKEWNESS      6.77771   KURTOSIS     49.8046       25% Q1         0.3       10%   0.0239999             0(E34      )            21(G9       )
USS           14196.5   CSS            12875        0% MIN          0        5%           0             0(E32      )            24(C5       )
CV             314.65   STD MEAN     1.81555                                 1%           0             0(E9       )           111(E17      )
T:MEAN=0      2.52256   PROB>|T|   0.0142308      RANGE           111
SGN RANK        826.5   PROB>|S|      0.0001      Q3-Q1           3.2
NUM ^= 0           57                             MODE              0
D:NORMAL     0.382069   PROB>D         <0.01
```

```
GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=SO4

                MOMENTS                                           QUANTILES(DEF=4)                                     EXTREMES

N                  62   SUM WGTS          62      100% MAX         24       99%          24        LOWEST    ID           HIGHEST    ID
MEAN          2.86339   SUM           177.53       75% Q3        3.35       95%       16.31             0(I15      )           7.2(I9       )
STD DEV       4.73413   VARIANCE      22.412       50% MED        1.3       90%        5.34             0(E36      )          15.8(A5       )
SKEWNESS        3.146   KURTOSIS     10.2096       25% Q1         0.3       10%   0.0179998             0(E34      )          16.4(C3       )
USS           1875.47   CSS          1367.13        0% MIN          0        5%           0             0(E32      )            21(G9       )
CV            165.333   STD MEAN    0.601235                                 1%           0             0(E9       )            24(C5       )
T:MEAN=0      4.76251   PROB>|T|      0.0001      RANGE            24
SGN RANK          798   PROB>|S|      0.0001      Q3-Q1          3.05
NUM ^= 0           56                             MODE              0
D:NORMAL     0.276422   PROB>D         <0.01

 MISSING VALUE        .
         COUNT        1
  % COUNT/NOBS     1.59
```

TABLE D-7

```
ALL GW DATA

UNIVARIATE

VARIABLE=P04

               MOMENTS                                          QUANTILES(DEF=4)                                              EXTREMES

N                 63   SUM WGTS          63     100% MAX       15.1       99%         15.1          LOWEST     ID           HIGHEST     ID
MEAN        0.521667   SUM           32.865      75% Q3        0.02       95%         1.76              0(I17        )          0.2(I1         )
STD DEV      2.55398   VARIANCE     6.52281      50% MED          0       90%         0.07              0(I15        )          1.6(C23        )
SKEWNESS     5.38197   KURTOSIS     28.3111      25% Q1           0       10%            0              0(I13        )          1.8(A36        )
USS          421.559   CSS          404.414       0% MIN          0        5%            0              0(I9         )         13.7(E17        )
CV           489.581   STD MEAN    0.321771                                1%            0              0(I7         )         15.1(E5         )
T:MEAN=0     1.62123   PROB>|T|    0.110043      RANGE         15.1
SGN RANK       115.5   PROB>|S|      0.0001      Q3-Q1         0.02
NUM ¬= 0          21                             MODE             0
D:NORMAL    0.490821   PROB>D         <0.01
```

```
GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=P04

               MOMENTS                                          QUANTILES(DEF=4)                                              EXTREMES

N                 61   SUM WGTS          61     100% MAX        1.8       99%          1.8          LOWEST     ID           HIGHEST     ID
MEAN       0.0666393   SUM            4.065      75% Q3       0.015       95%     0.186999              0(I17        )         0.07(A5         )
STD DEV     0.305181   VARIANCE   0.0931356      50% MED          0       90%         0.04              0(I15        )         0.07(C5         )
SKEWNESS     5.33608   KURTOSIS     27.7553      25% Q1           0       10%            0              0(I13        )          0.2(I1         )
USS          5.85902   CSS          5.58814       0% MIN          0        5%            0              0(I9         )          1.6(C23        )
CV            457.96   STD MEAN   0.0390745                                1%            0              0(I7         )          1.8(A36        )
T:MEAN=0     1.70545   PROB>|T|    0.093284      RANGE          1.8
SGN RANK          95   PROB>|S| .000137327      Q3-Q1        0.015
NUM ¬= 0          19                             MODE             0
D:NORMAL    0.452812   PROB>D         <0.01

 MISSING VALUE       .
         COUNT       2
  % COUNT/NOBS    3.17
```

TABLE D-8

```
ALL GW DATA

UNIVARIATE

VARIABLE=SI

               MOMENTS                                          QUANTILES(DEF=4)                                         EXTREMES

N                   63  SUM WGTS           63     100% MAX       15.7       99%        15.7          LOWEST    ID          HIGHEST    ID
MEAN           4.18397  SUM            263.59      75% Q3        4.72       95%     9.72195            0.08(C9        )         6.38(G23       )
STD DEV        2.61034  VARIANCE      6.81387      50% MED        3.6       90%       5.972            1.04(I15       )         6.61(E5        )
SKEWNESS       2.96119  KURTOSIS      11.1765      25% Q1         2.9       10%       2.489            1.59(C19       )         10.5(C5        )
USS            1525.31  CSS            422.46       0% MIN       0.08        5%         1.6            1.64(G21       )         15.7(A5        )
CV             62.3891  STD MEAN     0.328872                                1%        0.08            2.16(C17       )         15.7(C3        )
T:MEAN=0       12.7222  PROB>|T|       0.0001     RANGE         15.62
SGN RANK          1008  PROB>|S|       0.0001     Q3-Q1          1.82
NUM ^= 0            63                            MODE              3
D:NORMAL      0.185576  PROB>D          <0.01




GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=SI

               MOMENTS                                          QUANTILES(DEF=4)                                         EXTREMES

N                   61  SUM WGTS           61     100% MAX       10.5       99%        10.5          LOWEST    ID          HIGHEST    ID
MEAN           3.80639  SUM            232.19      75% Q3        4.56       95%       6.342            0.08(C9        )         5.93(E9        )
STD DEV        1.57335  VARIANCE      2.47542      50% MED       3.45       90%       5.878            1.04(I15       )            6(E19       )
SKEWNESS       1.23376  KURTOSIS      4.52265      25% Q1        2.88       10%       2.482            1.59(C19       )         6.38(G23       )
USS            1032.33  CSS           148.525       0% MIN       0.08        5%       1.595            1.64(G21       )         6.61(E5        )
CV             41.3343  STD MEAN     0.201447                                1%        0.08            2.16(C17       )         10.5(C5        )
T:MEAN=0       18.8953  PROB>|T|       0.0001     RANGE         10.42
SGN RANK         945.5  PROB>|S|       0.0001     Q3-Q1          1.68
NUM ^= 0            61                            MODE              3
D:NORMAL      0.117636  PROB>D          0.035

 MISSING VALUE         .
         COUNT         2
  % COUNT/NOBS      3.17
```

TABLE D-9

```
ALL GW DATA

UNIVARIATE

VARIABLE=NA

                 MOMENTS                                           QUANTILES(DEF=4)                                        EXTREMES

N                  63   SUM WGTS         63      100% MAX        40.1       99%        40.1           LOWEST     ID           HIGHEST     ID
MEAN          8.13092   SUM         512.248       75% Q3          7.3       95%       32.22              1.6(C11         )          30(E17        )
STD DEV       8.68899   VARIANCE    75.4985       50% MED        5.55       90%       20.64              1.7(A11         )        31.7(C3         )
SKEWNESS      2.54631   KURTOSIS    5.84455       25% Q1         4.01       10%         2.4            1.718(C21         )        32.3(G7         )
USS           8845.95   CSS         4680.91        0% MIN         1.6        5%      1.8144              2.2(E13         )        38.8(C9         )
CV            106.863   STD MEAN    1.09471                                  1%         1.6             2.27(C19         )        40.1(I13        )
T:MEAN=0      7.42747   PROB>|T|     0.0001       RANGE          38.5
SGN RANK         1008   PROB>|S|     0.0001       Q3-Q1          3.29
NUM ^= 0           63                             MODE            2.9
D:NORMAL     0.316912   PROB>D        <0.01
```

```
GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=NA

                 MOMENTS                                           QUANTILES(DEF=4)                                        EXTREMES

N                  61   SUM WGTS         61      100% MAX        32.3       99%        32.3           LOWEST     ID           HIGHEST     ID
MEAN          7.10407   SUM         433.348       75% Q3         6.71       95%     29.1599              1.6(C11         )        19.2(I15        )
STD DEV       6.65078   VARIANCE    44.2328       50% MED        5.32       90%          14              1.7(A11         )        21.6(A5         )
SKEWNESS       2.7529   KURTOSIS    7.50707       25% Q1        4.005       10%        2.35            1.718(C21         )          30(E17        )
USS            5732.5   CSS         2653.97        0% MIN         1.6        5%      1.7662              2.2(E13         )        31.9(C3         )
CV            93.6193   STD MEAN   0.851545                                  1%         1.6             2.27(C19         )        32.3(G7         )
T:MEAN=0      8.34256   PROB>|T|     0.0001       RANGE          30.7
SGN RANK        945.5   PROB>|S|     0.0001       Q3-Q1         2.705
NUM ^= 0           61                             MODE            2.9
D:NORMAL     0.289087   PROB>D        <0.01

 MISSING VALUE         .
         COUNT         2
  % COUNT/NOBS      3.17
```

TABLE D-10

```
ALL GW DATA

UNIVARIATE

VARIABLE=K

                MOMENTS                                        QUANTILES(DEF=4)                                      EXTREMES

N                63   SUM WGTS            63    100% MAX       110       99%         110        LOWEST     ID          HIGHEST     ID
MEAN        3.91968   SUM             246.94     75% Q3        2.8       95%       10.02          0.18(A32        )         6.6(A5         )
STD DEV     13.8681   VARIANCE       192.324     50% MED      1.27       90%       4.988          0.19(G32        )         8.5(G9         )
SKEWNESS    7.46961   KURTOSIS       57.7037     25% Q1       0.89       10%        0.38          0.26(G34        )        10.4(E17        )
USS           12892   CSS            11924.1      0% MIN      0.18        5%       0.268           0.3(A21        )        18.4(A11        )
CV          353.806   STD MEAN       1.74721                              1%        0.18          0.33(A23        )         110(I13        )
T:MEAN=0    2.24339   PROB>|T|     0.0284539     RANGE      109.82
SGN RANK       1008   PROB>|S|        0.0001     Q3-Q1        1.91
NUM ¬= 0         63                              MODE         0.77
D:NORMAL    0.39371   PROB>D           <0.01




GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=K

                MOMENTS                                        QUANTILES(DEF=4)                                      EXTREMES

N                62   SUM WGTS            62    100% MAX      18.4       99%        18.4        LOWEST     ID          HIGHEST     ID
MEAN        2.20871   SUM             136.94     75% Q3     2.6875       95%     8.21498          0.18(A32        )        5.18(C9         )
STD DEV     2.83285   VARIANCE       8.02503     50% MED      1.25       90%       4.553          0.19(G32        )         6.6(A5         )
SKEWNESS    3.79127   KURTOSIS       18.0518     25% Q1       0.88       10%        0.37          0.26(G34        )         8.5(G9         )
USS         791.988   CSS            489.527      0% MIN      0.18        5%       0.266           0.3(A21        )        10.4(E17        )
CV          128.258   STD MEAN      0.359772                              1%        0.18          0.33(A23        )        18.4(A11        )
T:MEAN=0    6.13919   PROB>|T|        0.0001     RANGE       18.22
SGN RANK      976.5   PROB>|S|        0.0001     Q3-Q1      1.8075
NUM ¬= 0         62                              MODE         0.77
D:NORMAL   0.244657   PROB>D           <0.01

 MISSING VALUE        .
         COUNT        1
  % COUNT/NOBS     1.59
```

TABLE D-11

```
ALL GW DATA

UNIVARIATE

VARIABLE=CA

                 MOMENTS                                           QUANTILES(DEF=4)                                        EXTREMES

N                 63   SUM WGTS         63     100% MAX       105      99%        105          LOWEST    ID               HIGHEST    ID
MEAN         13.3695   SUM          842.28      75% Q3       17.2      95%    85.0395         0.09(A23        )            44.6(C3         )
STD DEV      23.0722   VARIANCE    532.324      50% MED       3.3      90%      40.94         0.12(A32        )              56(E17        )
SKEWNESS     2.80255   KURTOSIS    8.02215      25% Q1       1.05      10%      0.349         0.18(C30        )            92.3(A5         )
USS            44265   CSS         33004.1       0% MIN      0.09       5%      0.184          0.2(E21        )           102.3(C5         )
CV           172.573   STD MEAN    2.90682                              1%       0.09         0.32(C34        )             105(G23        )
T:MEAN=0     4.59937   PROB>|T|     0.0001     RANGE       104.91
SGN RANK        1008   PROB>|S|     0.0001     Q3-Q1        16.15
NUM ¬= 0          63                           MODE           3.1
D:NORMAL    0.284289   PROB>D        <0.01




GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=CA

                 MOMENTS                                           QUANTILES(DEF=4)                                        EXTREMES

N                 60   SUM WGTS         60     100% MAX        56      99%         56          LOWEST    ID               HIGHEST    ID
MEAN         9.04467   SUM          542.68      75% Q3      10.05      95%     42.305         0.09(A23        )            32.8(E1         )
STD DEV      12.5867   VARIANCE    158.425      50% MED       3.2      90%      28.74         0.12(A32        )            38.6(G7         )
SKEWNESS     2.00528   KURTOSIS    3.72424      25% Q1       0.93      10%      0.341         0.18(C30        )            42.5(E3         )
USS          14255.4   CSS         9347.06       0% MIN      0.09       5%      0.181          0.2(E21        )            44.6(C3         )
CV           139.161   STD MEAN    1.62493                              1%       0.09         0.32(C34        )              56(E17        )
T:MEAN=0     5.56617   PROB>|T|     0.0001     RANGE        55.91
SGN RANK         915   PROB>|S|     0.0001     Q3-Q1         9.12
NUM ¬= 0          60                           MODE           3.1
D:NORMAL    0.241907   PROB>D        <0.01

 MISSING VALUE        .
         COUNT        3
  % COUNT/NOBS     4.76
```

TABLE D-12

```
ALL GW DATA

UNIVARIATE

VARIABLE=BA

              MOMENTS                                        QUANTILES(DEF=4)                                         EXTREMES

N                63  SUM WGTS           63       100% MAX      21.1      99%         21.1          LOWEST     ID           HIGHEST     ID
MEAN       0.581476  SUM            36.633        75% Q3       0.38      95%        1.144               0(I9       )          0.83(E36      )
STD DEV     2.65755  VARIANCE      7.06257        50% MED      0.05      90%         0.83               0(I5       )          0.92(G1       )
SKEWNESS    7.66629  KURTOSIS      59.9947        25% Q1       0.01      10%            0               0(I1       )           1.2(C36      )
USS         459.181  CSS           437.879         0% MIN         0       5%            0               0(G36      )           2.4(I17      )
CV          457.035  STD MEAN      0.33482                                1%            0               0(G32      )          21.1(I13      )
T:MEAN=0    1.73668  PROB>|T|    0.0874094       RANGE         21.1
SGN RANK        770  PROB>|S|       0.0001       Q3-Q1         0.37
NUM -= 0         55                              MODE          0.01
D:NORMAL   0.413402  PROB>D          <0.01
```

```
GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=BA

              MOMENTS                                        QUANTILES(DEF=4)                                         EXTREMES

N                62  SUM WGTS           62       100% MAX       2.4      99%          2.4          LOWEST     ID           HIGHEST     ID
MEAN       0.250532  SUM            15.533        75% Q3       0.38      95%     0.906499               0(I9       )          0.83(E34      )
STD DEV    0.406486  VARIANCE      0.16523        50% MED      0.05      90%         0.83               0(I5       )          0.83(E36      )
SKEWNESS    2.95361  KURTOSIS      12.1071        25% Q1       0.01      10%            0               0(I1       )          0.92(G1       )
USS         13.9706  CSS           10.0791         0% MIN         0       5%            0               0(G36      )           1.2(C36      )
CV          162.249  STD MEAN    0.0516237                                1%            0               0(G32      )           2.4(I17      )
T:MEAN=0    4.85305  PROB>|T|       0.0001       RANGE          2.4
SGN RANK      742.5  PROB>|S|       0.0001       Q3-Q1         0.37
NUM -= 0         54                              MODE          0.01
D:NORMAL   0.268836  PROB>D          <0.01

 MISSING VALUE        .
         COUNT        1
  % COUNT/NOBS     1.59
```

TABLE 5-13

```
ALL GW DATA

UNIVARIATE

VARIABLE=SR

                  MOMENTS                                             QUANTILES(DEF=4)                                        EXTREMES

N                  63   SUM WGTS          63     100% MAX       0.61       99%         0.61         LOWEST    ID           HIGHEST    ID
MEAN         0.113937   SUM            7.178      75% Q3        0.14       95%        0.508              0(G32       )        0.48(I13      )
STD DEV      0.159519   VARIANCE   0.0254463      50% MED       0.04       90%     0.399999              0(G13       )         0.5(C36      )
SKEWNESS      1.79798   KURTOSIS     2.39201      25% Q1        0.01       10%       0.0014              0(E21       )        0.51(G23      )
USS            2.3955   CSS          1.57767       0% MIN          0        5%            0              0(C19       )         0.6(A5       )
CV            140.007   STD MEAN   0.0200975                                1%            0          0.001(A23       )        0.61(C5       )
T:MEAN=0      5.66919   PROB>|T|      0.0001      RANGE         0.61
SGN RANK          885   PROB>|S|      0.0001      Q3-Q1         0.13
NUM ^= 0           59                             MODE          0.01
D:NORMAL     0.243428   PROB>D         <0.01
```

```
GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=SR

                  MOMENTS                                             QUANTILES(DEF=4)                                        EXTREMES

N                  61   SUM WGTS          61     100% MAX       0.51       99%         0.51         LOWEST    ID           HIGHEST    ID
MEAN        0.0978361   SUM            5.968      75% Q3       0.135       95%        0.474              0(G32       )        0.37(C7       )
STD DEV      0.134134   VARIANCE    0.017992      50% MED       0.03       90%     0.321999              0(G13       )        0.42(G7       )
SKEWNESS      1.81087   KURTOSIS     2.54949      25% Q1        0.01       10%       0.0012              0(E21       )        0.48(I13      )
USS            1.6634   CSS          1.07952       0% MIN          0        5%            0              0(C19       )         0.5(C36      )
CV            137.101   STD MEAN   0.0171741                                1%            0          0.001(A23       )        0.51(G23      )
T:MEAN=0      5.69672   PROB>|T|      0.0001      RANGE         0.51
SGN RANK        826.5   PROB>|S|      0.0001      Q3-Q1        0.125
NUM ^= 0           57                             MODE          0.01
D:NORMAL     0.237937   PROB>D         <0.01

  MISSING VALUE       .
          COUNT       2
   % COUNT/NOBS    3.17
```

TABLE D-14

```
ALL GW DATA

UNIVARIATE

VARIABLE=FE

              MOMENTS                                      QUANTILES(DEF=4)                                     EXTREMES

N                63   SUM WGTS          63    100% MAX     112.8      99%      112.8         LOWEST    ID          HIGHEST    ID
MEAN        3.00789   SUM          189.497     75% Q3       0.03      95%    10.8238             0(G36     )       0.67(G30      )
STD DEV     16.0668   VARIANCE     258.144     50% MED     0.013      90%   0.191999             0(G34     )       0.84(I1       )
SKEWNESS    6.08077   KURTOSIS     38.5313     25% Q1          0      10%          0             0(G28     )      13.32(C19      )
USS         16574.9   CSS          16004.9      0% MIN         0       5%          0             0(G23     )       60.6(G7       )
CV          534.157   STD MEAN     2.02423                             1%          0             0(G1      )      112.8(G21      )
T:MEAN=0    1.48594   PROB>|T|    0.142362    RANGE        112.8
SGN RANK        495   PROB>|S|      0.0001    Q3-Q1         0.03
NUM ^= 0         44                           MODE             0
D:NORMAL   0.506047   PROB>D         <0.01




GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=FE

              MOMENTS                                      QUANTILES(DEF=4)                                     EXTREMES

N                61   SUM WGTS          61    100% MAX     13.32      99%      13.32          LOWEST    ID          HIGHEST    ID
MEAN       0.263885   SUM           16.097     75% Q3      0.025      95%   0.624997             0(G36     )       0.15(C9       )
STD DEV     1.70508   VARIANCE     2.90731     50% MED     0.012      90%   0.109999             0(G34     )       0.22(G9       )
SKEWNESS    7.73414   KURTOSIS     60.1681     25% Q1          0      10%          0             0(G28     )       0.67(G30      )
USS         178.686   CSS          174.438      0% MIN         0       5%          0             0(G23     )       0.84(I1       )
CV          646.146   STD MEAN    0.218314                             1%          0             0(G1      )      13.32(C19      )
T:MEAN=0    1.20874   PROB>|T|    0.231503    RANGE        13.32
SGN RANK      451.5   PROB>|S|      0.0001    Q3-Q1        0.025
NUM ^= 0         42                           MODE             0
D:NORMAL   0.461086   PROB>D         <0.01

  MISSING VALUE        .
          COUNT        2
   % COUNT/NOBS     3.17
```

WELL D-15

```
ALL GW DATA

UNIVARIATE

VARIABLE=MG

               MOMENTS                                           QUANTILES(DEF=4)                                         EXTREMES

N                 63  SUM WGTS          63      100% MAX        39.9      99%         39.9         LOWEST     ID            HIGHEST     ID
MEAN         1.71397  SUM           107.98       75% Q3          1.5      95%      4.14599              0(C19       )           3.4(A5       )
STD DEV      5.05264  VARIANCE     25.5292       50% MED        0.66      90%         3.05           0.03(A11       )          3.53(G9       )
SKEWNESS      7.2094  KURTOSIS     54.8558       25% Q1         0.35      10%        0.174           0.14(C21       )           4.3(G21      )
USS          1767.88  CSS          1582.81        0% MIN           0       5%         0.14           0.14(G28       )           7.6(I15      )
CV           294.792  STD MEAN    0.636573                                 1%            0           0.15(A36       )          39.9(G7       )
T:MEAN=0     2.69249  PROB>|T| 0.00910809       RANGE           39.9
SGN RANK       976.5  PROB>|S|      0.0001       Q3-Q1           1.15
NUM ¬= 0          62                            MODE            0.34
D:NORMAL    0.367221  PROB>D         <0.01
```

TABLE D-16

```
ALL GW DATA

UNIVARIATE

VARIABLE=DO

                MOMENTS                                       QUANTILES(DEF=4)                                 EXTREMES

N                 63    SUM WGTS          63     100% MAX         9       99%           9           LOWEST     ID           HIGHEST    ID
MEAN         5.90159    SUM            371.8      75% Q3        7.5       95%         8.6              1.5(G7        )           8.6(A32       )
STD DEV      2.04722    VARIANCE     4.19113      50% MED         6       90%        8.52              1.7(G23       )           8.6(C17       )
SKEWNESS   -0.417779    KURTOSIS   -0.753336      25% Q1        4.5       10%        2.64                2(E30       )           8.6(C34       )
USS          2454.06    CSS           259.85       0% MIN       1.5        5%        2.08              2.4(C9        )           8.7(G34       )
CV           34.6894    STD MEAN    0.257926                               1%         1.5              2.5(I5        )             9(I17       )
T:MEAN=0     22.8809    PROB>|T|      0.0001      RANGE         7.5
SGN RANK        1008    PROB>|S|      0.0001      Q3-Q1           3
NUM ^= 0          63                              MODE          8.6
D:NORMAL   0.0779735    PROB>D         >0.15
```

TABLE D-17

```
ALL GW DATA

UNIVARIATE

VARIABLE=TEMP

                 MOMENTS                                   QUANTILES(DEF=4)                                      EXTREMES

N                 63   SUM WGTS           63     100% MAX        22       99%         22          LOWEST    ID            HIGHEST    ID
MEAN         19.4762   SUM              1227      75% Q3         21       95%         21             15(I5        )          21(G23       )
STD DEV      1.71903   VARIANCE      2.95507      50% MED        20       90%         21             15(A3        )          21(G28       )
SKEWNESS   -0.838777   KURTOSIS    -0.196702      25% Q1         18       10%         17             16(I9        )          21(G30       )
USS          24080.5   CSS           183.214       0% MIN        15        5%       16.2             17(I17       )          21(G36       )
CV           8.82632   STD MEAN     0.216578                                1%         15             17(I15       )          22(C19       )
T:MEAN=0     89.9271   PROB>|T|       0.0001      RANGE           7
SGN RANK        1008   PROB>|S|       0.0001      Q3-Q1           3
NUM ¬= 0          63                              MODE           21
D:NORMAL    0.225007   PROB>D          <0.01
```

TABLE D-18

```
ALL GW DATA

UNIVARIATE

VARIABLE=COND

                  MOMENTS                                            QUANTILES(DEF=4)                                        EXTREMES

N                   63   SUM WGTS          63     100% MAX        530        99%          530        LOWEST      ID          HIGHEST     ID
MEAN           93.1111   SUM             5866      75% Q3         100        95%      431.999            15(C21       )            310(E17       )
STD DEV        113.916   VARIANCE     12976.9      50% MED         47        90%          262            20(E13       )            360(A5        )
SKEWNESS       2.56577   KURTOSIS     6.29069      25% Q1          31        10%           25            20(C15       )            450(I13       )
USS            1350756   CSS           804566       0% MIN         15         5%           20            20(C11       )            500(G21       )
CV             122.344   STD MEAN     14.3521                                 1%           15            20(A21       )            530(G7        )
T:MEAN=0       6.48764   PROB>|T|      0.0001     RANGE           515
SGN RANK          1008   PROB>|S|      0.0001     Q3-Q1            69
NUM ^= 0            63                            MODE             30
D:NORMAL      0.262925   PROB>D         <0.01



GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=COND

                  MOMENTS                                            QUANTILES(DEF=4)                                        EXTREMES

N                   60   SUM WGTS          60     100% MAX        360        99%          360        LOWEST      ID          HIGHEST     ID
MEAN              73.1   SUM             4386      75% Q3       88.75        95%          269            15(C21       )            170(C5        )
STD DEV        70.9188   VARIANCE     5029.48      50% MED       45.5        90%        139.8            20(E13       )            250(C9        )
SKEWNESS       2.44703   KURTOSIS     6.24624      25% Q1       30.25        10%           25            20(C15       )            270(G23       )
USS             617356   CSS           296739       0% MIN         15         5%           20            20(C11       )            310(E17       )
CV             97.0162   STD MEAN     9.15558                                 1%           15            20(A21       )            360(A5        )
T:MEAN=0        7.9842   PROB>|T|      0.0001     RANGE           345
SGN RANK           915   PROB>|S|      0.0001     Q3-Q1          58.5
NUM ^= 0            60                            MODE             30
D:NORMAL      0.234099   PROB>D         <0.01

 MISSING VALUE        .
         COUNT        3
  % COUNT/NOBS     4.76
```

TABLE D-19

```
ALL GW DATA

UNIVARIATE

VARIABLE=PH

              MOMENTS                                          QUANTILES(DEF=4)                                    EXTREMES

N                  63   SUM WGTS          63    100% MAX      7.17      99%        7.17        LOWEST    ID           HIGHEST    ID
MEAN          5.31825   SUM           335.05     75% Q3       6.04      95%        6.85          2.38(E7        )       6.64(C9        )
STD DEV      0.974551   VARIANCE     0.94975     50% MED      5.39      90%       6.576          2.91(E13       )       6.81(A5        )
SKEWNESS    -0.504402   KURTOSIS    0.467874     25% Q1       4.67      10%       4.162          3.45(G36       )       6.86(C5        )
USS           1840.77   CSS          58.8845      0% MIN      2.38       5%       3.536          3.88(E9        )       6.95(A3        )
CV            18.3246   STD MEAN    0.122782                             1%        2.38          4.07(E30       )       7.17(E19       )
T:MEAN=0      43.3146   PROB>|T|      0.0001     RANGE        4.79
SGN RANK         1008   PROB>|S|      0.0001     Q3-Q1        1.37
NUM ^= 0           63                            MODE          5.4
D:NORMAL    0.0567663   PROB>D         >0.15
```

```
GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=PH

              MOMENTS                                          QUANTILES(DEF=4)                                    EXTREMES

N                  62   SUM WGTS          62    100% MAX      7.17      99%        7.17        LOWEST    ID           HIGHEST    ID
MEAN          5.36565   SUM           332.67     75% Q3     6.0575      95%      6.8525          2.91(E13       )       6.64(C9        )
STD DEV       0.90637   VARIANCE    0.821507     50% MED     5.395      90%       6.582          3.45(G36       )       6.81(A5        )
SKEWNESS     -0.22275   KURTOSIS   -0.195682     25% Q1      4.685      10%       4.192          3.88(E9        )       6.86(C5        )
USS            1835.1   CSS          50.1119      0% MIN      2.91       5%      3.9085          4.07(E30       )       6.95(A3        )
CV            16.8921   STD MEAN    0.115109                             1%        2.91          4.15(E23       )       7.17(E19       )
T:MEAN=0      46.6135   PROB>|T|      0.0001     RANGE        4.26
SGN RANK        976.5   PROB>|S|      0.0001     Q3-Q1      1.3725
NUM ^= 0           62                            MODE          5.4
D:NORMAL    0.0459026   PROB>D         >0.15

 MISSING VALUE        .
         COUNT        1
  % COUNT/NOBS     1.59
```

TABLE D-20

```
ALL GW DATA

UNIVARIATE

VARIABLE=MV

               MOMENTS                                         QUANTILES(DEF=4)                                    EXTREMES

N                   63   SUM WGTS          63    100% MAX       683      99%         683       LOWEST    ID           HIGHEST    ID
MEAN           513.016   SUM            32320     75% Q3        567      95%       660.2          172(G7        )          642(I7        )
STD DEV        94.4788   VARIANCE     8926.24     50% MED       536      90%         630          264(G9        )          653(I5        )
SKEWNESS     -0.971987   KURTOSIS     2.00051     25% Q1        452      10%       392.8          345(C19       )          662(I1        )
USS           17134100   CSS           553427      0% MIN       172       5%       349.8          369(G21       )          668(G17       )
CV             18.4163   STD MEAN     11.9032                             1%         172          379(C5        )          683(C13       )
T:MEAN=0        43.099   PROB>|T|      0.0001     RANGE         511
SGN RANK          1008   PROB>|S|      0.0001     Q3-Q1         115
NUM ^= 0            63                            MODE          541
D:NORMAL      0.135181   PROB>D         <0.01
```

```
GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=MV

               MOMENTS                                         QUANTILES(DEF=4)                                    EXTREMES

N                   62   SUM WGTS          62    100% MAX       683      99%         683       LOWEST    ID           HIGHEST    ID
MEAN           518.516   SUM            32148     75% Q3        568      95%      660.65          264(G9        )          642(I7        )
STD DEV        84.4713   VARIANCE      7135.4     50% MED       537      90%         631          345(C19       )          653(I5        )
SKEWNESS     -0.470591   KURTOSIS    0.351507     25% Q1        455      10%       396.4          369(G21       )          662(I1        )
USS           17104516   CSS           435259      0% MIN       264       5%       370.5          379(C5        )          668(G17       )
CV              16.291   STD MEAN     10.7279                             1%         264          392(C7        )          683(C13       )
T:MEAN=0       48.3336   PROB>|T|      0.0001     RANGE         419
SGN RANK         976.5   PROB>|S|      0.0001     Q3-Q1         113
NUM ^= 0            62                            MODE          541
D:NORMAL      0.127929   PROB>D         0.012

 MISSING VALUE         .
         COUNT         1
  % COUNT/NOBS      1.59
```

TABLE D-21

```
ALL GW DATA

UNIVARIATE

VARIABLE=TC

                MOMENTS                                   QUANTILES(DEF=4)                                  EXTREMES

N                63   SUM WGTS          63     100% MAX       245      99%         245         LOWEST    ID            HIGHEST    ID
MEAN        20.8587   SUM           1314.1      75% Q3       26.7      95%     69.5799              0(G17      )          60.5(E3      )
STD DEV     34.4365   VARIANCE     1185.87      50% MED      10.4      90%     51.8999              0(G13      )          63.5(C5      )
SKEWNESS    4.83086   KURTOSIS      29.296      25% Q1          5      10%         2.4              0(E36      )          71.1(A5      )
USS          100935   CSS          73524.2       0% MIN         0       5%           0              0(E32      )            93(G7      )
CV          165.094   STD MEAN     4.33859                              1%           0              1(C32      )           245(G21     )
T:MEAN=0    4.80772   PROB>|T|      0.0001      RANGE         245
SGN RANK        885   PROB>|S|      0.0001      Q3-Q1        21.7
NUM ^= 0         59                             MODE            0
D:NORMAL   0.272352   PROB>D         <0.01
```

TABLE D-22

```
ALL GW DATA

UNIVARIATE

VARIABLE=TOC

                MOMENTS                                             QUANTILES(DEF=4)                                    EXTREMES

N                 63  SUM WGTS          63    100% MAX        225       99%          225          LOWEST    ID           HIGHEST    ID
MEAN         7.72381  SUM            486.6     75% Q3           6       95%        20.32              0(I15      )          16(I1        )
STD DEV      28.7192  VARIANCE     824.791     50% MED          1       90%        12.02              0(G34      )          18(C21       )
SKEWNESS     7.24187  KURTOSIS     55.1153     25% Q1           0       10%            0              0(G32      )        20.9(C17       )
USS          54895.4  CSS            51137      0% MIN          0        5%            0              0(G28      )          45(G7        )
CV           371.827  STD MEAN     3.61828                               1%            0              0(G23      )         225(G21       )
T:MEAN=0     2.13467  PROB>|T|   0.0367488     RANGE          225
SGN RANK         315  PROB>|S|      0.0001     Q3-Q1            6
NUM ¬= 0          35                           MODE             0
D:NORMAL    0.393987  PROB>D         <0.01
```

```
GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=TOC

                MOMENTS                                             QUANTILES(DEF=4)                                    EXTREMES

N                 62  SUM WGTS          62    100% MAX         45       99%           45          LOWEST    ID           HIGHEST    ID
MEAN         4.21935  SUM            261.6     75% Q3         5.7       95%         17.7              0(I15      )        12.1(C30       )
STD DEV      7.20503  VARIANCE     51.9124     50% MED        0.7       90%        11.63              0(G34      )          16(I1        )
SKEWNESS     3.46258  KURTOSIS     16.4598     25% Q1           0       10%            0              0(G32      )          18(C21       )
USS          4270.44  CSS          3166.66      0% MIN          0        5%            0              0(G28      )        20.9(C17       )
CV           170.761  STD MEAN    0.915039                               1%            0              0(G23      )          45(G7        )
T:MEAN=0     4.61112  PROB>|T|      0.0001     RANGE           45
SGN RANK       297.5  PROB>|S|      0.0001     Q3-Q1          5.7
NUM ¬= 0          34                           MODE             0
D:NORMAL    0.279068  PROB>D         <0.01

 MISSING VALUE        .
         COUNT        1
  % COUNT/NOBS     1.59
```

TABLE D-23

```
ALL GW DATA

UNIVARIATE

VARIABLE=TIC

               MOMENTS                                        QUANTILES(DEF=4)                                           EXTREMES

N                   63   SUM WGTS          63     100% MAX        71.1       99%         71.1          LOWEST     ID             HIGHEST     ID
MEAN           13.1349   SUM            827.5      75% Q3           17       95%      54.9199               0(G36      )              48(G7      )
STD DEV        16.3409   VARIANCE     267.024      50% MED           7       90%        38.64               0(G17      )            48.6(E3      )
SKEWNESS       1.94032   KURTOSIS     3.35109      25% Q1            3       10%            0               0(G13      )            56.5(C3      )
USS            27424.6   CSS          16555.5       0% MIN           0        5%            0               0(E36      )            63.5(C5      )
CV             124.408   STD MEAN     2.05875                                 1%            0               0(E32      )            71.1(A5      )
T:MEAN=0       6.38003   PROB>|T|      0.0001      RANGE          71.1
SGN RANK           770   PROB>|S|      0.0001      Q3-Q1            14
NUM ¬= 0            55                             MODE              0
D:NORMAL      0.226862   PROB>D         <0.01
```

```
GW DATA WITH 3-SIGMA VALUES MISSING

UNIVARIATE

VARIABLE=TIC

               MOMENTS                                        QUANTILES(DEF=4)                                           EXTREMES

N                   61   SUM WGTS          61     100% MAX        56.5       99%         56.5          LOWEST     ID             HIGHEST     ID
MEAN            11.359   SUM            692.9      75% Q3        14.55       95%      47.0999               0(G36      )            38.1(E5      )
STD DEV        13.2076   VARIANCE     174.441      50% MED           7       90%      35.5799               0(G17      )              39(C9      )
SKEWNESS       1.80874   KURTOSIS     2.82856      25% Q1            3       10%            0               0(G13      )              48(G7      )
USS            18337.1   CSS          10466.5       0% MIN           0        5%            0               0(E36      )            48.6(E3      )
CV             116.274   STD MEAN     1.69106                                 1%            0               0(E32      )            56.5(C3      )
T:MEAN=0       6.71708   PROB>|T|      0.0001      RANGE          56.5
SGN RANK         715.5   PROB>|S|      0.0001      Q3-Q1         11.55
NUM ¬= 0            53                             MODE              0
D:NORMAL      0.213108   PROB>D         <0.01

 MISSING VALUE        .
         COUNT        2
  % COUNT/NOBS     3.17
```

APPENDIX E

A SUMMARY OF THE CHEMICALLY ANOMALOUS WELLS

(INCLUDING TABLES E1 AND E2)

TABLE E1. Five most extreme values: Including all wells outside of three standard deviations from the mean of the calculated distributions.*

F	A-19	C-9	E-17	E-3	C-15
(ppm F)	0.5	0.42	0.4	0.4	0.4
Cl	I-13	G-7	I-15	G-21	I-17
(ppm Cl)	152	45	35	33	16.3
NO_3	I-15	E-32	I-1	G-34	G-15
(ppm NO_3)	28	25.2	22	18.2	16.8
SO_4	E-17	C-5	G-9	C-3	A-5
(ppm SO_4)	111	24	21	16.4	15.8
PO_4	E-5**	E-17**	A-36	C-23	I-1
(ppm PO_4)	15.1	13.7	1.8	1.6	0.2
H_2SiO_3	C-3	A-5	C-5	E-5	G-23
(ppm Si)	15.7	15.7	10.5	6.61	6.38
Na	I-13	C-9	G-7	C-3	E-17
(ppm Na)	40.1	38.8	32.3	31.9	30

* These values are underlined. See Appendix D for distributions.

** Detected only as phosphorus by plasma emission, presence of phosphate could not be confirmed.

TABLE E1. Cont'd

A Summary of Extreme Values: Including all wells outside of three standard deviations from the mean of the calculated distributions.

K	I-13	A-11	E-17	G-9	A-5
(ppm K)	110	18.4	10.4	8.5	6.6
Ca	G-23	C-5	A-5	E-17	C-3
(ppm Ca)	105	102	92	56	44
Ba	I-13	I-17	C-36	G-1	E-36
(ppm Ba)	21.1	2.4	1.2	0.92	0.83
Sr	C-5	A-5	G-23	C-36	I-13
(ppm Sr)	0.61	0.6	0.51	0.5	0.48
Fe	G-21	G-7	C-19	I-1	G-30
(ppm Fe)	113	61	13	0.8	0.7
Mg	G-7	I-15	C-19	I-1	G-30
(ppm Mg)	40	7.6	4.3	3.51	3.4
Do	G-7	G-23	E-30	C-9	I-5
(ppm O_2)	1.5	1.7	2.0	2.4	2.5

TABLE E1. Cont'd

A Summary of Extreme Values: Including all wells outside of three standard deviations from the mean of the calculated distributions.

Temp - not a continuous (variable) function

Conductivity	G-7	G-21	I-13	A-5	E-17
(mho/cm)	530	500	450	360	310
pH (low)	E-7	E-13	G-36	E-9	E-30
	2.38	2.91	3.45	3.88	4.07
(high)	E-19	A-3	C-5	A-5	C-9
	7.17	6.95	6.86	6.81	6.64
Mv	G-7	G-9	C-19	G-21	C-5
(millivolts vs. NHE)	172	264	345	369	379
TOC	G-21	G-7	C-17	C-21	I-1
(ppm C)	225	45	20.9	18	16
TIC	A-5	C-5	C-3	E-3	G-7
(ppm C)	71	64	57	49	48

TABLE E2
LOW-LEVEL COUNTING SUMMARY: COMPARISON WITH PREDICTIONS BASED ON TRANSPORT CHEMISTRY[a]

WELL	RADIONUCLIDES PREDICTED	RADIONUCLIDES DETECTED
A-1	None	None[b]
A-3	None	Sr-90, Pu-238[c]
A-9	None	None
C-1	None	Sr-90, Pu-238/239[c]
C-3	Cs-137	Pu-238
C-13	None	None[b]
C-15	None	Pu-238/239[d]
C-17	None	None
E-13	None	None
E-17	Pu(IV), Pu(VI) Sr-90, Cs-137	Cs-137, Co-60
E-19	None	None[b]
G-17	None	None[b]
G-21	Pu(IV), Sr-90 Cs-137	Pu-238/239, Cs-137 Sr-90
G-34	None	None[b]
I-1	None	None[b]
I-5	None	None[b]
I-7	None	None[b]
I-9	None	Pu-238[d]
I-13	Sr-90, Cs-137	Pu-238, Cs-137, Sr-90
I-17	None	Pu-238

a. Predictions are based on the 3-sigma criterion discussed in the text. See also Tables E-1 and D-1 and Appendix E.

b. Low-level alpha and gamma analyses, but Sr-90 was not measured.

c. Contamination apparently mobilized by complexants from equipment decontamination facility.

d. Contamination known to be related to solvent spill (see text, Operation's History).

APPENDIX E

A SUMMARY OF THE CHEMICALLY ANOMALOUS WELLS

(INCLUDING TABLES E1 AND E2)

SUMMARY OF SUSPECT WELLS

A-5 This well is anomalously high (more than 3 s.d. from the mean of all wells) in calcium carbonate, silica and strontium, probably all from natural sources. As described previously, calcium mobilizes Sr-90, and free carbonate (1.8 x 10^{-6} M CO_3^{2-} calculated for well A-5) could favor the formation of the mobile hydroxycarbonate species with any available Pu(VI). The hydroxycarbonate of Pu(VI) is favored over the insoluble Pu(VI) carbonate by 4 x 10^4. This places Well A-5 among the ten monitoring wells that are most susceptible to this complexation.

The most favorable oxidation states for plutonium in Well A-5 appear to be Pu(V) and Pu(III). Pu(V) is favored by over 10^5 with respect to Pu(III), and Pu(VI) is about 10^{10} more stable than Pu(IV) under the prevailing conditions (pH = 6.8, E_H = 0.5 volts). Pu(V), which is not well complexed, is about 10^7 times more stable than the free Pu(VI), but Pu(VI) hydroxycarbonate is also favored over PuO_2^{2+} by 8 x 10^{11}. The net increase in solubility of plutonium due to the hydroxycarbonate complex is nearly 10^5 with respect to Pu (V), which is the most thermodynamically stable form of free plutonium in this system.

In a soil-water equilibrium, the dissolution of plutonium is reduced by a factor that can be related to the operating distribution coefficient (usually 10^3 to 10^4). The predicted concentration of plutonium for a homogeneous system that is in equilibrium with solid plutonium oxide, along with water of the same composition as that in well A-5, and typical burial ground soil, is on the order of the solubility of plutonium in that water. This is because, as described above, most of the soluble plutonium would be present as the non-exchangeable (anionic) hydroxycarbonate complex.

Well A-5 is also among the top five in sulfate (15.8 ppm), potassium (6.6 ppm), pH (6.81), and magnesium (3.4 ppm) and is a "suspect" for its high conductivity (360 µmho/cm). These observations indicate the influence of a nearby trench and clearly mark A-5 as a well with the potential of being contaminated by fission product activity from that trench. However, radiochemical data on Well A-5 show low values (<60 pCi/L since 1976) for non-volatile beta-gamma activity, with a negative year-to-year slope (-5.8 pCi/L since 1976). This well is probably stabilizing, and it is unlikely to show a significant increase in fission products in the near future. The alpha activity in this well is 6th highest of all burial ground wells (4 pCi/L) and is increasing slightly, at the rate of 0.24 pCi/L-year.

A low-level pulse height analysis, a gamma scan, and Sr-90 analysis should all be performed on Well A-5 to determine the source of the activity. Regular monitoring of this well should continue along with the other "suspect" wells.

A-11 Well A-11 is mentioned here because it contains 18.4 ppm of potassium. This concentration is sufficiently large to exclude Well A-11 from a modified distribution of potassium values for all wells. The modified distribution was obtained, as described previously, by first excluding all wells that deviated by three standard deviations or more from the mean of the best* potassium values for all wells.

Potassium, being poorly hydrated and about the same size as cesium, competes relatively well with Cesium-137 for ion exchange sites on the soil. In situations of slow kinetics (or non-equilibrium) Cesium-137 might be moved at very low concentrations and could reach the groundwater at trace levels. Ultra-trace analysis of radioisotopes has not been performed yet, but the gross alpha and non-volatile beta-gamma measurements indicate no activity above background.

A-19 Well A-19 contains more fluoride than any other monitoring well (0.5 ppm). As described in the section on fluoride, even this small amount may be able to complex Plutonium (VI). The monitoring results of this well are also negative for gross alpha and non-volatile beta-gamma. Low-level analysis has not been performed.

The anionic fluoride complex ($PuO_2F_3^-$) is favored in this well by a factor of 1100 over the free plutonium (VI) cation. However, the hydroxycarbonate complex may be more important since it is favored in this well by roughly 10^{10} over the free Plutonium (VI) cation. Plutonium(V) is the most stable species at the pH and reduction potential of Well A-19, but 10% of Pu(III) would also be expected. The free Pu(VI) concentration would be on the order of one part per million of the dissolved Pu(V). Complexation of Pu(VI) mobilizes some plutonium, but soil adsorption and hydrolysis of Pu(III) have the opposite effect. Therefore, only traces of Pu(VI) complexes would be likely to remain in solution.

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* Best values denote averages of all available data for each individual well.

A-36 This well is considered a radionuclide migration "suspect" because 1.8 ppm of phosphate were detected there. (This concentration is also more than three standard deviations higher than the mean of the modified distribution.) Phosphate at this concentration complexes Pu(IV) effectively, the ratio of the tetra-anion complex to free Pu(IV) being 1.6×10^{19} at equilibrium. Pu(IV) is not stable in this well with respect to Pu(III), except at very small concentration ratios (1 part per million). However, the oxidized plutonium states are only slightly favored over Pu(IV) and Pu(III) in this well, and phosphate would shift the equilibrium toward Pu(IV).

If this water were at a source of plutonium, the complexation reaction would be limited by the phosphate concentration. During transport, the plutonium concentration would diminish through dilution, but soil adsorption would have little impact on such a strong anionic complex.

Therefore, well A-36 should be analyzed carefully for traces of plutonium.

C-3 This well was high in silica, sodium, and inorganic carbon. It was also among the highest in sulfate and calcium. Of these, the most significant are the high sodium and inorganic carbon, both of which exceeded the mean (of the modified distribution) by over three standard deviations. The sodium, and probably most of the carbonate, came from a man-made source (trench).

The sodium concentration of 31.9 ppm could be enough to enable a detectable quantity of Cs-137 to reach the groundwater under non-equilibrium conditions. However, the low-level count performed on a water sample from C-3 indicates that this has not happened so far. Since equilibrium strictly favors the irreversible retention of cesium by the soil column, very little, if any, Cesium-137 would be expected to emerge later. Traces of Sr-90 could appear later, however, due to gradual elution by sodium and calcium. Low-level analysis detected no Strontium-90 or Cesium-137 in Well C-3 (<6 pci/L and <8 pCi/L respectively).

Traces of Pu-238 (2±1 pCi) were detected in Well C-3, but no Pu-239 was detectable. This contamination may be due to the remaining traces of contamination from an equipment decontamination station that was once located in the northwest corner of the burial ground. Although the exact location of the station is not known, it is believed to have been just north of grid Well A-3 (see Section V, Burial Ground History).

The high pH of Well C-3 (6.14) favors the oxidized states of plutonium, but the oxidation-reduction potential is low. This means that Pu(V) is the most thermodynamically stable state for plutonium. The calculated equilibrium constant for hydroxycarbonate complexation of Pu(VI) is about 10^{10}, but so is the ratio of Pu(V)/Pu(VI). Plutonium would, therefore, move roughly as fast as the free Pu(V) in this system.

C-5 Total inorganic carbon, calcium, and strontium were high in Well C-5. Sulfate and silica also exceeded the criteria for suspecting chemical contamination. In addition, the pH of Well C-5 was the third highest of all wells.

These values lead one to expect Sr-90 to appear in Well C-5. Unfortunately, C-5 has not been analyzed for trace levels of this radionuclide. Traces of other cationic radionuclides could be mobilized by the high calcium concentration, but the most important variable is probably the large amount of free carbonate, which could theoretically mobilize Plutonium (VI). The likelihood of hydroxycarbonate plutonium complexation is about two orders of magnitude lower than for Well A-5 because the redox potential of Well C-5 is about 120 mv lower.

C-9 Monitoring Well C-9 is abnormally high in sodium for the lithology of the SRP Burial Ground, indicating that it, too, contains chemical contamination from the waste trenches. This concentration (1.7×10^{-3} M) is too low to significantly enhance the overall mobility of Cs-137 or even Sr-90 by the soil (see Sodium). However, this concentration could shift the surface potential of the soil positively enough to allow an extremely low concentration of Cs-137 to reach the groundwater. This scenario is not highly probable, but does have credibility, since similar effects seem to have occurred in wells E-17, G-21 and I-13. Well C-5 should therefore be analyzed carefully for traces of Cs-137 and Sr-90. Well C-9 is also more alkaline (pH 6.64) than all but four other wells and it contained 0.42 ppm of fluoride. Pu (VI) could therefore be mobilized at very low concentrations around this well.

C-19 Well C-19 contains 13 ppm of iron, presumably as Fe(II) rather than complexed Fe(III). The oxidation potential of this well was also low (0.345 V vs. SHE) as was the dissolved oxygen content (2.7 ppm at 22°C). Interestingly, the organic carbon content was below the detection limit (about 1 ppm in most determinations).

This well should be considered a candidate for low-level analysis, and is likely to contain traces of Sr-90 and Cs-137.

C-23 This well contains 1.6 ppm of phosphate, which could indicate a potential source of Pu (IV). Again, calculations (see Phosphate), predict a limited potential for this transport mechanism, but traces of plutonium might be detected in this monitoring well. (See well A-36)

E-5 High concentrations of phosphorus (15.1 ppm) were detected in one sample from this well in 1981. Subsequent analysis of this sample could detect no orthophosphate. Samples obtained in 1982 have shown no detectable phosphorus of any kind. Potential explanations include analytical error (plasma emission), but TBP-solvent contamination on the soil at the height of the water table (at that time) is also conceivable. Variations of this kind have been seen before in locations around the 1971 solvent spill (REF. DPST-77-495).

Well E-5 should be analyzed for traces of plutonium activity.

E-7 The pH in this well was exceptionally low (pH = 2.38). However, none of the other parameters, including conductivity, were significantly deviant. As expected, the total carbonate concentration in this well was lower than normal.

In low pH systems of this kind, the bulk migration rate of Ru-106, Sr-90, and Cs-137 are greatly enhanced. (see pH) This well should therefore be suspected of containing traces of these radionuclides. The levels of Sr-90 contamination would be expected to be higher than those of other fission products if pH does affect radionuclide transport around this well.

E-17 This well contains elevated concentrations of sodium (30 ppm), calcium (56 ppm), potassium (10 ppm), and sulfate (111 ppm). Like Well E-5, it also appeared to contain a phosphorus compound (13.7 ppm P) in 1981. The pH was relatively high (6.54), fluoride was present (0.4 ppm), and the conductivity was fifth highest of all wells (310 µmho/cm).

This well has a history of non-volatile beta-gamma contamination and it is in the vicinity of the TBP/ultrasene solvent spill that occurred in 1971 (see Operation's History). Both Cs-137 and Co-60 have been detected by low-level gamma spectrometry. No Sr-90 or plutonium isotopes were detected, but continued low-level monitoring should be performed.

G-7 This well was anomolous in more chemical parameters than any other well. The oxidation-reduction potential was the lowest (0.172 V vs. S.H.E.), conductivity the highest (530 µmho/cm), magnesium the highest (39.9 ppm), and iron the second highest (60.6 ppm) of all wells. Total organic carbon, sodium and chloride were all statistically abnormal, and the dissolved oxygen was only 1.5 ppm (at 21°C). The inorganic carbon was 43 ppm, which probably accounts for the relatively high pH (6.48).

Careful analysis of this well would probably detect Cs-137, Sr-90 and several other fission products. The extremely high concentration of dissolved organic carbon (45 ppm) that was detected in this well means that plutonium may also be present as an organic complex of Pu(IV). Aside from a detailed radiochemical analysis, this well is a prime candidate for a complete organic analysis.

G-9 Well G-9 is statistically high in sulfate (21 ppm). It also is slightly high in magnesium, potassium, and sodium. The presence of organic carbon and the low oxidation-reduction potential (0.264 V) could be signs of organic complexation strength. However, the conductivity of this well (140 μmho/cm) is only slightly higher than the average.

This well probably does not contain Sr-90 or Cs-137 at measurable levels. Plutonium could be present, but only in the form of an organic complex of Pu (IV).

G-21 This well has long been identified as a "bad actor" in the 643-G Burial Ground. It has a history of tritium contamination, plutonium contamination (Pu-238/239), and non-volatile beta-gamma contamination (see Tables B1-B2). Sr-90 has been detected at 1600 pCi/L in Well G-21.

The well water is reduced relative to the surrounding system (dissolved oxygen = 2.8 ppm, E_h=0.369 V). The pH is a moderate 5.03 but the iron concentration and dissolved organic carbon were the highest detected in the burial ground (Fe = 112.8 ppm, TOC = 225 ppm). Chloride was also relatively high at 33 ppm.

The chemical parameter most responsible for the strontium migration here may be the large amount of iron in solution. The plutonium is probably in the form of organic complexes of Pu (IV).

Additional low-level analyses of this well should be performed. The source of the contamination and the operating "K_d" for the radionuclides in this system should be identified through coring studies. A complete analysis of the organics in the system is also highly desirable.

G-23 This well contains calcium at 105 ppm, a concentration that is high enough to reduce the K_d of Sr-90 to about 10 for the soils in the burial ground region. The dissolved oxygen in this well was only 1.7 ppm (21°C, and conductivity was 270 μmho/cm. Other factors were approximately normal.

Samples of water from Well G-23 should be submitted for low-level analysis, especially Sr-90.

I-13 Although most of the activity is from naturally occurring sources, well I-13 has a history of alpha and beta-gamma contamination (see Table B1). However, given the high levels of chemical contamination that were measured, it is not surprising that Cs-137 (10 pCi/L), Sr-90 (19 pCi/L) and even trace levels of plutonium (2 ± 1 pCi/L of Pu-238) were identified in that well. Well I-13 is also one of the wells that were contaminated with TBP/ultrasene extraction solvent in 1971.

The conductivity of water samples from Well I-13 was 450 μmho/cm). Abnormally high levels of three cations were measured, including potassium (110 ppm), sodium (40 ppm) and barium (21 ppm). I-13 was one of only three wells that exceeded 1 ppm of barium, and it contained by far the most. The next highest was 2.4 ppm, detected in Well I-17. Chloride was 152 ppm in I-13, also the highest of any well.

This well should be analyzed in the future to verify the apparent downward trend in radionuclide contamination levels. Additional chemical monitoring should be employed to detect any correlation between changes in activity and the chemical contamination level. Trace level organic analysis should also be attempted, to test the hypothesis that plutonium contamination has come about solely due to the solvent spill which occurred in 1971.

I-15 This well was statistically abnormal in chloride (35 ppm). Nitrate (28 ppm), magnesium (7.60 ppm) and sodium (19 ppm) were also slightly high, but the overall conductivity was actually lower than average due to the lack of calcium carbonate.

Enhanced radionuclide transport is not predicted around this well.

I-17 This well is anomalous for its barium content (2.4 ppm). The magnesium to calcium concentration ratio is slightly high, but it is uncertain whether this indicates natural or "waste" calcium. Several other apparently normal wells were also slightly higher than average in magnesium/calcium. Other parameters were approximately normal. Radioactivity would not be expected in Well I-17, but the presence of barium (2.4 ppm) has not been adequately explained, and there could be some coincidence of radioactivity (Ra-226 etc; with barium).

Approximately 2 pCi/L of Pu-238 were detected in this well (Jan. 1981). Additional measurements are needed to confirm this finding, which could be caused by organic complexation. Even 1.6 ppm (the TOC in I-17) of a complexing agent could mobilize measurable concentrations of Pu (IV). Although it was not one of the wells that were affected originally, Well I-17 may have been contaminated slightly by the solvent spill that occurred in 1971 (see Operation's History).

APPENDIX F

RADIOACTIVE WELLS WITH "NORMAL" CHEMISTRY

A-3 Except for having the second highest pH (6.95) of all monitoring wells, Well A-3 was unremarkable in its chemistry, and therefore, was not a "suspect" well in that respect. However, it has had a history of beta-gamma activity since it was first sampled, so a low-level radiochemical analysis was performed. Alpha activity (Pu-238) and fission products (Cs-137 and Sr-90) were detected.

The source of the contamination probably lies in the fact that an equipment decontamination station once operated nearby. The predicted flow path of complexed material is from the old station, through A-3, through Wells C-1 and C-3 and then to the south and west (see Figure 5). The trend of data over the past five years on Well A-3 is toward lower and lower non-volatile beta-gamma activities (with a 5-year average slope of -138.5 pCi/L-yr). Present levels of activity are less than 70 pCi/L non-volatile beta-gamma so pCi/L of Sr-90 and 11 pCi/L of Pu-238. Since the decontamination station is no longer operating, this downward trend is expected to continue.

C-3 The only man made activity that was detectable in this well was a trace of plutonium (2 ± 1 pCi/L of Pu-238). Since no Pu-239 was detected, precise measurement of the 238/239 ratio could not be used to confirm or deny the theory that a common source was responsible for the contamination in Wells C-1, C-3, and A-3 (see Operations History).

C-15 This well does not show any unusual chemical characteristics other than the presence of 0.4 ppm of fluoride. As described previously (see Fluoride) this concentration could mobilize small amounts of Pu(VI).

Well C-15 has a history of alpha contamination (see radiochemical data, Table B1), and both Pu-238 and Pu-239 have been identified in that well. Although it is possible that flouride contributed to the mobility of the plutonium, the most likely transport mechanism is organic complexation, probably in the form of TBP and its degradation products. These complexes could have come from the solvent burning operations which took place in the trenches near there. An even more likely source, however, is the solvent spill, in 1971, which allowed several hundred gallons of solvent to reach the water table (see Operations History).

I-9 A trace of plutonium was detected in this well (2 ± 1 pCi/L of Pu-238). The source was probably TBP-kerosene extraction solvent that was spilled in 1971 (see Operations History).

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