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Uranium in Surface Soil, Surface Water, and Groundwater at the Rocky Flats Environmental Technology Site

June 2004

Environmental Media Management
Rocky Flats Environmental Technology Site

Kaiser-Hill Company, L.L.C.
Classification Exemption CEX-105-01

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1.0 PURPOSE

This report provides a general review and compilation of uranium data for surface soil, surface water, and groundwater at the Rocky Flats Environmental Technology Site (RFETS or Site). Data presented are from multiple investigations, conducted from 1993 through 2004, which are related to uranium in the RFETS environment. Data and information in this report supplement the Pathway Analysis Report, which addresses the transport of plutonium, americium, and uranium in the environment at RFETS (Kaiser-Hill, 2002a).

Background information is provided on different types of uranium, including the naturally-occurring form, as well as anthropogenic (man-made) types. In addition, information is provided on uranium chemistry, the environmental behavior of uranium, different analytical techniques used to measure uranium in the environment, and Site-specific information on historic uranium usage. Surface soil, surface water, and groundwater uranium data from RFETS are presented and compared with applicable Rocky Flats Cleanup Agreement (RFCA) standards and Action Levels. Finally, implications for Site closure are addressed.

2.0 BACKGROUND INFORMATION

Background information addressed includes:

- A description of natural and anthropogenic uranium (Section 2.1);
- A brief summary of uranium environmental chemistry (Section 2.2);
- Information on analytical techniques pertinent to uranium (Section 2.3);
- Site-specific information on historic uranium operations (Section 2.4);
- Background concentrations of uranium and RFCA Action Levels and standards for soil, surface water, and groundwater (Section 2.5); and

- Information on RFETS treatment systems used to capture and treat uranium in groundwater (Section 2.6).

2.1 NATURAL AND ANTHROPOGENIC FORMS OF URANIUM

2.1.1 Natural Uranium

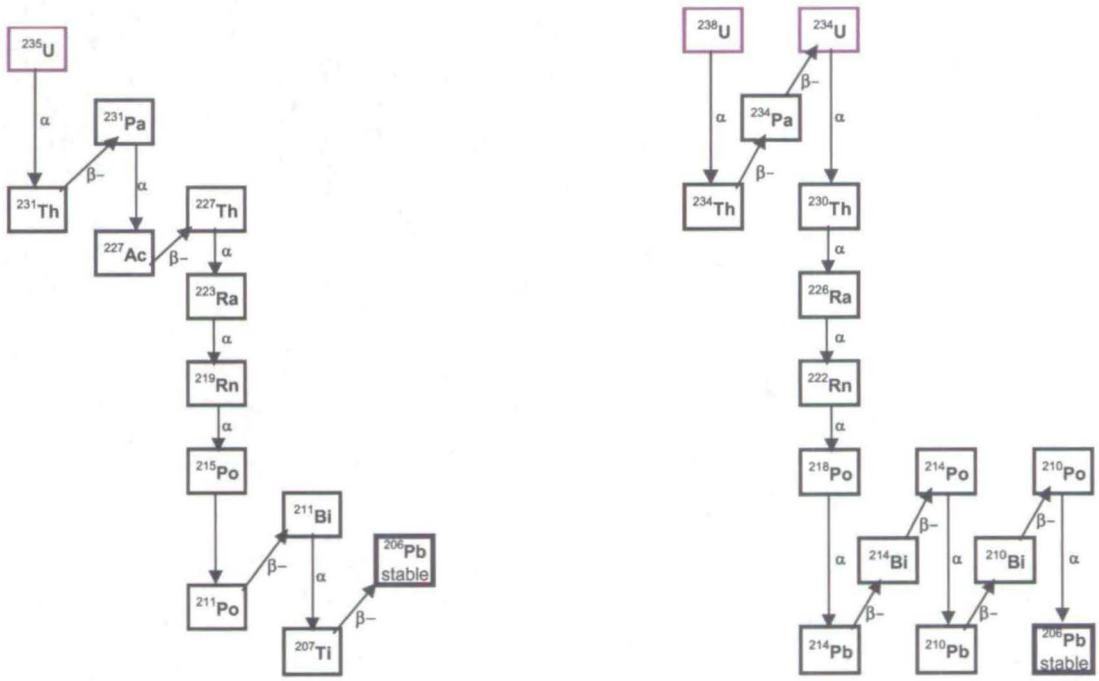
Uranium occurs naturally in the Earth's crust, averaging 2.3 milligrams per kilogram (mg/kg)(1.6 picoCuries per gram [pCi/g]) (Langmuir, 1997). It is most abundant in granites (averaging 5 mg/kg [3.5 pCi/g]) and shales (averaging 3.5 mg/kg [2.5 pCi/g]) (Krauskopf, 1979). In a relative sense, natural uranium is not particularly radioactive compared to other radioactive elements. For example, it requires approximately 2,800,000 grams of natural uranium to emit the same radioactivity as one gram of radium-226 (Filov and Ivin, 1993).

Natural uranium activity is ubiquitous in the Front Range of Colorado (Langmuir, 1997). The Ralston Buttes Uranium District boundary is located within two miles west of RFETS, and the Schwartzwalder mine, used as a source of uranium ore, is located approximately 10 miles from the Site near Ralston Reservoir. From the watershed perspective, the South Platte River is known to be "...anomalously rich in uranium compared to most other rivers of its size" (Bolivar et al., 1978).

The presence of relatively large amounts of naturally-occurring uranium in the region complicates studies to identify uranium from anthropogenic sources at the Site. Consequently, a relatively high concentration of uranium detected in the RFETS environment can, in some cases, be attributed to natural sources; it does not necessarily indicate the uranium is contamination from industrial sources.

Natural uranium is a mixture of three α-emitting isotopes, U-234, U-235, and U-238. Figure 2-1 presents decay chains for these isotopes. U-235 belongs to the actinium series, while U-238 and U-234 belong to the uranium series. As shown in Figure 2-1, U-234 is a decay product of U-238.

Figure 2-1. Natural Uranium Decay Chains



The naturally-occurring uranium isotope U-235 belongs to the actinium series (left), while the other naturally-occurring uranium isotopes, U-238 and U-234, belong to the uranium series (right). Each α (alpha particle) decay involves the emission of 2 neutrons and 2 protons, resulting in a daughter product with a molecular weight reduced by 4 atomic mass units. Each β (beta particle) decay involves the emission of 1 electron, which results in a negligible reduction in atomic mass for the daughter product. The stable lead isotope, Pb-206, is the end product of both decay chains. (Figure Source: Efurd, et. al., 1993a).

While the concentration of natural uranium varies significantly in different locations, the proportions of uranium isotopes in natural uranium are essentially fixed, although the proportions do change very slowly over time, because of varying decay rates of the different isotopes. Natural uranium is a mixture of three α-emitting isotopes: U-234, U-235, and U-238. These isotopes exist in the mass percentages listed in Table 2-1.

Table 2-1. Naturally-Occurring Uranium - Isotopic Composition

Uranium Isotope	Percent by Mass (%)	Half Life (years)	Specific Activity (Curies/gram)
U-238	99.274	4.47×10^{9}	3.33×10^{-7}
U-235	0.7202	7.08×10^{8}	2.14×10^{-6}
U-234	0.0057	2.45×10^{5}	6.25×10^{-3}

Source: (Clark et al., 1997)

Although U-234 comprises only 0.0057 % of the mass of natural uranium, its specific activity is nearly four orders of magnitude higher than that of U-238 (see Table 1). Therefore, although there is much less mass of U-234 compared to U-238, the activity emitted from U-234 and U-238 in natural uranium is approximately equal.

2.1.2 Anthropogenic Uranium

2.1.2.1 Enriched and Depleted Uranium

Enriched and depleted uranium are the two main types of anthropogenic uranium used historically at RFETS in the production of nuclear weapons components. These two forms of uranium are discussed below:

Enriched uranium - Uranium enriched with U-235 creates an atomic mass fraction of U-235 that is larger than the 0.72 % of U-235 found naturally. Enrichment may be achieved by UF_6 gaseous diffusion or centrifugation processes, using UF_6 atomic vapors (AVLIS) or molecular vapors (MLIS) laser isotope separation (Grant and Grant, 1987). The enrichment processes were conducted elsewhere and the enriched uranium was transported to RFETS.

There is a distinction between enriched uranium and highly enriched uranium. Enriched uranium is commonly used in nuclear reactors, whereas highly enriched uranium refers to material used for special applications, such as compact naval reactors or for the weapons-grade material historically used at RFETS for weapons component fabrication. The mass percents of uranium isotopes in highly enriched uranium are presented in Table 2-2.

Depleted uranium – Uranium from which most of the fissile material, U-235, has been removed is referred to as depleted uranium. Therefore, depleted uranium has a U-235 mass fraction below the 0.72 % found naturally. The mass percents of uranium isotopes in depleted uranium are presented in Table 2-2.

Table 2-2. Natural, Highly Enriched, and Depleted Uranium Mass Percents

	Mass Percent				
	U-233	**U-234**	**U-235**	**U-236**	**U-238**
Natural	0	0.0057	0.720	0	99.274
Highly Enriched	< 0.002	1.007	95 - 97	0.4	3 -5
Depleted	< 0.005	< 0.01	0.25	< 0.01	99.75

Sources: Clark et al., 1997; DOE, 1997.

2.1.2.2 Indicators of Anthropogenic Uranium

U-236 - The presence of U-236 indicates the existence of uranium from a source that has been produced in a nuclear reactor (neutron capture on U-235). Alternatively, U-236 can be generated from the decay of Pu-240 ($t_{1/2}$ = 6,537 years).

It is possible, though unlikely, to have anthropogenic uranium *without* U-236. For example, U-236 is not produced when U-238 is refined by gaseous diffusion. However, the majority of uranium used at RFETS (from the late 1950s until the end of production in 1989) did contain U-236 (Gregory-Frost, 2003).

To determine if anthropogenic uranium is present, U-236 is a better marker than U-233 (see below), because U-233 is generally present in very low concentrations and is difficult to detect. Nevertheless, detection of U-236 requires the use of a suitably accurate analytical method, such as Inductively Coupled Plasma/Mass Spectrometry or Thermal Ionization Mass Spectrometry (see Section 2.3).

U-233 – U-233 is an anthropogenic uranium isotope produced in reactors by neutron capture. The U-233 material contains an admixture of U-232 whose decay chain produces gamma rays. Environmental sample results at RFETS often report U-233 and

U-234 together because the analytical method frequently used (alpha spectroscopy) does not distinguish between U-233 and U-234.

2.2 URANIUM CHEMISTRY

2.2.1 Introduction

Uranium has an atomic number of 92 and 19 isotopes, all radioactive, with atomic masses of 218, 222, 225 through 240, and 242. The radioactive half-lives for the isotopes range from 1×10^{-6} seconds for U-222, to 4.468×10^{9} years for U-238, the main naturally-occurring isotope (Clark et al., 1997; Holden, 1997).

2.2.2 Oxidation States

The oxidation state of an actinide has a controlling effect on its environmental behavior. Oxidation states, in turn, are determined by each actinide's unique electronic structure and the geochemical conditions of the surrounding soil and water. The actinide oxidation states of environmental interest are III, IV, V, and VI. Different oxidation states can form various molecular complexes, each with a characteristic solubility and chemical reactivity. Actinides in the lower oxidation states (III and IV) tend to form complexes with very low solubilities and the strongest sorption to mineral and rock surfaces. Actinides in the higher oxidation states (V and VI) tend to form complexes with much higher solubilities and weaker sorption to mineral and rock surfaces.

Uranium can be stable in the III, IV, V, and VI oxidation states, but is most commonly found in the environment as U(IV) and U(VI). This condition exists because dissolved U(III) readily oxidizes to U(IV) under most environmental conditions, and the U(V) state readily disproportionates to U(IV) and U(VI) in the presence of water or hydrolytic compounds (Clark et al., 1997). Therefore, U(IV) and U(VI) are the most common oxidation states for uranium in natural waters and soils.

Of the oxidation states observed in the environment, U(IV) tends to hydrolyze and form strong hydrolytic complexes, form precipitates that are sparingly soluble, and adsorb strongly to mineral surfaces. These reactions cause U(IV) to be largely immobile in

groundwater. The solubility of U(IV) can be increased somewhat by complexes with naturally-occurring organic materials, particularly in areas with high concentrations of dissolved organic materials.

In contrast to U(IV), U(VI) is more soluble, undergoes weaker specific sorption and tends to be more mobile (Salomons and Foerstner, 1984). The solubility of U(VI) accounts for its wide distribution in fresh water, sea water, and hydrothermal deposits (Clark et al., 1997).

2.2.3 Natural Uranium Sources

As discussed in Section 2.1.1, uranium is widely distributed in natural geologic deposits. Approximately 155 minerals are known to contain uranium as a major constituent, and another 60 minerals contain minor amounts of uranium, or uranium as an impurity (Clark et al., 1997). Important sources of U(IV) and U(VI) include:

U(IV)

- uraninite (UO_2 through $UO_{2.25}$); and
- coffinite ($USiO_4$).

U(VI)

- carnotite [$K_2(UO_2)_2(VO_4)_2$] ;
- schoepite ($UO_3 \cdot 2H_2O$);
- rutherfordine (UO_2CO_3);
- tyuyamunite [$Ca(UO_2)_2(VO_4)_2$] ;
- autunite [$Ca(UO_2)_2(PO_4)_2$];
- potassium autunite [$K_2(UO_2)_2(PO_4)_2$]; and
- uranophane [$Ca(UO_2)_2(SiO_3OH)_2$].

Commercially recoverable concentrations of uranium are also found in phosphate rock and lignite. In the presence of lignite and other sedimentary carbonaceous substances, uranium enrichment is believed to be the result of uranium reduction to form insoluble precipitates, such as uraninite (Langmuir, 1978; Clark et al., 1997).

2.2.4 Geochemical Processes

2.2.4.1 Aqueous Speciation of Uranium

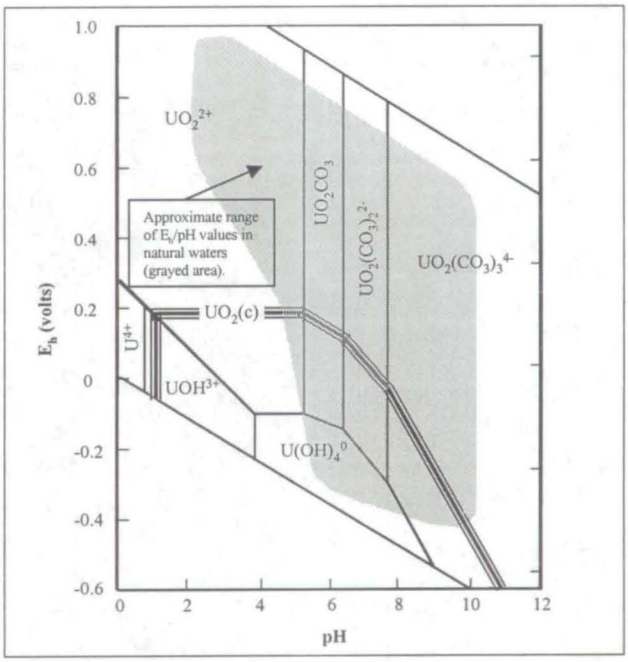
Uranium ions in aqueous solution can generate very complex species. Anions, such as carbonate, nitrate, chloride, fulvic acid, humic acid, and ethylenediaminetetraacetic acid (EDTA), form complexes with both U(IV) and U(VI), thereby increasing the amount of uranium that can remain in solution and, consequently, increasing the overall mobility of uranium. Uranium is generally least mobile in reducing (anaerobic) environments free of complexing anions and most mobile in oxidizing (aerobic) environments with high concentrations of complexing anions.

U(VI) species dominate in oxidizing environments, where U(VI) is not bound tightly to soils in alkaline conditions because of the predominance of neutral or negatively charged species above pH 5 (see Figure 2-2). In addition, an increase in CO_2 pressure in soil solutions can further reduce U(VI) adsorption by promoting the formation of poorly sorbing carbonate complexes.

A large number of additional U(VI) species will exist in a chemically more complicated system containing inorganic complexing ligands as shown in Figure 2-3. The water in Figure 2-3 has a U(VI) concentration of 1,000 µg/L (details of the water composition are provided in EPA, 1999). At pH values less than 5, the UO_2F^+ species dominates the system, whereas at pH values greater than 5, carbonate complexes [$UO_2CO_3^0$(aq), $UO_2(CO_3)_2^{2-}$, $UO_2(CO_3)_3^{4-}$] dominate the system. These calculations clearly show the importance of carbonate chemistry on U(VI) speciation. For the water composition shown in Figure 2-3, complexes with chloride, sulfate, and phosphate were relatively less important.

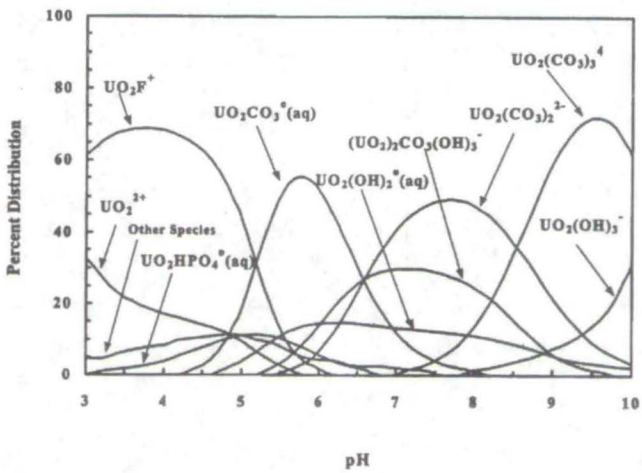
For perspective, the Upper Hydrostratigraphic Unit at RFETS is primarily a calcium-bicarbonate water (EG&G, 1995a; Ball, 2000). Therefore, in general, ample quantities of carbonate should generally be available to complex with uranium and promote its mobility in groundwater.

Figure 2-2. Calculated Pourbaix predominance diagram for aqueous species in the U-O_2-CO_2-H_2O system



Conditions are pure water at 25°C, 1 bar total pressure, $\Sigma U = 10^{-8}$ M and $p(CO_2) = 0.01$ bar. The $UO_2(c)$ solid/solution boundary for $\Sigma U = 10^{-8}$ M is shown as a tripled line overlay. The approximate range of E_h/pH values found in natural waters is shown within the grayed area. (Adapted from Langmuir, 1997.)

Figure 2-3. Calculated distribution of U(VI) aqueous species as a function of pH



Water containing inorganic complexing ligands and a U(VI) concentration of 1,000 µg/L. The distribution of U(VI) species was calculated using the MINTEQA2 code using thermodynamic data from Grenthe et al., 1993. Figure reproduced from EPA, 1999.

Organic complexes are also important to uranium aqueous chemistry. The uncomplexed U(VI) ion has a greater tendency to form complexes with fulvic and humic acids than many other metals with a +2 valence (Kim, 1986, Shanbhag and Choppin, 1981). Kim (1986) concluded that, in general, VI actinides, including U(VI), would have approximately the same tendency to form humic- or fulvic-acid complexes as to hydrolyze or form carbonate complexes. This suggests that the dominant reaction of the uranyl ion depends on the relative concentrations of hydroxide, carbonate, and organic materials. Importantly, U(VI) can form stable organic complexes, thereby increasing its solubility and mobility.

2.2.5 Dissolution, Precipitation and Coprecipitation of Uranium

Dissolution, precipitation, and coprecipitation generally have a greater effect on the concentrations of U(IV) than on the concentration of U(VI) in groundwaters, because of the lower solubility of U(IV) compounds. However, U(VI) mobility can be somewhat limited by precipitation where aqueous U(VI) is in contact with the lower solubility phosphate and vanadate uranyl minerals, and U(VI) solution-mineral equilibria impose precipitation control on uranium migration (Clark et al., 1997; Langmuir, 1997).

In cases with low dissolved U(VI) concentrations under oxidizing conditions, as in oxygenated groundwaters far from a uranium source, solution-mineral equilibria will not limit the concentration of U(VI). However, near a uranium source, where U(VI) concentrations are higher, or in reducing environments, the precipitation processes become increasingly important and, several coprecipitates may form, depending on the environmental conditions (Falck, 1991; Frondel, 1958).

2.2.6 Sorption/Desorption of Uranium

2.2.6.1 Sorption and Ion Exchange Processes

In low ionic strength solutions, with low U(VI) concentrations, dissolved uranyl concentrations are likely to be controlled by adsorption processes. The uranyl ion and its complexes adsorb onto clays (Ames et al., 1982; Chisholm-Brause et al., 1994), organics

(Borovec et al., 1979; Read et al., 1993; Shanbhag and Choppin, 1981) and oxides (Hsi and Langmuir, 1985; Waite et al., 1994).

As the ionic strength of an oxidized solution increases, other ions, mainly Ca^{2+}, Mg^{2+} and K^{+}, displace the uranyl ion from soil exchange sites, thereby forcing the uranyl ion into solution. Not only do other cations dominate the uranyl ion in competition for exchange sites, but carbonate ions form strong soluble complexes with the uranyl ion, increasing the total amount of uranium in solution (Yeh and Tripathi, 1991). For this reason, the uranyl ion is particularly mobile in high ionic strength solutions.

2.2.6.2 Uranium Uptake by Minerals

Some of the uranyl sorption processes are not completely reversible. Sorption onto iron and manganese oxides can be a major process for extraction of uranium from solution (Hsi and Langmuir, 1985; Waite et al., 1994). These oxide phases act as a relatively irreversible sink for uranium in soils. Uranium bound in these phases is not generally in isotopic equilibrium with dissolved uranium in the same system, suggesting that the reaction rate mediating the transfer of the metal between the two phases is slow.

2.2.6.3 Uranium Uptake by Organics

Naturally-occurring organic matter is another possible sink for U (VI) in soils and sediments. The mechanisms by which uranium is sequestered by organic matter have not been determined in detail (EPA, 1999). A possible process involves adsorption of uranium to humic substances through rapid ion-exchange and complexation with carboxylic and other acidic functional groups (Boggs et al., 1985; Borovec et al., 1979; Idiz et al., 1986; Shanbhag and Choppin, 1981; Szalay, 1964). These groups can coordinate with the uranyl ion, displacing waters of hydration, to form stable complexes. This could account for a significant fraction of the organically-bound uranium in surface and subsurface soils.

Alternatively, sedimentary organic matter may act to reduce dissolved U(VI) species to U(IV) (Nash et al., 1981). Uranium sorption to iron oxide minerals and smectite clay has been shown to be extensive in the absence of dissolved carbonate (Ames et al., 1982; Hsi

and Langmuir, 1985). However, in the presence of carbonate and organic complexing agents, sorption has been shown to be substantially reduced, thereby increasing the mobility (Hsi and Langmuir, 1985).

2.3 ANALYTICAL METHODS FOR URANIUM ANALYSES

Data presented in this report include the use of three analytical methods, including: 1) alpha spectroscopy, 2) Inductively Coupled Plasma/Mass Spectrometry (ICP/MS), and 3) Thermal Ionization Mass Spectrometry (TIMS). These methods, and their application for analyzing uranium in surface water and groundwater, are described in this section.

2.3.1 Activity-Based Analytical Method

2.3.1.1 Alpha Spectroscopy

Alpha spectroscopy involves preliminary treatment of the sample, followed by complex radiochemical separation. The sample is passed through an ion exchange column where the element is isolated and concentrated prior to flaming and counting. Although alpha spectroscopy measurements produce relatively high sensitivity results, they have somewhat limited utility for determining isotopic ratios. Minimum detection limits are sample specific. Approximately 13% variability was determined in a study performed by the State of California, Department of Health Services (Liu et al., 2002).

2.3.2 Mass-Based Analytical Methods

2.3.2.1 Mass Spectrometry

Mass spectrometry (MS) involves the separation of matter according to atomic and molecular mass. Materials are ionized and analyzed according to their mass-to-charge ratio. Mass spectrometers are frequently coupled with state-of-the-art separation techniques, such as inductively coupled plasma (ICP) or thermal ionization (TI), to allow accurate determination of a compound at very low levels in complex matrices.

2.3.2.2 Inductively Coupled Plasma / Mass Spectrometry

ICP is used to analyze isotopes introduced to the instrument in solution. The process involves high frequency currents flowing in a coil that generates oscillating magnetic fields. Electrons flow through the oscillating electromagnetic field and inside a quartz tube space. Collisions cause heating and the ions give varying thermal temperatures associated with their individual properties. The differing temperatures, due to differences in ionization potentials among the elements and their isotopes, are measured. High resolution ICP/MS has a more sensitive detection limit than traditional ICP/MS and is more comparable to the results from TIMS analysis.

2.3.2.3 Thermal Ionization Mass Spectrometry

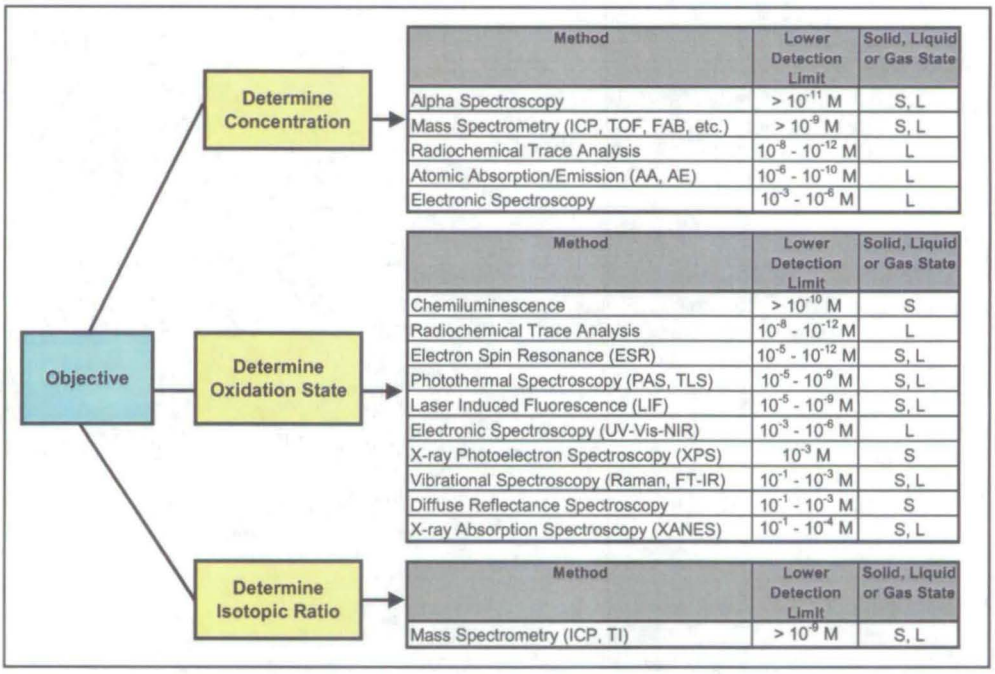
TI is used to analyze amounts of elemental isotopes evaporated from material precipitated on a filament. The analyte (e.g., uranium) is electrodeposited on a filament surface and then ionized. The process is affected by heating rates, filament temperature, and sample composition. Reproducing the fractionation condition is difficult. The most accurate results are achieved by repeated analysis of reference materials.

The amount of time required to chemically process water samples using alpha spectroscopy and TIMS is comparable. However, TIMS is a more accurate technique than alpha spectroscopy and provides more certain isotopic ratios. TIMS is best used for low-level measurements below the detection limits of the alpha spectroscopy measurements (Efurd et al., 1993b). High resolution (HR) ICP/MS and TIMS methods are comparable for low-level measurements and have similar detection limits, although TIMS tends to be less destructive on the molecule (Weiner, 2000).

2.3.3 Selection of Appropriate Analytical Method

Determination of which analytical method to use is dependent on the specific question asked. For example, determination of the oxidation state of uranium in a sample requires a different analytical method than one used to determine isotopic ratios. Figure 2-4 diagrams different options for analytical methods based on the question to be answered.

Figure 2-4. Analytical Method Options for Uranium Analyses



Method	Lower Detection Limit	Solid, Liquid or Gas State
Alpha Spectroscopy	> 10^{-11} M	S, L
Mass Spectrometry (ICP, TOF, FAB, etc.)	> 10^{-9} M	S, L
Radiochemical Trace Analysis	10^{-8} - 10^{-12} M	L
Atomic Absorption/Emission (AA, AE)	10^{-6} - 10^{-10} M	L
Electronic Spectroscopy	10^{-3} - 10^{-6} M	L

Method	Lower Detection Limit	Solid, Liquid or Gas State
Chemiluminescence	> 10^{-10} M	S
Radiochemical Trace Analysis	10^{-8} - 10^{-12} M	L
Electron Spin Resonance (ESR)	10^{-5} - 10^{-12} M	S, L
Photothermal Spectroscopy (PAS, TLS)	10^{-5} - 10^{-9} M	S, L
Laser Induced Fluorescence (LIF)	10^{-5} - 10^{-9} M	S, L
Electronic Spectroscopy (UV-Vis-NIR)	10^{-3} - 10^{-6} M	L
X-ray Photoelectron Spectroscopy (XPS)	10^{-3} M	S
Vibrational Spectroscopy (Raman, FT-IR)	10^{-1} - 10^{-3} M	S, L
Diffuse Reflectance Spectroscopy	10^{-1} - 10^{-3} M	S
X-ray Absorption Spectroscopy (XANES)	10^{-1} - 10^{-4} M	S, L

Method	Lower Detection Limit	Solid, Liquid or Gas State
Mass Spectrometry (ICP, TI)	> 10^{-9} M	S, L

2.4 HISTORIC URANIUM OPERATIONS AT RFETS

Uranium operations at RFETS were numerous and varied. Highly enriched uranium was employed in the manufacturing of components for nuclear weapons, and depleted uranium was used in other military applications. Descriptions of how natural uranium, highly enriched uranium, and depleted uranium were used at RFETS are provided in Sections 2.4.1 through 2.4.3, respectively. A summary of locations where uranium wastes were disposed at RFETS is provided in Section 2.4.4, and a map with locations of uranium operations and waste disposal sites is provided in Figure 2-5.

2.4.1 Natural Uranium Operations

Naturally-occurring uranium was not associated with major production activities at RFETS and was therefore only used in small quantities. According to Nuclear Materials Control/Nuclear Materials Accountability personnel, Rocky Flats did introduce some naturally-occurring uranium onsite; however, it was not used in accountable quantities, which is one kilogram for natural uranium (Gregory-Frost, 2003).

2.4.2 Highly Enriched Uranium Operations

The primary buildings used for historic highly enriched uranium operations are identified in Table 2-3.

Table 2-3. Highly Enriched Uranium Operations - Primary Buildings Used

Uranium Type	Building	Description of Operation
Highly Enriched Uranium	881	Original process building for highly enriched uranium. Processes included casting, forming, machining, assembly, recovery, and purification, in addition to handling the Site's waste solvent. 881 was equipped with a plenum and High Efficiency Particulate Air (HEPA) filtration system to filter the air before it was released through a stack. In addition, the 881 floors were surfaced with stainless steel sheeting to contain spills and facilitate cleaning (ChemRisk 3&4, 1992). Closeout of enriched uranium processes in 881 was completed in 1967.
	865	Supported production for research and development of metal-working operations.
	883	Produced formed parts that were sent to other buildings for machining. Operations involved the use of presses, mills, salt baths, and annealing furnaces. Work involved both highly enriched uranium and depleted uranium.
	886	Received uranyl nitrate solutions from 881. Used highly enriched uranium to conduct criticality experiments.
	771	Used for chemical decontamination processing of highly enriched uranium components.
	774	Treatment of liquid wastes, including those containing highly enriched uranium.
	Other 700 complex buildings	Assembly of components, including parts with highly enriched uranium.

Source: DOE, 2000.

2.4.3 Depleted Uranium Operations

The primary buildings used for historic depleted uranium operations are identified in Table 2-4.

Table 2-4. Depleted Uranium Operations - Primary Buildings Used

Uranium Type	Building	Description of Operation
Depleted uranium	444	Operations included casting, machining, milling, welding, plating, and part cleaning (degreasing).
	447	Used for manufacturing processes and waste handling, including processing of depleted uranium chips and turnings from other buildings.
	865	Produced formed parts which were sent to other buildings for machining. Operations involved the use of presses, mills, salt baths, and annealing furnaces. Work involved both depleted uranium and highly enriched uranium.
	883	Produced formed parts that were sent to other buildings for machining. Operations involved the use of presses, mills, salt baths, and annealing furnaces. Work involved both depleted uranium and highly enriched uranium.
	774	Treatment of liquid wastes, including those containing depleted uranium.
	Other 700 complex buildings	Assembly of components, including parts with depleted uranium.

Source: DOE, 2000.

2.4.4 Historic Uranium Disposal Areas

Areas that were historically used for on-Site disposal of depleted and/or enriched uranium are listed in Table 2-5 and shown on Figure 2-5.

Table 2-5. RFETS Areas Used for Historic Uranium Disposal

Disposal Area	Dates of Disposal	Description[1]	Remedial Action Taken
Trench T-1	1952 -1962	Drums with depleted uranium chips were disposed of in trench and covered with approximately 2 feet of dirt.	1998 - Remediated (soil excavation). Over 170 drums, and over 30 tons of soil were removed.
Trenches T-3 and T-4	1960s	Sanitary sewage sludge, flattened drums, debris, with some uranium contamination.	1996 - Remediated (soil excavation), involving approx. 3,000 cubic yards (cy) of soil and 300 cy of debris.
Trenches T-2 and T-5 – T-11	1954 – 1968	Disposal of flattened drums contaminated with uranium.	1999 - East Trenches Plume groundwater treatment system constructed (treats T-1 – T-4).

(Table continued)

Table 2-5 continued

Disposal Area	Dates of Disposal	Disposal Description[1]	Remedial Action Taken
Mound Area	1954 – 1958	Drums with oil and solid waste were buried. Most had depleted uranium, some with enriched uranium.	1997 – Remediated (soil excavation). Approx. 700 cubic yards (cy) of soil excavated.
Solar Evaporation Ponds	1953 - 1986	The ponds were used to store and evaporate process waste water, including liquid wastes containing depleted and enriched uranium.	1971 – Original groundwater interceptor trench installed 1995 - Pond sludges removed. 1999 – New groundwater collection and treatment system completed. 2002 – Buildings and facilities removed and area regraded. (Kaiser-Hill, 2003a)
Original Landfill	1952- 1968	Original waste disposal area for site. An estimated 20 kg of depleted uranium ash is buried along with other waste.	Accelerated action proposed via IM/IRA.
Ash Pits I-1 to I-4 (IHSS SW-1702, 133.1, 133.2, 133.4)	1952 - 1968	An estimated 100 grams of depleted uranium was burned with general combustible waste in the nearby incinerator. The ash was diposed of in these pits, which were covered with fill.	No Further Accelerated Action (NFAA) approved 6/12/03 (Kaiser-Hill, 2003b).
881 Hillside (OU 1)	1951 - 1972	Multiple disposal sites on hillside.	Groundwater treatment system installed and then dismantled in accordance with OU1 CAD/ROD.
Oil Burn Pit #1 (IHSS 300-128)	1957 and 1961 - 1965	Approximately 1,100 drums of oil containing uranium were burned. The resulting residues and some flattened drums were covered with backfill.	No Further Accelerated Action (NFAA) approved 6/20/03 (Kaiser-Hill, 2003c).
Oil Burn Pit #2 (IHSS 153)	1956	Ten drums of oil containing depleted uranium were burned in 1956. The residue was covered with backfill and later covered by Building 335.	Accelerated Action, if any required, not determined as of May 2004.

Source: DOE, 1992; ChemRisk, 1992.

Notes:

[1]Descriptions address uranium disposal only. Other contaminants potentially disposed of are not addressed in this table.

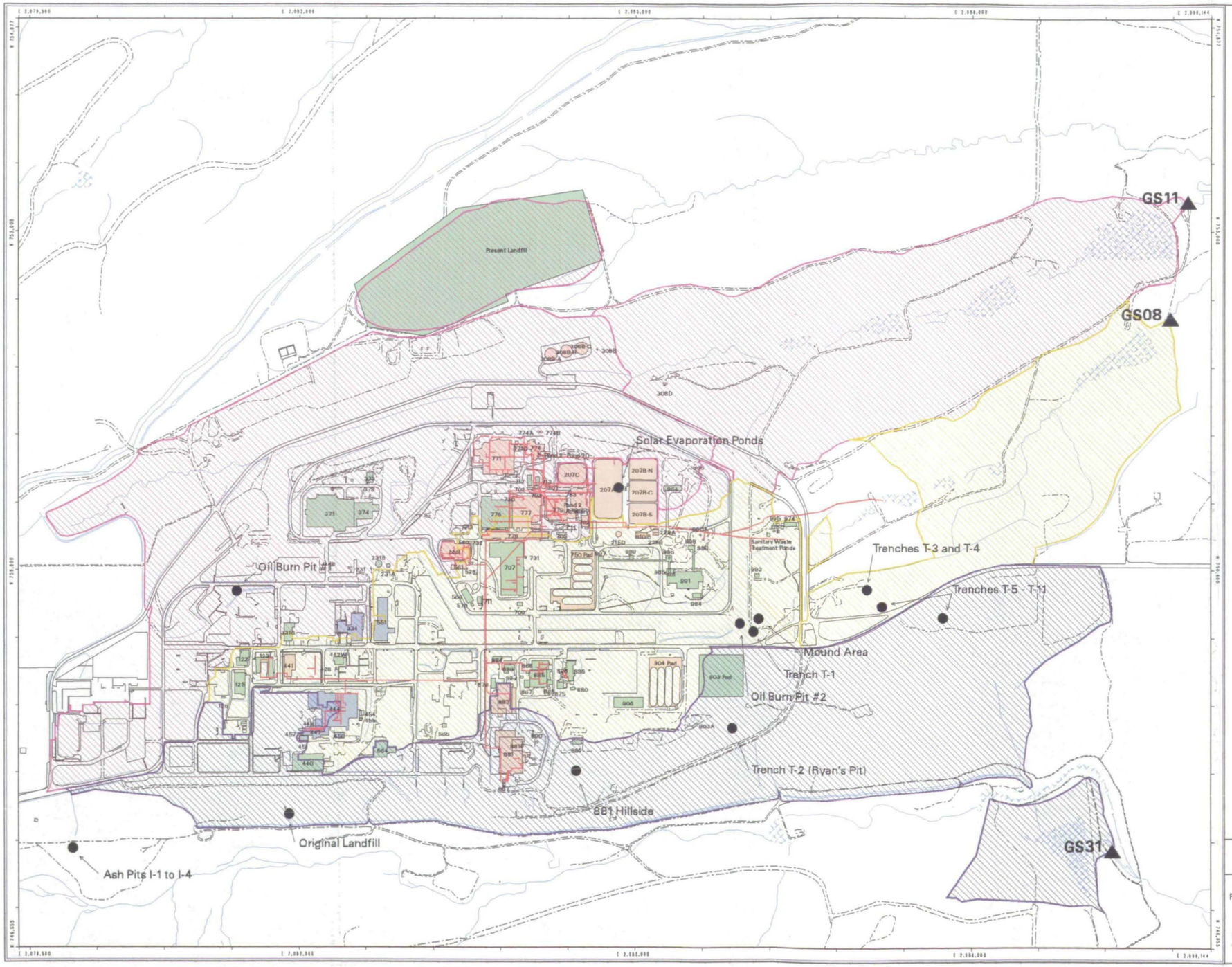


Figure 2-5

Uranium Usage in Buildings, Historic Disposal Areas, and Gage Locations with Drainage Basins

EXPLANATION

- Depleted Uranium
- Depleted and Enriched Uranium
- Depleted, Enriched and U-233
- North Walnut Creek Basin
- South Walnut Creek Basin
- South Interceptor Ditch Basin (Pond C-2)
- Original Process Waste Lines (OPWL)
- Historic Uranium Disposal Areas

Standard Map Features

- Lakes and ponds
- Streams, ditches, or other drainage features
- Paved roads
- Dirt roads

DATA SOURCE BASE FEATURES:
Buildings, fences, hydrography, roads and other structures from 1994 aerial flyover data captured by EG&G RSL Las Vegas. Digitized from the orthophotographs. 1/95

Scale = 1 : 10610
1 inch represents approximately 884 feet

250 0 500 1000ft

State Plane Coordinate Projection
Colorado Central Zone
Datum: NAD27

U.S. Department of Energy
Rocky Flats Environmental Technology Site

GIS Dept. 303-966-7707

Prepared by: CH2MHILL

Prepared for: KAISER-HILL COMPANY

June 02, 2004

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2.5 URANIUM IN THE ENVIRONMENT – BACKGROUND LEVELS AND RFCA REQUIREMENTS

Background concentrations of uranium in the environment, attributed to uranium from natural sources, were determined for different RFETS environmental media (EG&G, 1993; EG&G, 1995b). The background uranium concentrations are presented, by isotope, for surface soil, surface water, and groundwater (Upper Hydrostratigraphic Unit [UHSU] groundwater only) in Sections 2.5.1, 2.5.2, and 2.5.3, respectively. In addition, for each environmental media, the relevant RFCA Action Level or standard is also presented.

2.5.1 Soil

2.5.1.1 Soil - Background Uranium Concentrations

Background activities in surface soil for the uranium isotopes are displayed in Table 2-6. The sum of the mean concentrations of the isotopes, or total uranium mean background activity in surface soil, is approximately 2.2 pCi/g, with a total standard deviation of more than 1 pCi/g.

Table 2-6. Background Surface Soil Uranium Concentrations

Isotope	Sample Size	Maximum (pCi/g)	Minimum (pCi/g)	Std. Deviation (pCi/g)	Mean (pCi/g)
U-234	20	3.10	0.60	0.58	1.10
U-235	20	0.11	0.03	0.02	0.05
U-238	20	2.60	0.74	0.46	1.09

Source: *Geochemical Characterization of Background Surface Soils: Background Soils Characterization Report* (EG&G, 1995b).
Note: Surface soil defined in RFCA as soil from 0 to 6 inches in depth.

2.5.1.2 Soil – RFCA Action Levels

RFCA soil Action Levels are defined in Attachment 5 of RFCA (DOE, 2003). Soil Action Levels for uranium are isotope-specific, and are defined for both the Wildlife Refuge Worker and Ecological Receptor, as shown in Table 2-7. The Soil Action Levels

for uranium apply to the top six inches of soil. Below six inches, if uranium activity exceeds the Soil Action Level, then the Sub-Surface Soil Risk Screen, defined in RFCA Attachment 5, is applied to evaluate if a need exists for remedial action (DOE, 2003).

Table 2-7. Soil – RFCA Action Levels for Uranium Isotopes

Uranium Isotope	Wildlife Refuge Worker Action Level (pCi/g)	Ecological Receptor Action Level (pCi/g)
U-233/234	300	4,980
U-235	8	2,770
U-238	351	1,580

2.5.2 Surface Water

2.5.2.1 Surface Water - Background Uranium Concentrations

Background activities in surface water for the uranium isotopes are displayed in Table 2-8. The sum of the mean concentrations of the isotopes, or mean background activity in surface water for total uranium, is approximately 0.9 pCi/L, with a total standard deviation of more than 1 pCi/L.

Table 2-8. Background Surface Water Uranium Concentrations

Isotope	Sample Size (n)	Maximum (pCi/L)	Minimum (pCi/L)	Std. Deviation (pCi/L)	Mean (pCi/L)
U-234	79	3.21	-0.01	0.55	0.49
U-235	75	0.38	-0.03	0.07	0.05
U-238	55	1.82	0.00	0.43	0.36

Source: *Background Geochemical Characterization Report* (EG&G, 1993).

2.5.2.2 Surface Water – RFCA Standards

RFCA surface water standards are defined in Attachment 5 of RFCA (DOE, 2003). For uranium, the Site-specific standard is found in Table 2 of 5, Colorado Code of Regulations (CCR) 1002-8, §3.8.0. The surface water standard is for total uranium; it is not isotope-specific. The uranium standard varies by drainage and is based on a 30-day,

volume-weighted moving average. Table 2-9 presents the RFCA standard and Points-of-Compliance for Walnut and Woman Creeks.

Table 2-9. Surface Water – RFCA Uranium Standards and Point-of-Compliance Locations

Drainage	RFCA Surface Water Standard (pCi/L)	RFCA Point-of-Compliance Locations
Walnut Creek	10	GS11 (Below Pond A-4) GS08 (Below Pond B-5) GS03 (Walnut Ck. at Indiana Street)
Woman Creek	11	GS31 (Below Pond C-2) GS01 (Woman Ck. at Indiana Street)

Notes:
Uranium surface water standards are Site-specific. The values in the table reflect the classifications and standards approved by the Water Quality Control Commission (WQCC) effective March 2, 1997. Values apply as standards in Segments 4a and 4b and as Action Levels in Segment 5.

For perspective, in December 2000, the Environmental Protection Agency (EPA) established the final maximum contaminant level (MCL) for uranium at 30 micrograms per liter (μg /L) (40 CFR, Parts 9, 141 and 142, Final Rule). Per the EPA, the best estimate of a geometric average mass:activity ratio is 0.9 pCi/μg of natural uranium, for values near the MCL (Federal Register, 2000). Therefore, the 30 μg /L uranium MCL is equivalent to approximately 27 pCi/L; in other words, the MCL allows for nearly three times the amount of uranium in drinking water than the RFCA surface water standard.

2.5.3 Groundwater

2.5.3.1 Groundwater - Background Uranium Concentrations

Background uranium activities in groundwater, both dissolved and total, by isotope, are displayed in Table 2-10. In addition to the mean values for the uranium isotope concentrations, the 99/99 Upper Tolerance Limit (UTL) is provided, which indicates the UTL for 99 % of the sample population with 99% level of confidence. It is noted that significant differences in background uranium activity exist for different geologic formations, with groundwater in the Colluvium having the highest background uranium

activity and groundwater in the Rocky Flats Alluvium have the lowest background uranium activity.

Table 2-10. Background Uranium Concentrations in Groundwater

Isotope	Geologic Formation	Dissolved			Total		
		Sample Size (n)	Mean (pCi/L)	Upper Toler. Limit (99/99) (pCi/L)	Sample Size (n)	Mean (pCi/L)	Upper Toler. Limit (99/99) (pCi/L)
U-233/234	Rocky Flats Alluvium	78	0.23	0.88	12	0.48	2.58
	Colluvium	30	31.82	226.34	8	58.74	446.99
	Valley-Fill Alluvium	60	2.05	10.80	7	1.58	8.01
	Weathered Claystone	39	8.59	77.33	8	7.49	44.13
U-235	Rocky Flats Alluvium	78	0.03	0.23	12	0.12	1.05
	Colluvium	30	0.86	5.63	8	2.14	16.03
	Valley-Fill Alluvium	60	0.08	0.47	7	0.10	0.75
	Weathered Claystone	39	0.20	1.88	8	0.28	1.81
U-238	Rocky Flats Alluvium	69	0.14	0.56	11	0.40	2.83
	Colluvium	24	26.70	180.03	6	36.04	376.92
	Valley-Fill Alluvium	49	1.66	8.92	2	1.23	223.18
	Weathered Claystone	35	3.54	14.17	3	5.11	123.65

Source: Background Geochemical Characterization Report (EG&G, 1993).

2.5.3.2 Groundwater – RFCA Action Levels

RFCA groundwater Action Levels for uranium, displayed in Table 2-11, are defined in Attachment 5 of RFCA (DOE, 2003). The Tier II groundwater Action Levels for radionuclides are the corresponding residential ground water ingestion Preliminary Remediation Goals, and Tier I Action Levels are one hundred times the Tier II levels.

Table 2-11. Groundwater – RFCA Uranium Action Levels

Analyte	Tier II (pCi/L)	Tier I (pCi/L)
U-233/234	1.06	106
U-235	1.01	101
U-238	0.768	76.8

2.6 GROUNDWATER TREATMENT SYSTEMS FOR URANIUM

Two groundwater treatment systems at RFETS exist that were constructed to remove uranium, as well as other contaminants, from groundwater. These systems are described in Section 2.6.1 and Section 2.6.2.

2.6.1 Solar Ponds Plume Treatment System

In September 1999, a groundwater collection trench and treatment system was completed on the hillside downgradient from (northeast of) the site of the Solar Evaporation Ponds (Rocky Mountain Remediation Systems [RMRS], 2000a). The collection trench extends approximately 1,100 feet in an east-west direction, and cuts across previously existing trench systems constructed in 1971 and 1981. The 1981 system enhances recovery by the new collection trench (RMRS, 2000a). Water from the collection trench flows into a two-celled concrete treatment vessel. The treatment vessel contains zero-valent iron, to facilitate reduction of the uranyl ions and removal of uranium in the groundwater via sorption to the reactive media (see Section 2.2.6). Organic media are also present in the treatment cell to remove nitrate in the groundwater.

2.6.2 East Trenches Plume Treatment System

A groundwater collection and treatment system was installed in an east-west direction on the south side of South Walnut Creek, above Ponds B-1, B-2, and B-3. The purpose of the system is to capture, redirect, and treat contaminated groundwater (RMRS, 2000b). Installation of the East Trenches Plume Treatment System began in February 1999 and was completed on September 23, 1999. The collection trench is approximately 1,200 feet long, varies in depth from approximately 16 to 26 feet below grade, and is dug into the claystone at least 6 inches, and on average 3 feet. An impermeable barrier was installed in the trench, consisting of 80-mil high density polyethylene (HDPE) panels. The treatment system consists of two HDPE tanks containing reactive iron. The reactive iron reduces the uranyl ions and removes the uranium from the groundwater via sorption to the reactive media.

3.0 DATA

3.1 SURFACE SOIL

Surface soil, as defined in RFCA, is the upper six inches of soil (DOE, 2003). Surface soil maps of uranium isotopes are presented here to illustrate potential sources of uranium for surface water and groundwater. A total of 1,182 surface soil samples, collected from throughout the Site, were used in a geostatistical analysis, called kriging, of isotopic uranium data (Kaiser-Hill, 2002b). In the kriging analysis, the average concentration for each isotope is calculated for individual "blocks" of 75-feet by 75-feet over the entire Site. The blocks are shaded with colors representing the estimated soil concentration of each isotope over each block area. Surface soil kriged data for U-233/234, U-235, and U-238 are included on Figure 3-1, Figure 3-2, and Figure 3-3.

3.2 SURFACE WATER

Three sources of RFETS surface water uranium data are presented. They are:

- Alpha spectroscopy data from the RFCA surface water monitoring stations, collected by Site personnel from October 1, 1996 through Spring 2004 (Section 3.2.1);
- TIMS uranium isotope data from a Los Alamos National Laboratory (LANL) study on the RFETS retention ponds and drainages, conducted in the early 1990s (Efurd et al., 1993a) (Section 3.2.2); and
- TIMS uranium isotope data from a LANL study on selected surface water locations, conducted in 2002 (Section 3.2.3).

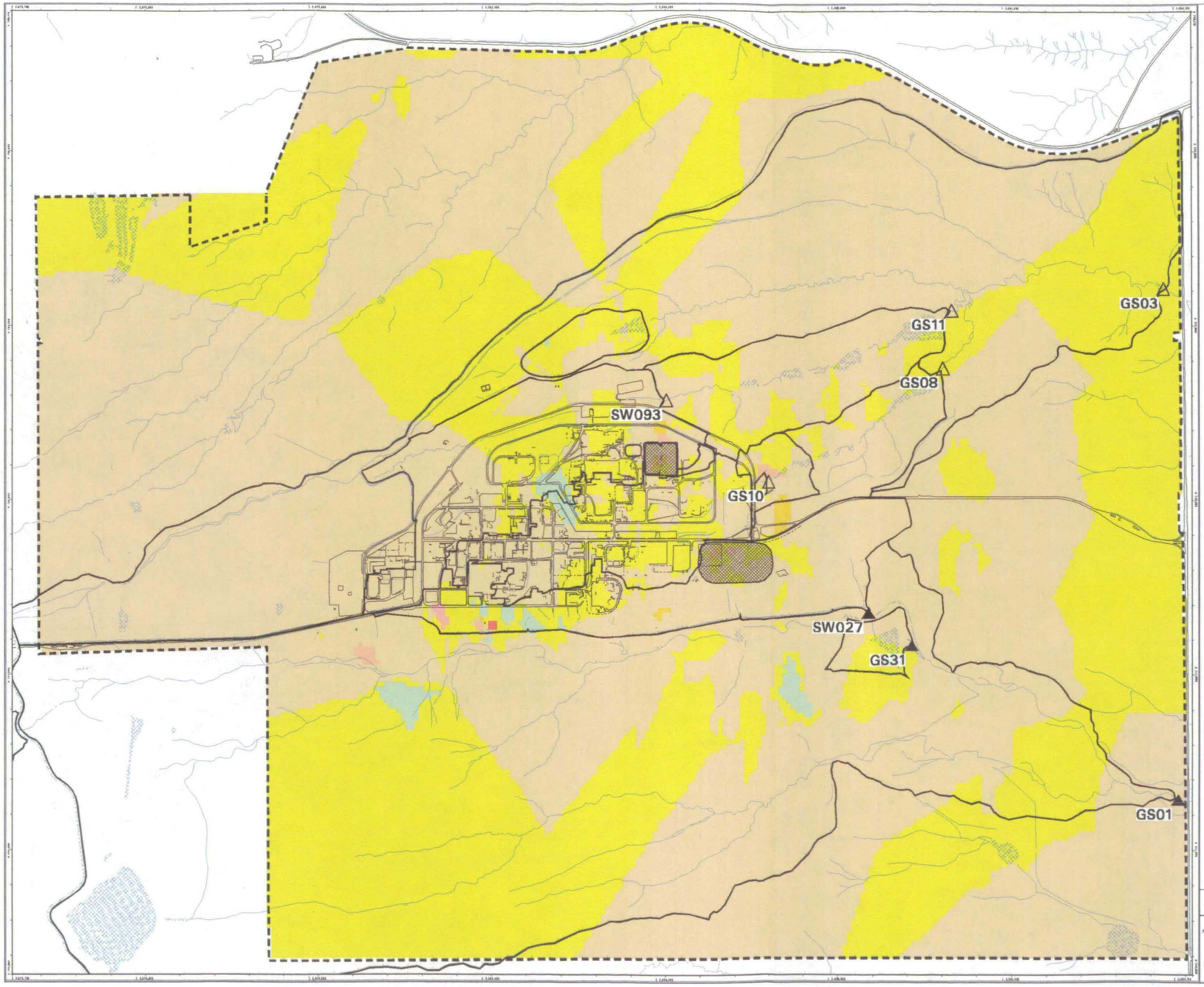

Figure 3-1

U-233/234 Activity in Surface Soils

Kriging Analysis Isoplot (pCi/g)

EXPLANATION

- Result < = 0.01
- 0.01 < Result < = 0.05
- 0.05 < Result < = 0.1
- 0.1 < Result < = 0.5
- 0.5 < Result < = 1.0
- 1.0 < Result < = 5.0
- 5.0 < Result < = 10.0
- 10.0 < Result < = 100.0
- 100.0 < Result < = 1000.0
- Result > 1000.0
- Areas altered by remediation or recontouring (approx.)
- Drainage Basin Boundary
- Walnut Creek Basin Gaging Station (GS03, GS08, GS10, GS11 & SW093)
- Woman Creek Basin Gaging Station (GS01, GS31 & SW027)

Standard Map Features

- Lakes and ponds
- Streams, ditches, or other drainage features
- Fences and other barriers
- Rocky Flats Environmental Technology Site boundary
- Paved roads

DATA SOURCE BASE FEATURES:

Buildings, fences, hydrography, roads and other structures from 1994 aerial fly-over data captured by EG&G RSL, Las Vegas. Digitized from the orthophotographs. 1/95

Analytical Data provided by Mark Wood (KH, 303-966-6689).

Kriged data provided by Jeff Myers (Westinghouse-Akin, 803-502-9747).

Scale = 1 : 21330
1 inch represents approximately 1778 feet

250 0 500 1000ft

State Plane Coordinate Projection
Colorado Central Zone
Datum: NAD27

U.S. Department of Energy
Rocky Flats Environmental Technology Site

Prepared by: CH2MHILL

Prepared for: KH

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Figure 3-2

U-235 Activity in Surface Soils

Kriging Analysis Isoplot (pCi/g)

EXPLANATION

- Result < = 0.01
- 0.01 < Result < = 0.05
- 0.05 < Result < = 0.1
- 0.1 < Result < = 0.5
- 0.5 < Result < = 1.0
- 1.0 < Result < = 5.0
- 5.0 < Result < = 10.0
- 10.0 < Result < = 100.0
- 100.0 < Result < = 1000.0
- Result > 1000.0
- Areas altered by remediation or recontouring (approx.)
- Drainage Basin Boundary
- Walnut Creek Basin Gaging Station (GS03, GS08, GS10, GS11 & SW093)
- Woman Creek Basin Gaging Station (GS01, GS31 & SW027)

Standard Map Features

- Lakes and ponds
- Streams, ditches, or other drainage features
- Fences and other barriers
- Rocky Flats Environmental Technology Site boundary
- Paved roads

DATA & SOURCE BASE FEATURES:

Buildings, fences, hydrography, roads and other structures from 1994 aerial fly-over data captured by EG&G RSL, Las Vegas. Digitized from the orthophotographs. 1/95

Analytical Data provided by Mark Wood (KH, 303-966-6689).

Kriged data provided by Jeff Myers (Westinghouse-Akin, 803-502-9747).

Scale = 1 : 21330
1 inch represents approximately 1778 feet

250 0 500 1000 ft

State Plane Coordinate Projection
Colorado Central Zone
Datum: NAD27

U.S. Department of Energy
Rocky Flats Environmental Technology Site

GIM Dept. 808-966-7/0.

Prepared by: CH2M HILL

Prepared for: KH

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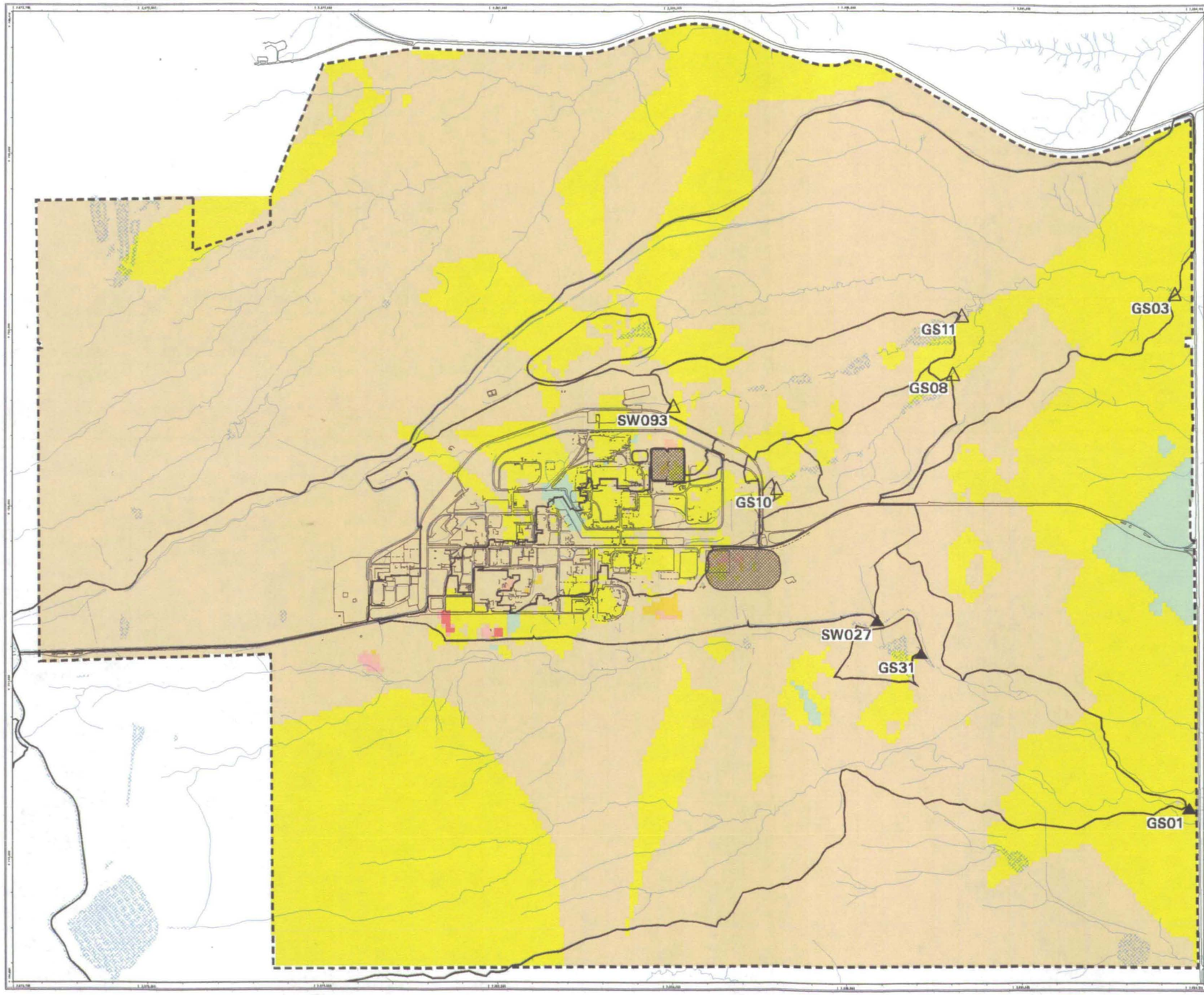

Figure 3-3

U-238 Activity in Surface Soils

Kriging Analysis Isoplot (pCi/g)

EXPLANATION

- Result < = 0.01
- 0.01 < Result < = 0.05
- 0.05 < Result < = 0.1
- 0.1 < Result < = 0.5
- 0.5 < Result < = 1.0
- 1.0 < Result < = 5.0
- 5.0 < Result < = 10.0
- 10.0 < Result < = 100.0
- 100.0 < Result < = 1000.0
- Result > 1000.0
- Areas altered by remediation or recontouring (approx.)
- Drainage Basin Boundary
- Walnut Creek Basin Gaging Station (GS03, GS08, GS10, GS11 & SW093)
- Woman Creek Basin Gaging Station (GS01, GS31 & SW027)

Standard Map Features

- Lakes and ponds
- Streams, ditches, or other drainage features
- Fences and other barriers
- Rocky Flats Environmental Technology Site boundary
- Paved roads

DATA SOURCE BASE FEATURES:

Buildings, fences, hydrography, roads and other structures from 1994 aerial fly-over data captured by EG&G RSL, Las Vegas. Digitized from the orthophotographs. 1/5b

Analytical Data provided by Mark Wood (KH, 303-966-6689).

Kriged data provided by Jeff Myers (Westinghouse-Akin, 803-502-9747).

Scale = 1 : 21330
1 inch represents approximately 1778 feet

250 0 500 1000 ft

State Plane Coordinate Projection
Colorado Central Zone
Datum: NAD27

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Rocky Flats Environmental Technology Site

URS Dept. 308-966-7/0.

Prepared by: CH2MHILL

Prepared for: KH

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3.2.1 Surface Water - Site Monitoring Data

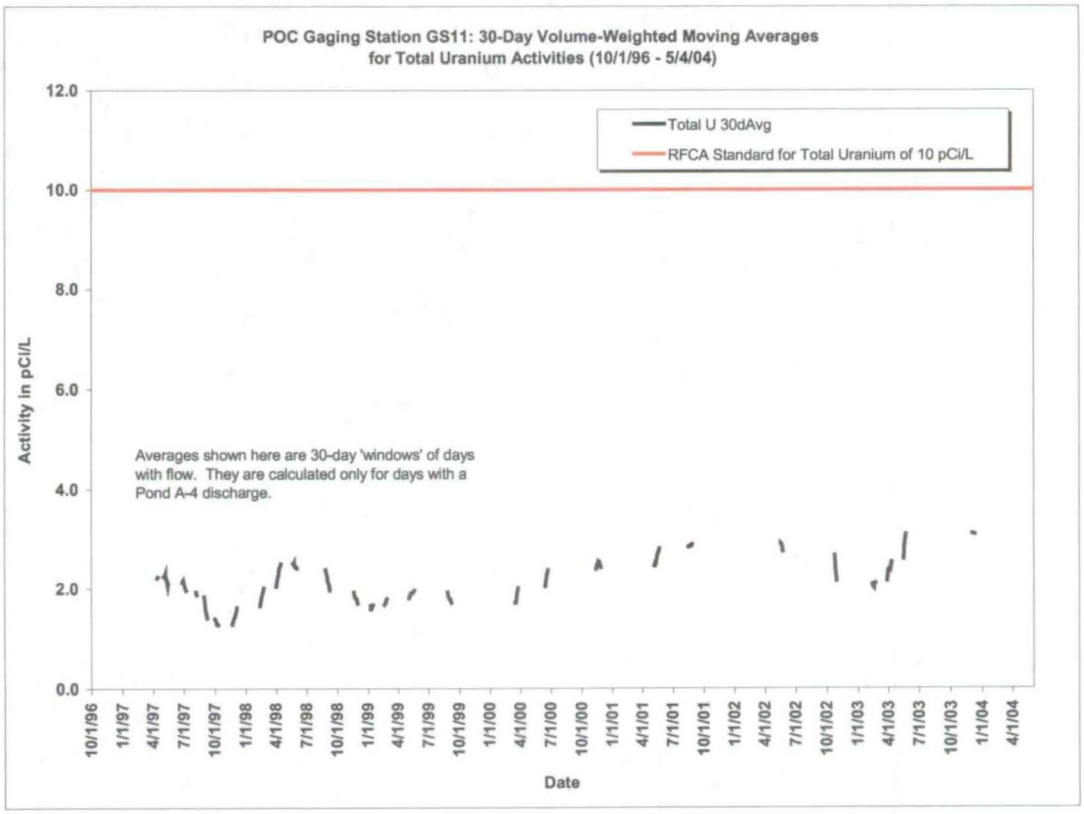
All RFCA surface water Point-of-Compliance (POC) monitoring stations are located downstream from the Site's Industrial Area (see listing of stations in Table 2-9). Since RFCA surface water monitoring was first implemented on October 1, 1996, the surface water quality at RFETS has been compliant with the applicable RFCA Action Levels in both Walnut and Woman Creeks. The Action Levels are 10 pCi/L for Walnut Creek and 11 pCi/L for Woman Creek.

Surface water samples are collected using automated sampling equipment and a flow-paced sampling protocol as specified in the RFETS *Integrated Monitoring Plan* (Kaiser-Hill, 2002c). Total uranium data for the five RFCA surface water POC stations are presented in Figure 3-4 through Figure 3-8.

In addition to the plots of the POC monitoring locations, a summary of total uranium data for all RFETS automated surface water sampling locations are presented in Table 3-1. Total uranium data presented include the median, 85th percentile, and maximum sample results. The median values are displayed on a map of the Site (Figure 3-9).

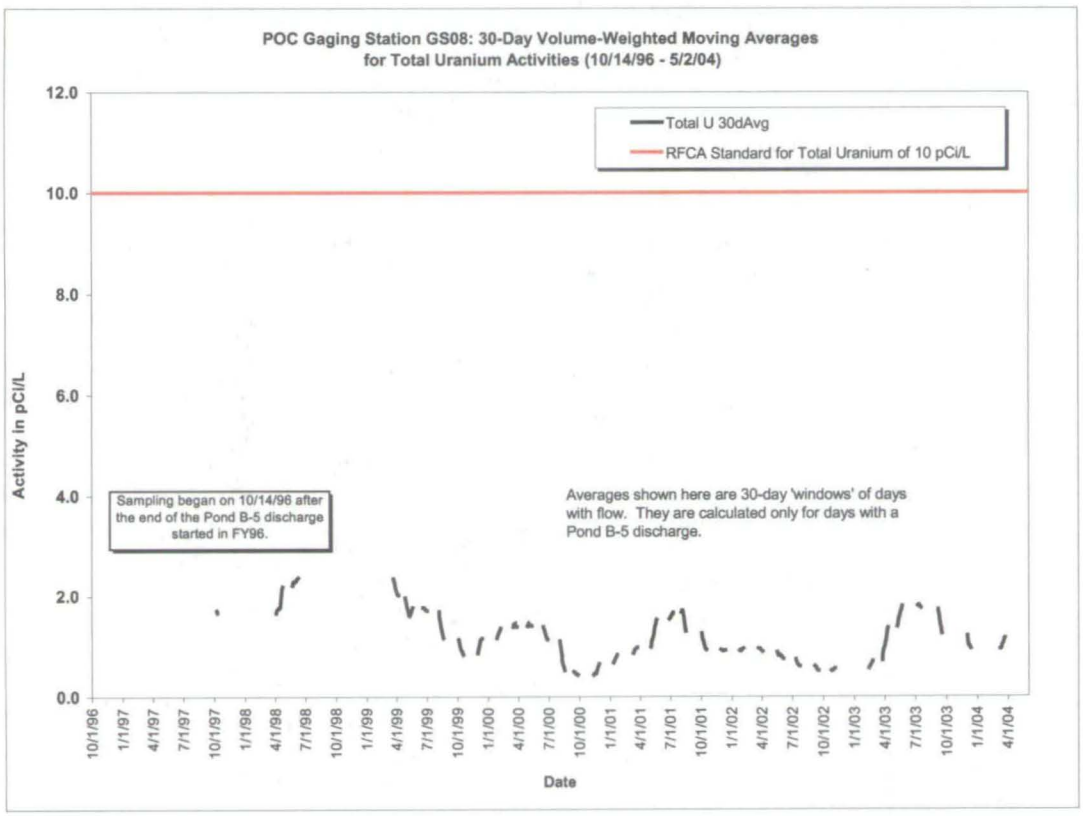
It is noted that all of the surface water monitoring locations listed in Table 3-1, except one, have a median total uranium concentration of less than 10 pCi/L (the RFCA standard in Walnut Creek). The one exception is station SW036, near the Original Landfill, with a median total uranium concentration of 30.430 pCi/L. For perspective on the amount of water contributed by the SW036 basin, the discharge volume at station SW036 has been less than 1 % of the volume measured at Woman Creek POC station GS01. Discharge volumes (188,416 cubic feet at SW036 versus 19,690,820 cubic feet at GS01), measured from 6/15/02 to 4/25/04, are based on data collected since station SW036 was installed. Therefore, the fraction of uranium load contributed by the SW036 watershed to the Woman Creek POC station is small, relative to the rest of the Woman Creek watershed.

Figure 3-4. North Walnut Creek Station GS11 (Pond A-4 Outfall) - Total Uranium (10/1/96 to 5/4/04) – 30-Day Volume-Weighted Average



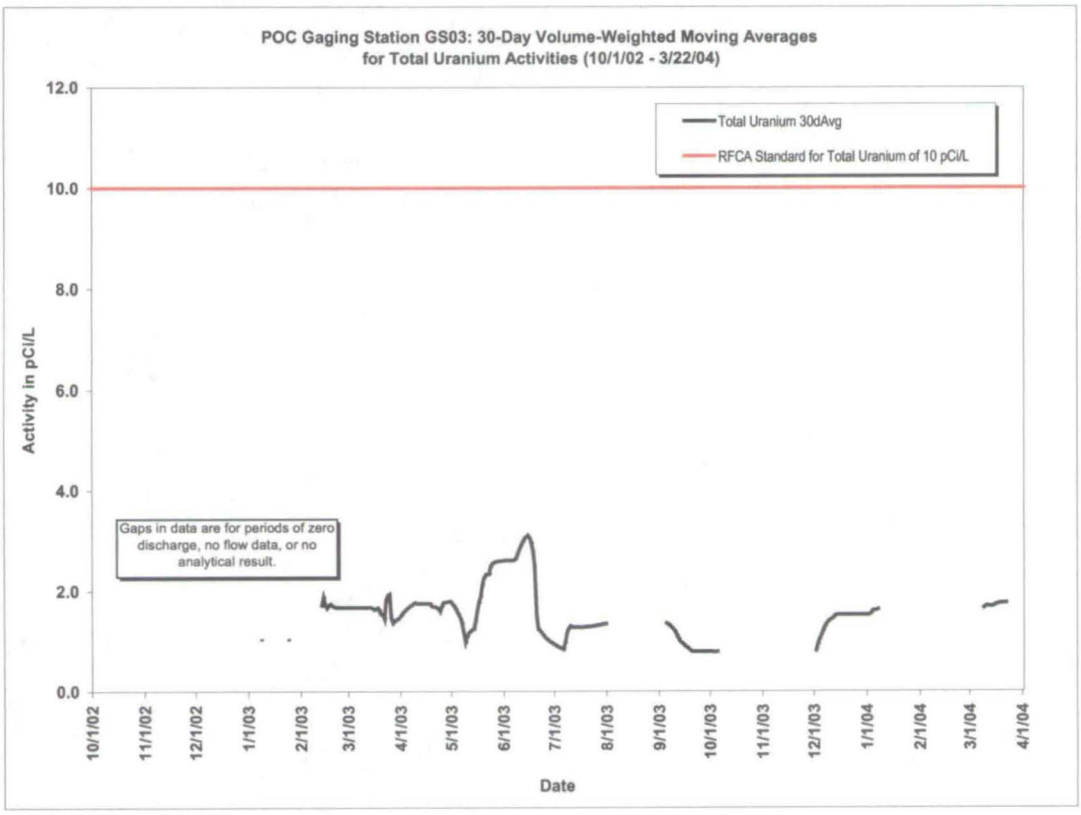
The RFCA standard for total uranium in Walnut Creek is 10 pCi/L (indicated by the red horizontal line). RFCA POC Monitoring location GS11 is located at the outfall of Pond A-4 on North Walnut Creek. The 30-day moving average of total uranium measured at GS11 has historically ranged from approximately 1 to 3 pCi/L. The GS11 data are intermittent because of the periodic nature of discharges from the pond.

Figure 3-5. South Walnut Creek Station GS08 (Pond B-5 Outfall) - Total Uranium (10/14/96 to 5/2/04) - 30-Day Volume-Weighted Average



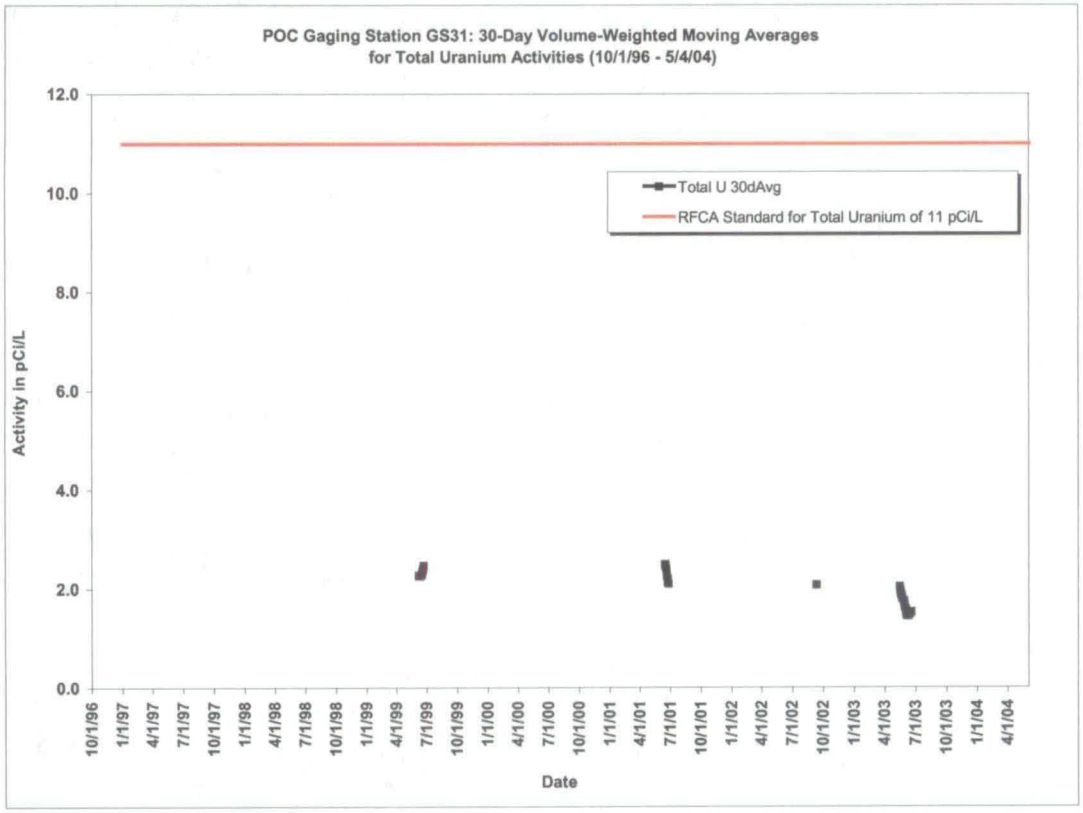
RFCA POC Monitoring location GS08 is located at the outfall of Pond B-5 on South Walnut Creek. The 30-day moving average of total uranium measured at GS08 is typically approximately 2 pCi/L or less. The GS08 data are intermittent because of the periodic nature of the discharges from the pond.

Figure 3-6. Walnut Creek Station GS03 (Woman Creek at Indiana Street) - Total Uranium (10/1/02 to 3/22/04) - 30-Day Volume-Weighted Average



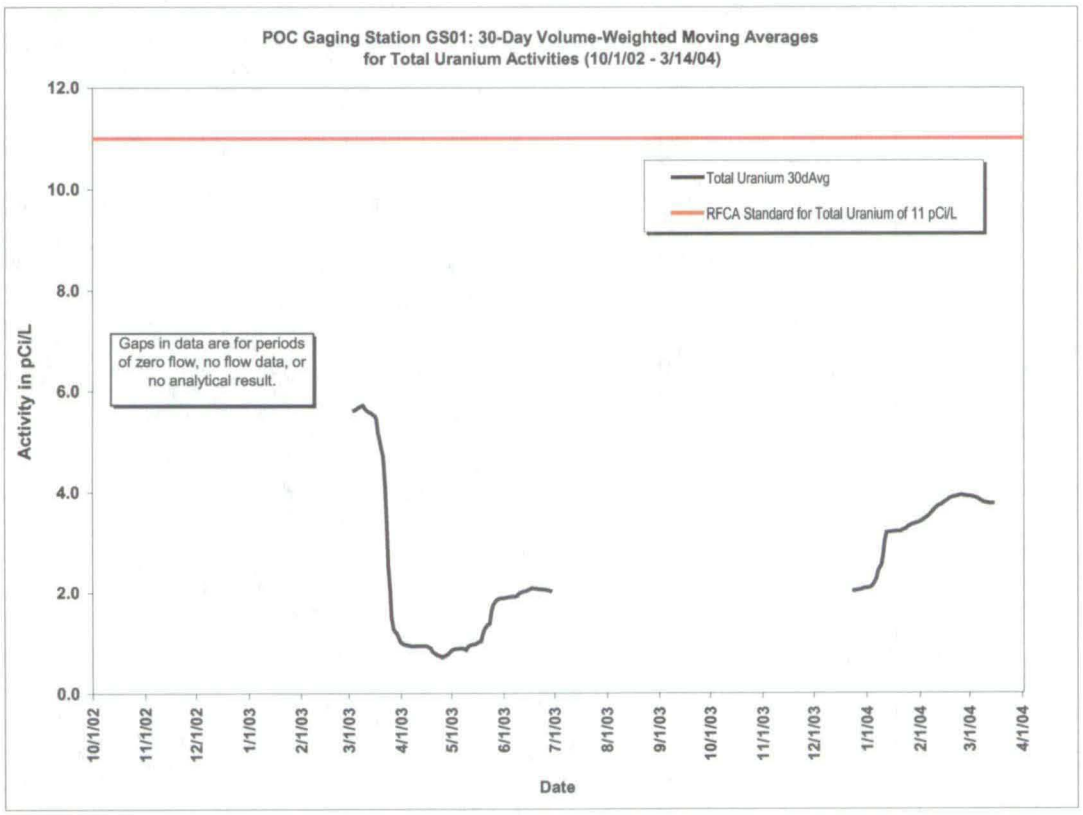
RFCA POC Monitoring location GS03 is located on Walnut Creek at Indiana Street. Uranium data were not collected at station GS03 using the flow-paced sampling protocol, as prescribed for the RFCA 30-day moving average value, until October 1, 2002. Data at GS03 are intermittent because of the periodic nature of discharges from Ponds A-4 and B-5 that are located upstream on North and South Walnut Creeks, respectively.

Figure 3-7. Station GS31 (Pond C-2 Outfall) - Total Uranium (10/1/96 to 5/4/04) - 30-Day Volume-Weighted Average



The RFCA standard for total uranium in Woman Creek is 11 pCi/L, as indicated by the red horizontal line. This compares to the Walnut Creek standard of 10 pCi/L. POC monitoring location GS31 is located in the Woman Creek watershed, at the outfall of Pond C-2, which receives flow from the South Interceptor Ditch. The GS31 data are intermittent because of the periodic nature of the discharges from the pond.

Figure 3-8. Station GS01 (Woman Creek at Indiana Street) - Total Uranium (10/1/02 to 3/14/04) - 30-Day Volume-Weighted Average



POC monitoring location GS01 is located on Woman Creek near Indiana Street at the RFETS boundary. Uranium data were not collected at this station using the flow-paced sampling protocol, as prescribed for the RFCA 30-day moving average value, until October 1, 2002. The GS01 uranium data are intermittent because of the periodic nature of the Woman Creek stream flows, as well as the periodic nature of the discharges from Pond C-2, which is located upstream.

Table 3-1. Surface Water Total Uranium – Data Summary for all RFETS Monitoring Locations

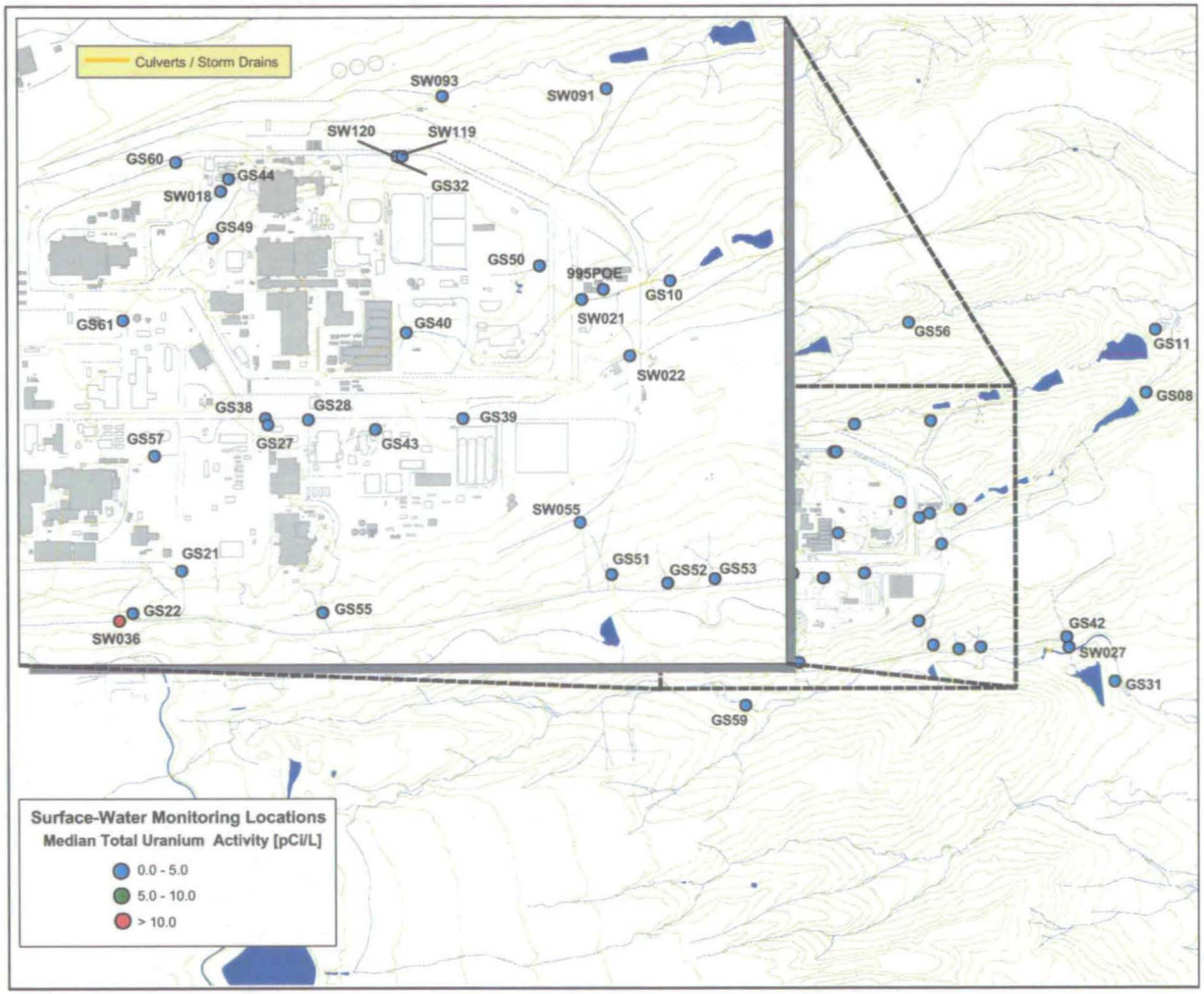
Location	POC Location	Samples [N]	Median [pCi/L]	85th Percentile [pCi/L]	Maximum [pCi/L]
GS01	x	25	2.505	4.124	5.885
GS03	x	39	1.572	2.188	3.266
GS08	x	97	1.204	1.965	4.579
GS10		225	3.055	4.345	7.200
GS11	x	84	2.045	3.072	4.055
GS21		13	0.593	1.423	28.660
GS22		24	1.167	3.344	7.315
GS27		71	0.359	1.179	3.889
GS28		10	0.819	1.083	1.150
GS31	x	22	2.283	2.633	3.917
GS32		75	1.906	3.962	21.219
GS38		22	0.593	0.868	1.324
GS39		16	0.379	1.206	2.997
GS40		53	3.097	4.183	11.264
GS42		6	0.189	0.292	0.300
GS43		24	2.042	10.550	23.088
GS44		37	2.343	4.455	5.240
GS49		28	0.384	1.475	2.933
GS50		8	0.313	0.398	0.438
GS51		10	1.261	2.538	2.758
GS52		6	2.659	3.150	3.778
GS53		4	1.221	2.667	3.668
GS54		2	0.113	0.126	0.131
GS55		34	3.253	5.271	9.448
GS56		8	1.554	2.960	3.702
GS57		27	0.500	0.816	5.014
GS59		13	0.570	1.303	3.866
GS60		5	0.444	0.587	0.625
GS61		4	0.593	0.668	0.726
SW018		4	2.161	2.488	2.584
SW021		8	3.705	6.077	13.249
SW022		38	0.809	1.266	2.987
SW027		55	1.615	3.017	4.476
SW036		13	30.430	37.086	39.620
SW055		14	1.208	2.941	4.502
SW091		14	2.745	4.248	5.878
SW093		246	2.694	4.166	6.640
SW119		15	2.701	7.480	10.628
SW120		30	3.544	7.394	10.338
995POE		40	0.371	1.079	1.835

Notes:

1) Data are preliminary and subject to review.

2) Dates of sample collection vary from location to location. Earliest sample date for any station is 10/1/96 (first day of RFCA sampling), through sample data received by May 24, 2004.

Figure 3-9. Median Total Uranium Concentration in Surface Water



3.2.2 Surface Water - LANL TIMS Analysis (Efurd et al.)

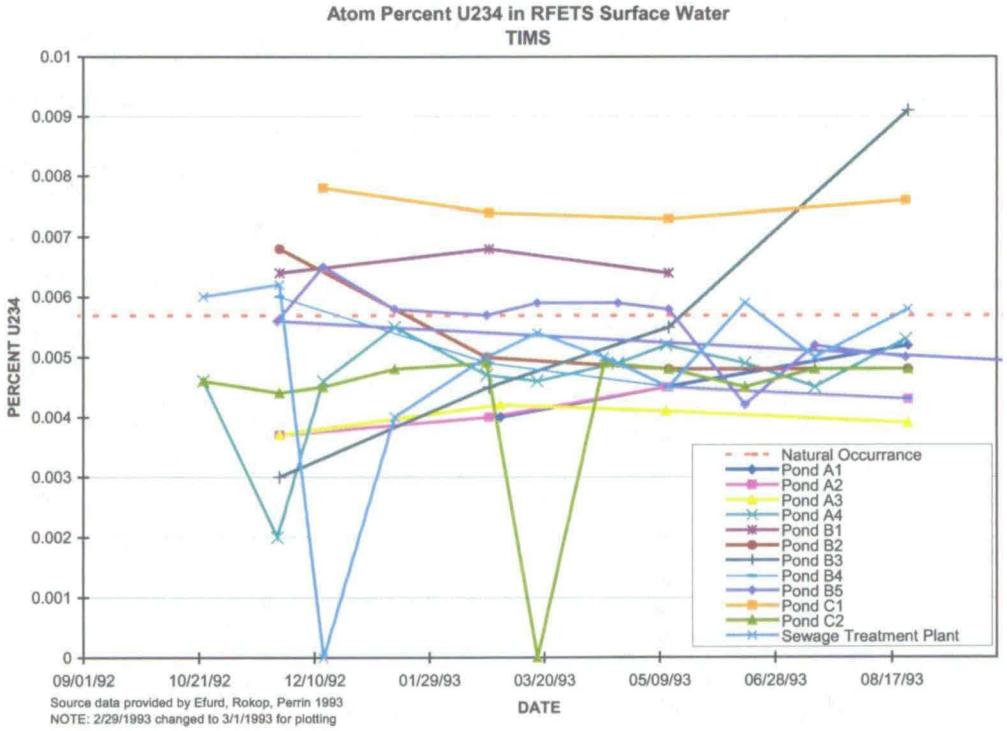
LANL researchers characterized the radioactivity in surface water and sediments of the RFETS ponds using TIMS (Efurd et al., 1993a). TIMS was used because it is more accurate than alpha-spectroscopy and provides more certain isotopic ratios. Data collection occurred in 1992 and 1993. Surface water samples were collected from Ponds A-1 through A-4, Ponds B-1 through B-5, Ponds C-1 and C-2, and the sewage treatment plant (STP) effluent. All uranium samples were analyzed using TIMS. Plots of sample results are presented for U-234, U-235, U-236, and U-238 in Figure 3-10 through Figure 3-13, respectively.

The authors concluded that upper Ponds A-1, A-2, A-3, B-1, B-2, B-3, and B-4 contain measurable quantities of depleted uranium. Essentially 100% of the uranium in Pond A-1 and Pond A-2 originated as depleted uranium. All of the other ponds, except Pond C-1, contained mixtures of naturally-occurring and depleted uranium, with concentrations ranging from 0.2 to 15.8 pCi/L. No depleted uranium was detected in Pond C-1.

In the terminal ponds, the authors concluded that the largest source of radioactivity is naturally-occurring uranium and its decay product, radium. For anthropogenic uranium, the largest source in the terminal ponds is depleted uranium. In the Walnut Creek basin, approximately 50% of the uranium in Pond A-4 and 20 % of the uranium in Pond B-5 water originated as depleted uranium. In the Woman Creek basin, approximately 50% of the uranium detected in the Pond C-2 water and 90% of the uranium detected in the sediment sample was associated with depleted uranium. Depleted uranium was also detected in soil samples collected from the South Interceptor Ditch.

In the effluent from the STP, approximately one third of the uranium present originated from depleted uranium. However, no anthropogenic uranium was detected in the influent or effluent from the STP (Building 124). For perspective, data collected since the Efurd study indicate the average annual volume-weighted concentration of total uranium in the STP effluent was 0.515 pCi/L for Water Years 2001 and 2002 (Kaiser-Hill, 2003d).

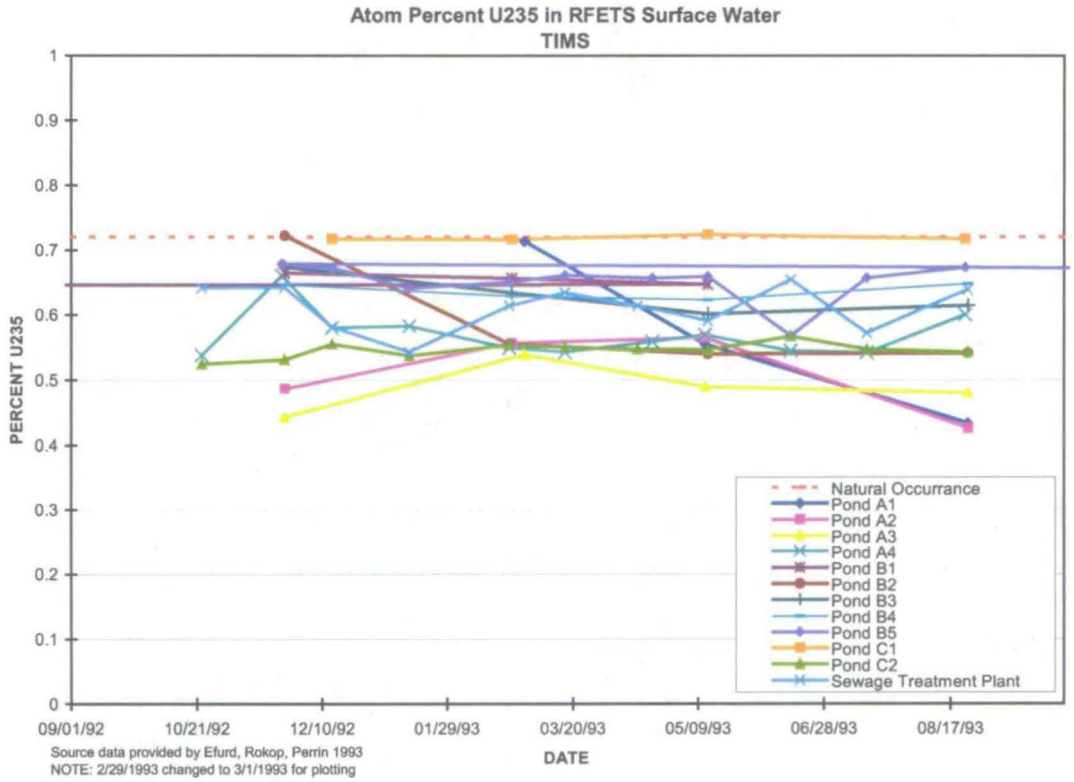
Figure 3-10. U-234 in RFETS Ponds – TIMS Analysis



TIMS analyses of water collected from the RFETS ponds provide data on the percentage, by mass, of U-234 in the different ponds. Naturally-occurring uranium has approximately 0.0057% U-234 by mass. For man-made forms, highly enriched uranium has approximately 1% U-234 by mass, whereas depleted uranium has less than 0.01%.

(Data source: Efurd et al., 1993a)

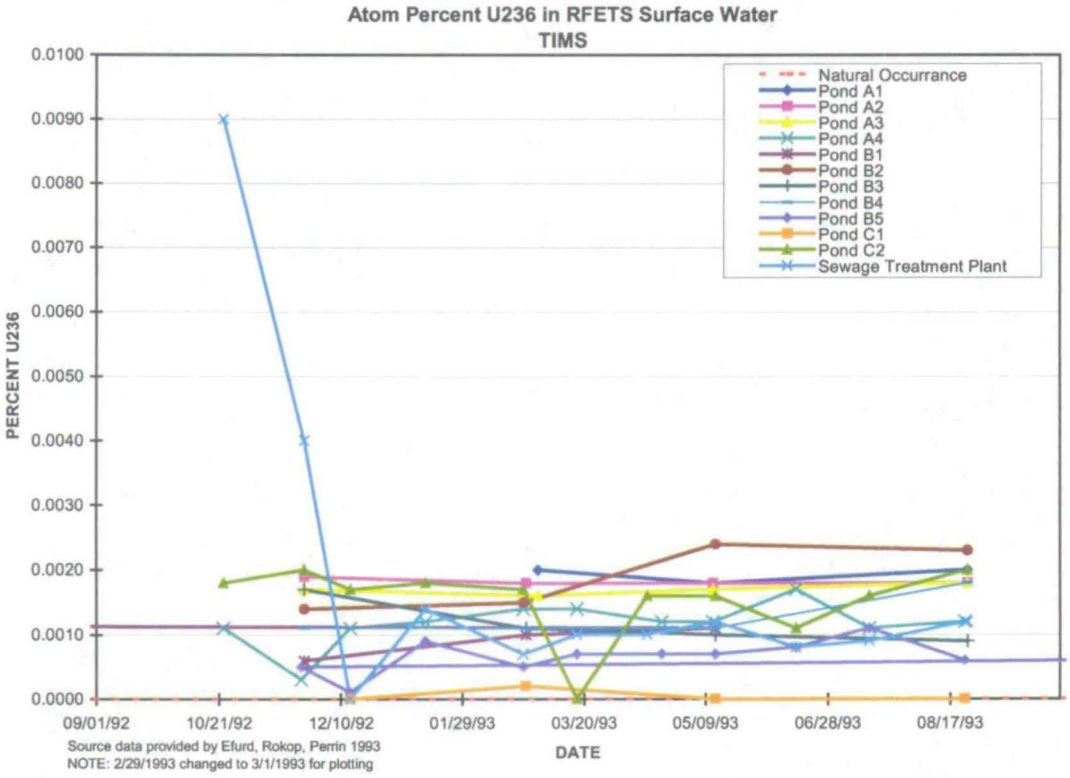
Figure 3-11. U-235 in RFETS Ponds – TIMS Analysis



TIMS analyses of water collected from the RFETS ponds provide data on the percentage, by mass, of U-235 in the different ponds. Naturally-occurring uranium has approximately 0.72% U-235 by mass. For man-made forms, highly enriched uranium has greater than 93% U-235 by mass, whereas depleted uranium has approximately 0.2%. Pond C-1 (orange line) appears to have the same approximate mass percentage of U-235 as is found naturally. The other ponds analyzed have mass percentages of U-235 that are less than occurs naturally and less than the percentage associated with highly enriched uranium, thereby indicating a depleted uranium signature.

(Data source: Efurd et al., 1993a)

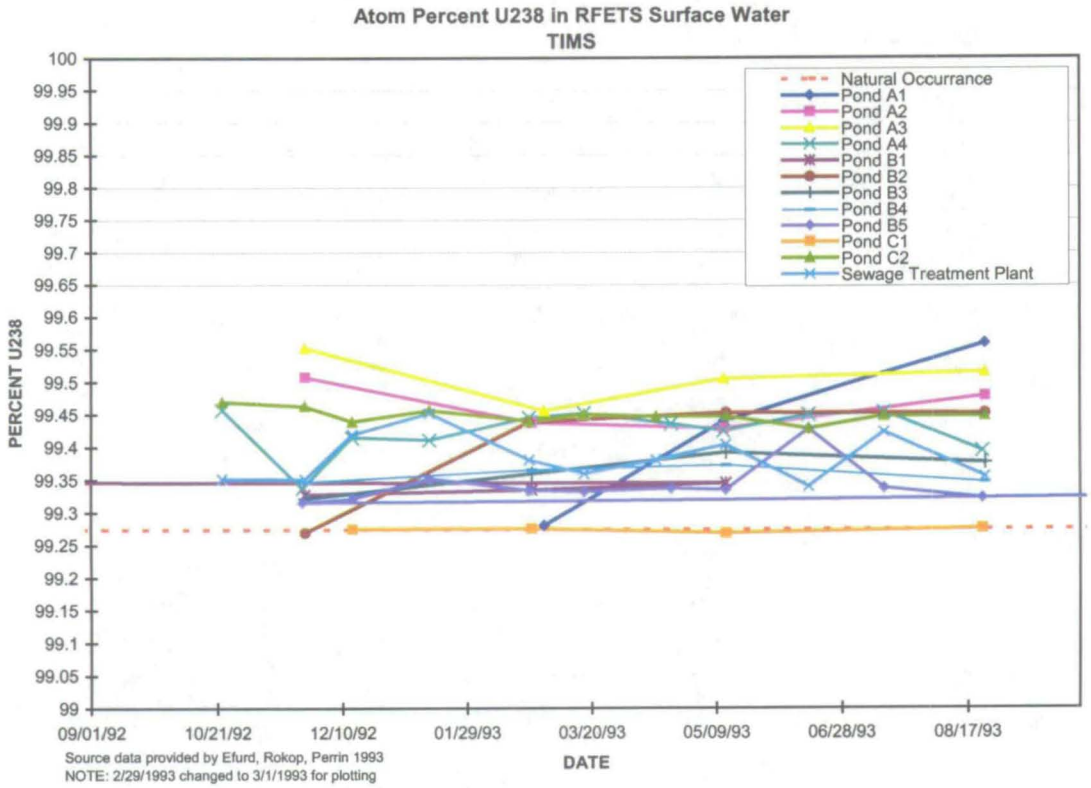
Figure 3-12. U-236 in RFETS Ponds – TIMS Analysis



TIMS analyses of water collected from the RFETS ponds provide data on the percentage, by mass, of U-236 in the different ponds. Naturally-occurring uranium has no U-236. Therefore, the presence of U-236 indicates uranium from a man-made source. Pond C-1 samples (orange line along the bottom) appear to have either no U-236 or very minor amounts. The other ponds analyzed have mass fractions of U-236 that are greater than zero, thereby indicating those ponds likely have uranium from a man-made source.

(Data source: Efurd et al., 1993a)

Figure 3-13. U-238 in RFETS Ponds – TIMS Analysis



TIMS analyses of water collected from the RFETS ponds provide data on the percentage, by mass, of U-238 in the different ponds. Naturally-occurring uranium has approximately 99.3% U-238 by mass. For man-made forms, highly enriched uranium has approximately 5.4% U-238 by mass, whereas depleted uranium has approximately 99.8%. Pond C-1 (orange line) appears to have the same approximate mass percentage of U-238 as is found naturally. The other ponds analyzed have mass percentages with more U-238 than occurs naturally, thereby indicating a depleted uranium signature.

(Data source: Efurd et al., 1993a)

3.2.3 Surface Water - LANL ICP/MS Analysis (Murrell and Brink)

Surface water samples collected at RFETS in January 2002 were analyzed by Murrell and Brink at LANL using High Resolution ICP/MS (Murrell and Brink, 2002a). Based on isotopic ratios, the authors determined whether uranium with an anthropogenic signature was detectable in the samples. Three of the sample locations, all in the Walnut Creek basin, had signatures associated with depleted uranium (see Table 3-2).

Of the two surface water POC locations analyzed using High Resolution ICP/MS (GS01 [Woman Creek at Indiana Street] and GS03 [Woman Creek at Indiana Street]), only GS03 has a signature associated with depleted uranium. The total uranium concentration of the GS03 sample was approximately 2.2 µg/L (approximately 1.5 pCi/L). For reference, this compares to the RFCA standard in that drainage of 10 pCi/L.

Table 3-2. Results Summary – LANL TIMS Analysis of Surface Water, 2002[1]

Sample	Location	Location Description	236/238	235/238	U (µg/l)	Man-Made U Signature?[2]
514-001	GS01	Woman Creek at Indiana St.	1.922E-06	0.007074	10.136891	No
514-002	GS03	Walnut Creek at Indiana St.	8.813E-06	0.006451	2.197284	Yes
514-003	GS04	Rock Creek at Hwy. 128	1.376E-07	0.007172	1.785421	No
514-004	GS05	Woman Creek at west boundary	-1.888E-06	0.006608	0.164187	No
514-005	GS10	S. Walnut Ck. Above Pond B-1	1.050E-05	0.006417	9.386207	Yes
514-009	GS10 (Duplicate)	S. Walnut Ck. Above Pond B-1	9.953E-06	0.006333	9.431062	Yes
514-006	GS13	N. Walnut Ck. Above Pond A-1	1.245E-05	0.006272	15.415099	Yes
514-007	GS17	Woman Creek above Pond C-1	1.727E-06	0.007106	2.613553	No
514-008	SW18	Ditch northeast of Building 371	2.690E-07	0.007336	4.864854	No

Notes:

[1]Analyses conducted in conformance with EPA Method 6020A. It is noted that method conformance for U-234 and U-236 was not readily achievable.

[2]Anthropogenic signature as noted in LANL report (Murrell and Brink, 2002a). All anthropogenic signatures associated with depleted uranium.

3.3 GROUNDWATER

Four sources of RFETS groundwater uranium data are presented. They are:

1) LANL analysis of groundwater samples using ICP/MS (Murrell and Brink, 2000a; 2000b; and 2002b) (Section 3.3.1);
2) LANL analysis of groundwater samples using TIMS (Section 3.3.2);
3) USGS model analysis of groundwater uranium transport in the Solar Evaporation Ponds area (Ball, 2000) (Section 3.3.3); and
4) Isotopic uranium groundwater data from the Soil/Water Database (Section 3.3.4).

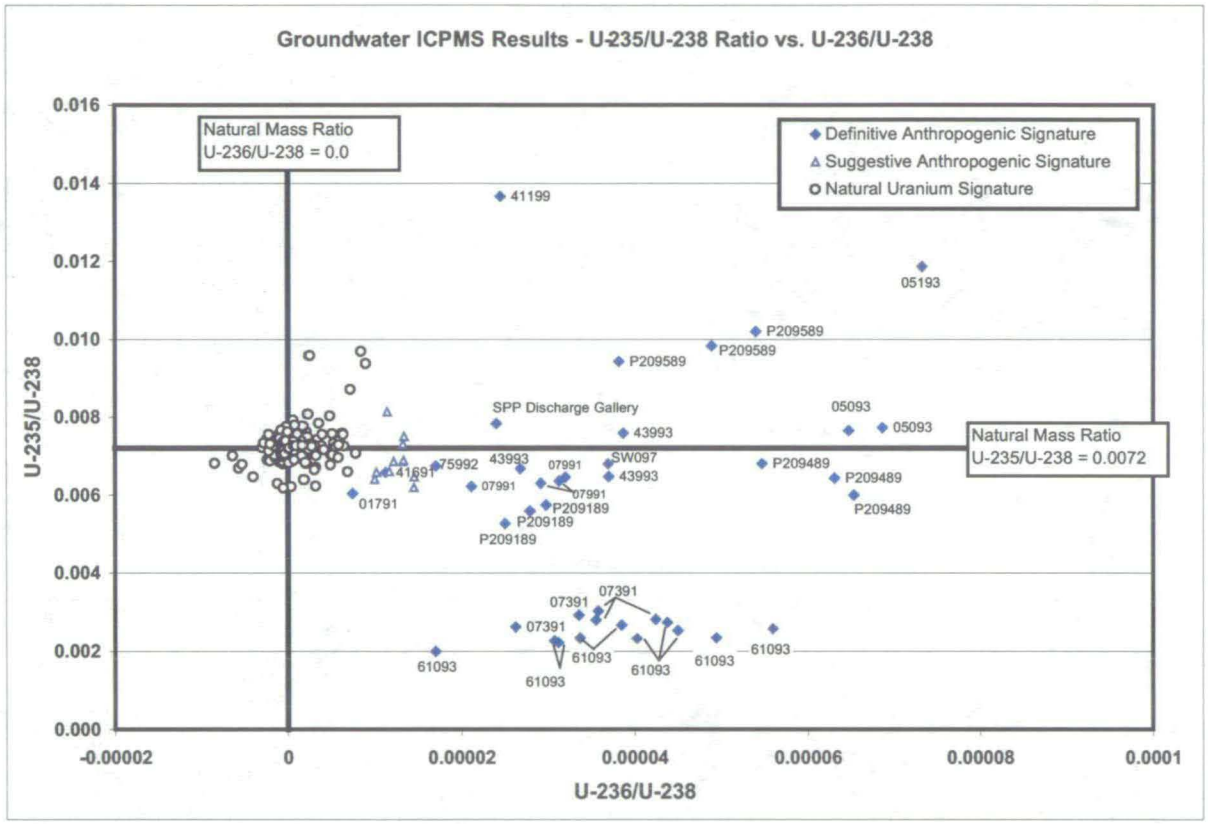
3.3.1 Groundwater - LANL ICP/MS Analysis (Murrell and Brink)

To determine anthropogenic versus natural uranium in groundwater, a sampling and analysis plan was developed by the Colorado Department of Public Health and Environment (CDPHE), in coordination with Site personnel, to analyze samples using High Resolution ICP/MS (Pottorff, 2004). Groundwater samples were collected in 1999, 2000, and 2002, and analyzed at LANL (Murrell and Brink, 2000a; Murrell and Brink, 2000b; Murrell and Brink, 2001; Murrell and Brink, 2002b).

A three-isotope plot of the ICP/MS data is displayed on Figure 3-14. A description of the plot, and the interpretation of the data, is provided with the figure. For each sample result, LANL personnel determined whether the uranium isotopic signature was "definitive anthropogenic", "suggestive anthropogenic", or "non-anthropogenic [i.e., "natural"]. On Figure 3-14, sample locations with a "definitive anthropogenic" signature are labeled with the well number from where the sample was collected. Some wells were sampled multiple times. The majority of data points on Figure 3-14 are clustered near the intersection of the natural ratios, indicating the uranium is from a natural source.

Figure 3-15 is a map of RFETS with the High Resolution ICP/MS sample results, with a symbol for each location indicating a "definitive", "suggestive", or "natural" signature.

Figure 3-14. Groundwater ICP/MS Analysis – Uranium Three-Isotope Plot



The bold horizontal line indicates the natural uranium ratio of U-235/U-238 – a greater distance above the line indicates a higher proportion of U-235 (i.e., enriched uranium signature). A greater distance below the line indicates a higher proportion of U-238 (i.e., depleted uranium signature). The bold vertical line indicates the natural uranium ratio of U-236/U-238. Since natural uranium has no U-236, points further from the vertical represent a more definitive man-made signature. The intersection of the vertical and horizontal lines, with the large cluster of data points, is representative of natural uranium.

Notes: The "Definitive Anthropogenic Signature", "Suggestive Anthropogenic Signature", and "Natural Uranium Signature" symbols are based on the interpretation of LANL personnel, with the following modifications: 1) Location 05193 (upper right of plot) was assigned a "definitive" symbol, instead of the "natural" label originally assigned, because the ratios indicated "definitive", 2) Location SW097 (in the center of plot) was assigned a "definitive" symbol, instead of a "suggestive signature", because the ratios were similar to other "definitive signature" locations; and 3) Location B208189 (in cluster of "natural" data points) was assigned a "natural" symbol, instead of a "definitive" symbol, because the ratios (U236/U238 ratio: -8.43E-7, and U235/U238 ratio: 0.007271) placed it at the intersection of the natural ratios.

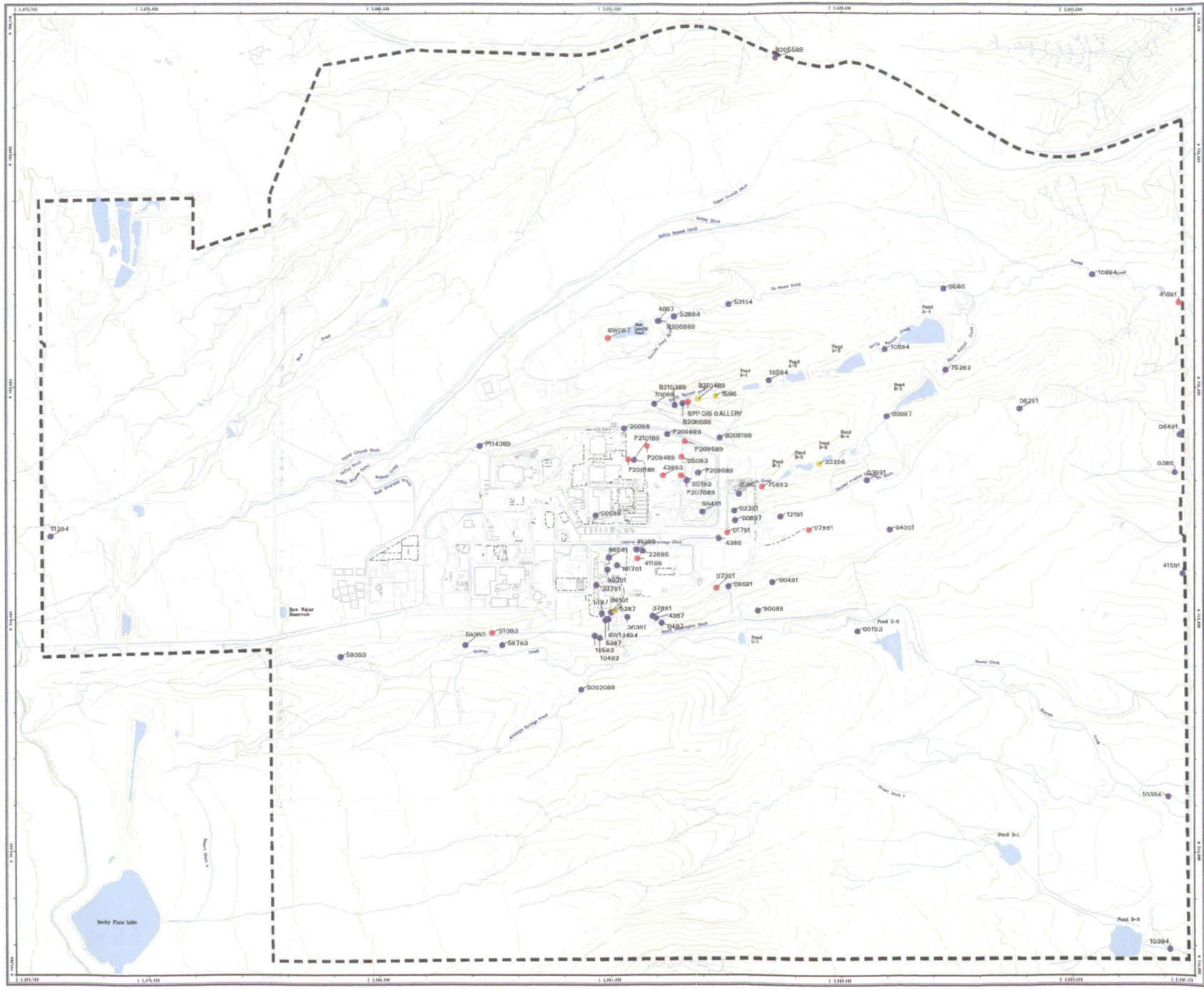


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Figure 3-15

Groundwater Analysis
LANL High Resolution ICP/MS

EXPLANATION

- Natural Uranium Signature (with Sample Location)
- Suggestive Anthropogenic Signature (with Sample Location)
- Definitive Anthropogenic Signature (with Sample Location)

Standard Map Features

- Buildings and other structures
- Lakes and ponds
- Streams, ditches, or other drainage features
- Fences and other barriers
- Topographic Contour (20-Foot)
- Rocky Flats Environmental Technology Site boundary
- Roads

DATA SOURCE BASE FEATURES:
Buildings, fences, hydrography, roads and other structures from 1994 aerial flyover data captured by EG&G RSL, Las Vegas. Digitized from the orthophotographs. 1995.
Topographic contours were derived from digital elevation model (DEM) data by Morrison Knudsen (MK) using ESRI Arc TIN and LATTICE to process the DEM data to create 5-foot contours. The DEM data was captured by the Remote Sensing Lab, Las Vegas, NV, 1994 Aerial Flyover, at ~ 30 meter resolution. DEM post processing performed by MK, Winter 1997.

NOTES:
Detection of an anthropogenic uranium signature only indicates the nature of the uranium source material. It does not indicate the concentration of uranium observed in the groundwater.
The "Natural", "Suggestive", and "Definitive" symbols are based on the interpretation of LANL personnel, with the following modifications:

- Location 05166 assigned a "definitive" symbol, instead of "natural" label originally assigned, because isotopic ratios indicated "definitive."
- Location SW087 assigned a "definitive" symbol, instead of a "suggestive signature", because isotopic ratios similar to other "definitive" wells.
- Location B206189 assigned a "natural" symbol, instead of definitive" symbol, because the ratios (U236/U238: -8.42E-7, and U235/U238: 0.007271) indicated "natural."
- Locations 05287 and 01586 assigned a "suggestive" symbol, instead of "natural", based on TIMS analysis results.

Note: If more than one sample was analyzed for one location, with different results (i.e., different signature types), then the more stringent of the signature types was applied to this map.

Scale = 1 : 11330
1 inch represents approximately 1778 feet

State Plane Coordinate Projection
Colorado Central Zone
Datum: NAD27

U.S. Department of Energy
Rocky Flats Environmental Technology Site

GIS Dept 303-966-7707

Prepared by: CH2MHILL

Prepared for: KAISER-HILL

June 28, 2004

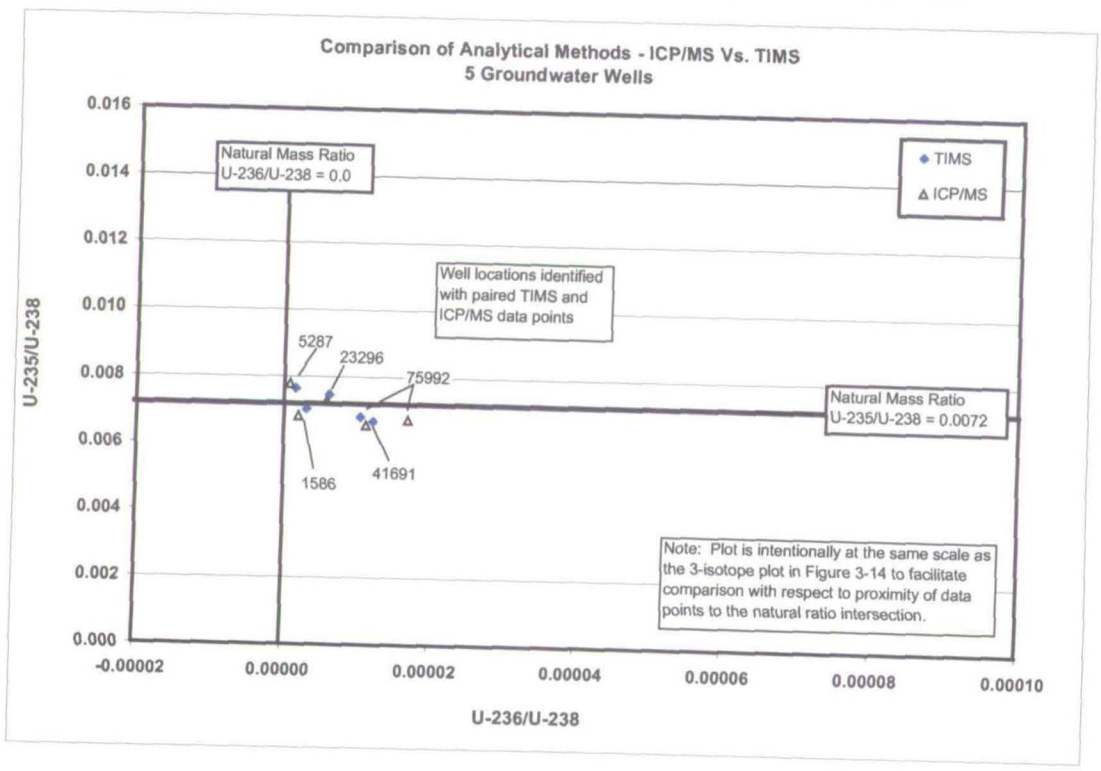
NT_Srv w:/projects/fy2003/03-0385/murrell_analysis_fig3-15.aml

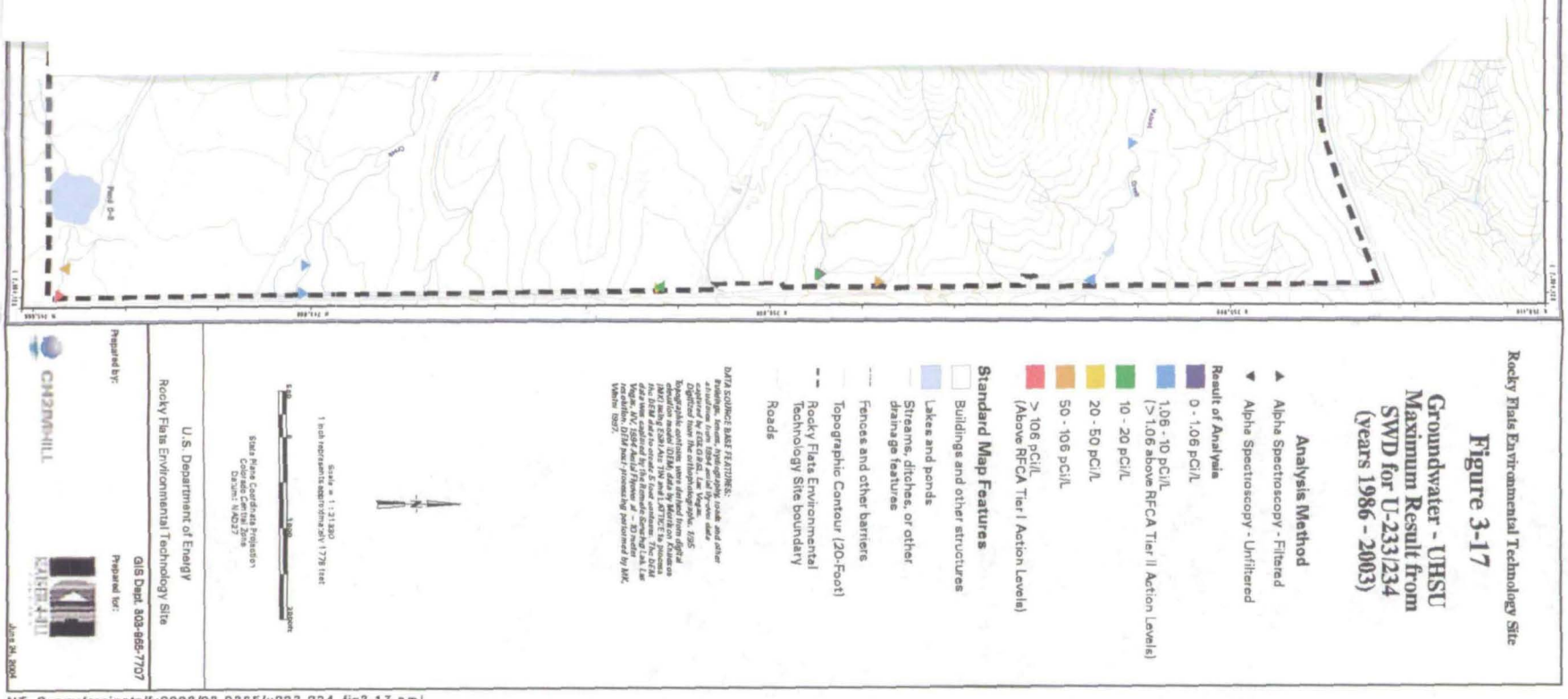
3.3.2 Groundwater – LANL TIMS Analysis (Murrell and Brink)

A second phase of the CDPHE anthropogenic uranium analysis involved analyzing groundwater wells with low concentrations of U-236, using both High Resolution ICP/MS and TIMS (Pottorff, 2004). The TIMS analysis was also performed at LANL. A three-isotope plot comparing the High Resolution ICP/MS and TIMS results from five wells is shown on Figure 3-16. As shown on the figure, the results for ICP/MS and TIMS are comparable, even at U-236/U-238 mass ratios in the 2×10^{-6} range.

The TIMS results support the High Resolution ICP/MS data that indicate slight signatures of anthropogenic uranium at these locations. Figure 3-16 is plotted with the same scales as Figure 3-14 to illustrate the relative proximity of these data points to the intersection of the natural ratios.

Figure 3-16. Groundwater Isotope Analysis – TIMS Versus ICP/MS





NT_Svr w:/projects/fy2003/03-0385/u233-234_fig3-17.aml

3.3.2 Groundwater – LANL TIMS Analysis (Murrell and Brink)

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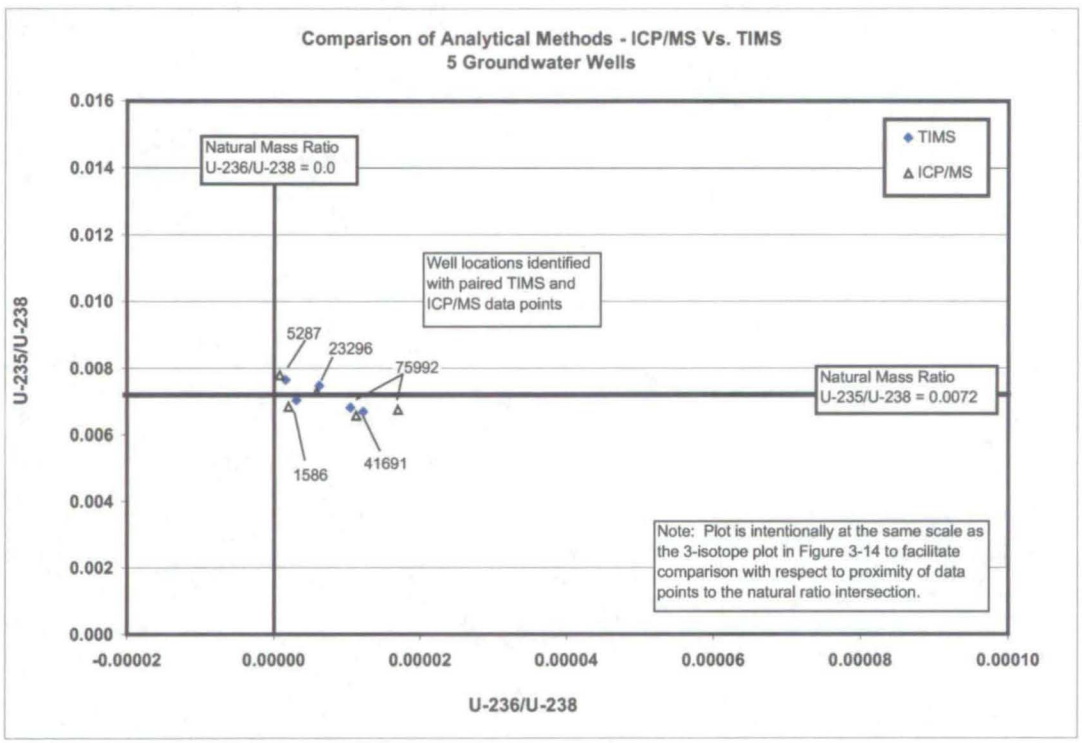

3.3.3 Groundwater - USGS Analysis (James W. Ball)

Ball reported on results of computer modeling uranium speciation in groundwater in the Solar Ponds plume area using a chemical speciation code (WATEQ4F) for natural waters (Ball, 2000 [unpublished USGS letter report]). The model used analyses of 950 groundwater well samples. Data were analyzed to determine saturation indices for major ions and to identify solubility controls for dissolved groundwater constituents. Study findings included:

- Uranium is migrating as a dissolved component of the groundwater plume;
- Above pH 6, all dissolved uranium species are undersaturated with respect to uranium minerals in the subsurface. This implies that, under the pH and redox conditions studied, there are no obvious solubility controls on mobility of uranium in the groundwater of the Solar Ponds Plume, and suggests that any transport retardation that may occur is due to sorption processes; and
- The dissolved uranium plume has not migrated as far from the Solar Ponds source area as has the dissolved nitrate plume. However, the extent this is due to retardation of the uranium plume is not known at present. Dates when nitrate disposal occurred are not known and may have been at different times than the uranium disposal. In addition, it is possible that other factors play a role, such as differing dispersivities of uranium and nitrate and selective microbial reduction of uranium.

3.3.4 Groundwater - Soil Water Database

Groundwater isotopic uranium data queried from the RFETS Soil/Water Database, for wells throughout the Site, are displayed in Figure 3-17 through Figure 3-20. Data are for samples collected from 1986 to 2003, depending on the isotope, as indicated on the figures. For each well, the value plotted is the maximum sample result measured at that location. In addition, for U-233/234, U-235, and U-238, the data are grouped to display values that exceed their respective RFCA Tier I and Tier II Action Levels, which vary for each isotope, as presented in Section 2.5.3.2.

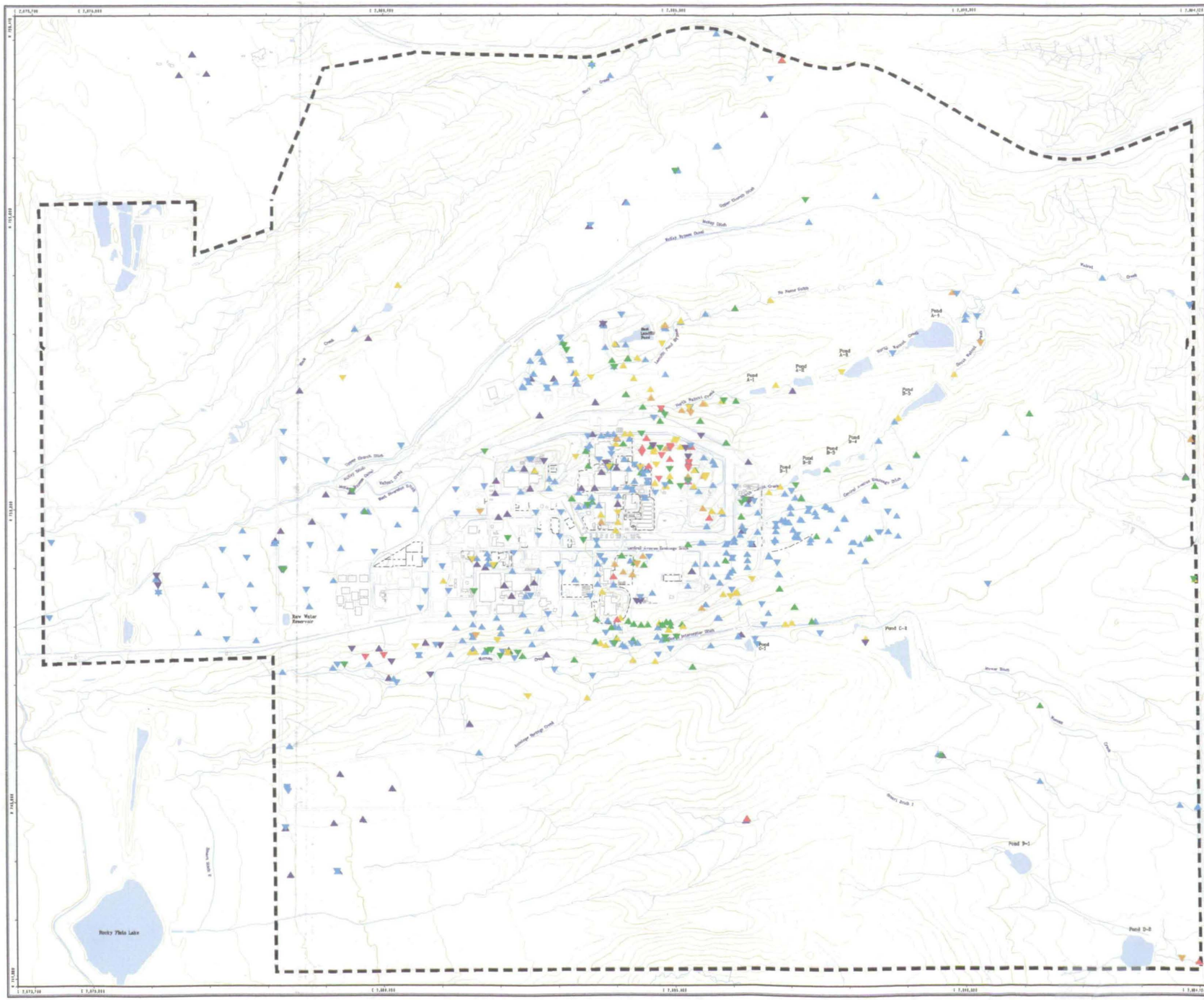

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Figure 3-17

Groundwater - UHSU Maximum Result from SWD for U-233/234 (years 1986 - 2003)

Analysis Method

- ▲ Alpha Spectroscopy - Filtered
- ▼ Alpha Spectroscopy - Unfiltered

Result of Analysis

- 0 - 1.06 pCi/L
- 1.06 - 10 pCi/L (> 1.06 above RFCA Tier II Action Levels)
- 10 - 20 pCi/L
- 20 - 50 pCi/L
- 50 - 106 pCi/L
- > 106 pCi/L (Above RFCA Tier I Action Levels)

Standard Map Features

- Buildings and other structures
- Lakes and ponds
- Streams, ditches, or other drainage features
- Fences and other barriers
- Topographic Contour (20-Foot)
- Rocky Flats Environmental Technology Site boundary
- Roads

DATA SOURCE BASE FEATURES:
Buildings, fences, hydrography, roads and other structures from 1994 aerial fly-over data captured by EG&G RSL, Las Vegas. Digitized from the orthophotographs. 1995 Topographic contours were derived from digital elevation model (DEM) data by Morrison Knudsen (MK) using ESRI Arc TIN and LATTICE to process the DEM data to create 5 foot contours. The DEM data was captured by the Remote Sensing Lab, Las Vegas, NV, 1994 Aerial Flyover at ~ 10 meter resolution. DEM post-processing performed by MK, Winter 1997.

Scale = 1 : 21,330
1 inch represents approximately 1778 feet

500 0 1000 2000ft

State Plane Coordinate Projection
Colorado Central Zone
Datum: NAD27

U.S. Department of Energy
Rocky Flats Environmental Technology Site

GIS Dept. 303-966-7707

Prepared by: CH2MHILL

Prepared for: KAISER-HILL

June 24, 2004

NT_Svr w:/projects/fy2003/03-0365/u233-234_fig3-17.aml

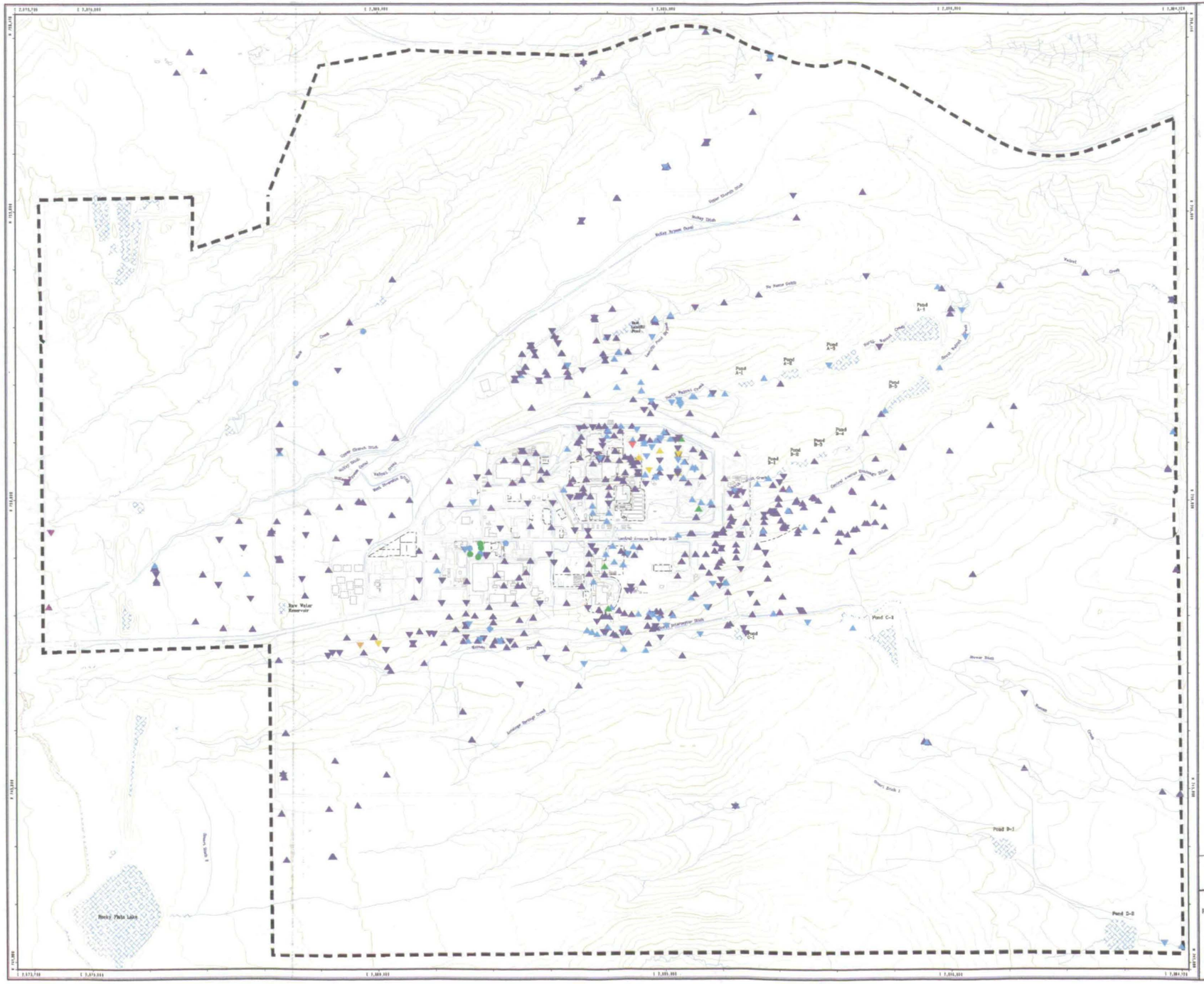

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Figure 3-18

Groundwater - UHSU Maximum Result from SWD for U-235 (years 1987 - 2003)

Analysis Method

- ▲ Alpha Spectroscopy - Filtered
- ▼ Alpha Spectroscopy - Unfiltered
- ◆ ICPMS (EPA SW-846 Method 6020A)
- ● Gamma Spectroscopy

Result of Analysis

- 0 - 1.01 pCi/L
- 1.01 - 10 pCi/L (> 1.01 above RFCA Tier II Action Levels)
- 10 - 20 pCi/L
- 20 - 50 pCi/L
- 50 - 101 pCi/L
- > 101 pCi/L (Above RFCA Tier I Action Levels)

Standard Map Features

- Buildings and other structures
- Lakes and ponds
- Streams, ditches, or other drainage features
- Fences and other barriers
- Topographic Contour (20-Foot)
- Rocky Flats Environmental Technology Site boundary
- Roads

DATA SOURCE BASE FEATURES:
Buildings, fences, hydrography, roads and other structures from 1994 aerial fly-over data captured by CGI/GPSL, Las Vegas. Digitized from the orthophotographs. 1:95
Topographic contours were derived from digital elevation model (DEM) data by Martin-Marietta Energy Systems (MK) using ESRI Arc TIN and LATTICE to process the DEM data to create 5-foot contours. The DEM data was acquired by the Remote Sensing Lab, Las Vegas, NV. 1994 Aerial Flyover at ~ 10 meter resolution. DEM post-processing performed by MK, Winter 1997.

Scale = 1 : 21,330
1 inch represents approximately 1778 feet

500 0 1000 2000ft

State Plane Coordinate Projection
Colorado Central Zone
Datum: NAD27

U.S. Department of Energy
Rocky Flats Environmental Technology Site

GIS Dept. 303-966-7707

Prepared by: CH2MHILL

Prepared for: KAISER-HILL

June 24, 2004

NT_Svr w:\projects\fy2003\03-0365\u235_fig3-18.aml

Rocky Flats Environmental Technology Site

Figure 3-19

Groundwater - UHSU Maximum Result from SWD for U-236 (years 1999 - 2000)

Analysis Method

- ICPMS (EPA SW-846 Method 6020A)

Result of Analysis

- 0
- 0.001 - 0.99 ppb
- 1.0 - 5.0 ppb

Standard Map Features

- Buildings and other structures
- Lakes and ponds
- Streams, ditches, or other drainage features
- Fences and other barriers
- Topographic Contour (20-Foot)
- Rocky Flats Environmental Technology Site boundary
- Roads

DATA SOURCE BASE FEATURES:
Buildings, fences, hydrography, roads and other structures from 1994 aerial fly-over data captured by EG&G RSL, Las Vegas.
Digitized from the orthophotographs. 1/95
Topographic contours were derived from digital elevation model (DEM) data by Morrison Knudsen (MK) using ESRI Arc TIN and LATTICE to process the DEM data to create 5 foot contours. The DEM data was captured by the Remote Sensing Lab, Las Vegas, NV, 1994 Aerial Flyover at ~ 10 meter resolution. DEM post-processing performed by MK, Winter 1997.

N

Scale = 1 : 21330
1 inch represents approximately 1778 feet

State Plane Coordinate Projection
Colorado Central Zone
Datum: NAD27

U.S. Department of Energy
Rocky Flats Environmental Technology Site

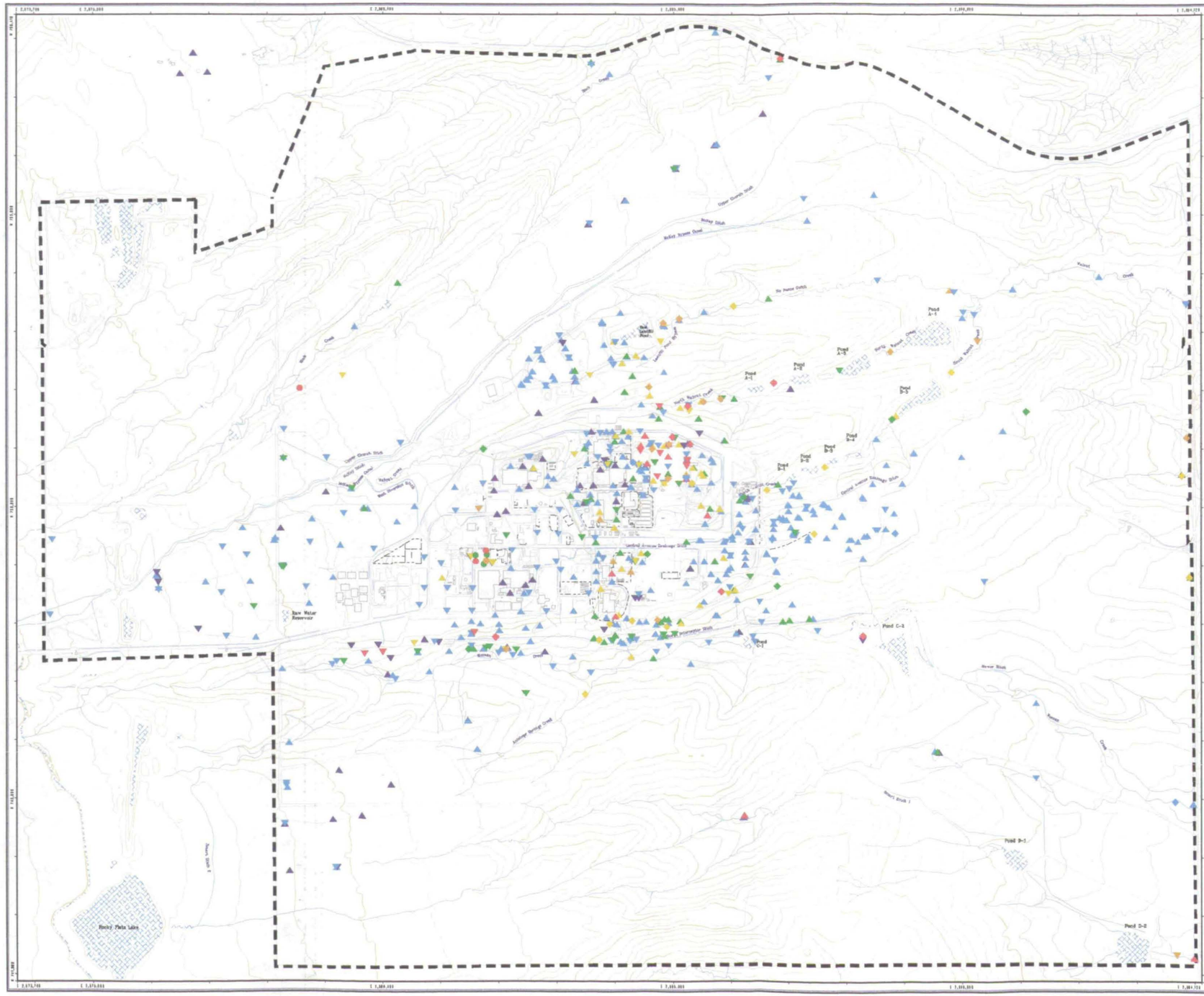
GIS Dept. 303-966-7707

Prepared by: CH2MHILL

Prepared for: KAISER-HILL

June 24, 2004

NT_Svr w:/projects/fy2003/03-0365/u236_fig3-19.aml



Rocky Flats Environmental Technology Site

Figure 3-20

Groundwater - UHSU Data Maximum Result from SWD for U-238 (years 1986 - 2003)

Analysis Method

- ▲ Alpha Spectroscopy - Filtered
- ▼ Alpha Spectroscopy - Unfiltered
- ◆ ICPMS (EPA SW-846 Method 6020A)
- ● Gamma Spectroscopy

Result of Analysis

- 0 - 0.768 pCi/L
- 0.768 - 10 pCi/L (>0.768 above RFCA Tier II Action Levels)
- 10 - 20 pCi/L
- 20 - 50 pCi/L
- 50 - 76.8 pCi/L
- > 76.8 pCi/L (Above RFCA Tier I Action Levels)

Standard Map Features

- Buildings and other structures
- Lakes and ponds
- Streams, ditches, or other drainage features
- Fences and other barriers
- Topographic Contour (20-Foot)
- Rocky Flats Environmental Technology Site boundary
- Roads

DATA SOURCE BASE FEATURES:
Buildings, fences, hydrography, roads and other structures from 1994 aerial fly-over data captured by EG&G R&L, Las Vegas. Digitized from the orthophotographs. 1/94.
Topographic contours were derived from digital elevation model (DEM) data by Morrison Knudsen (MK) using ESRI Arc TIN and LATTICE to process the DEM data to create 5-foot contours. The DEM data was captured by the Remote Sensing Lab, Las Vegas, NV. 1994 Aerial Flyover at ~ 10 meter resolution. DEM post-processing performed by MK, Vista 1997.

Scale = 1 : 21330
1 inch represents approximately 1778 feet

State Plane Coordinate Projection
Colorado Central Zone
Datum: NAD27

U.S. Department of Energy
Rocky Flats Environmental Technology Site

GIS Dept. 303-966-7707

Prepared by: CH2MHILL

Prepared for: KAISER-HILL

June 24, 2004

NT_Ser w:/projects/fy2003/03-0985/u238_fig3-20.aml

4.0 SUMMARY AND IMPLICATIONS FOR SITE CLOSURE

4.1 GENERAL URANIUM OCCURRENCE AND CHEMISTRY

Uranium occurs naturally in the Earth's crust and is ubiquitous in the Front Range of Colorado (Langmuir, 1997). The presence of relatively large amounts of naturally-occurring uranium can complicate studies to identify uranium from anthropogenic sources. High relative concentrations of uranium in the environment do not necessarily indicate an anthropogenic uranium source; high uranium concentrations in various environmental media at RFETS can frequently be attributed to natural sources.

Naturally-occurring uranium contains percentages of the isotopes U-234, U-235, and U-238 that are essentially fixed, regardless of the uranium concentration. These constant ratios provide insight whether uranium detected in a sample originates from a natural source, or an anthropogenic one. The mass ratio of U-235/U-238 provides the most reliable determination of the source of the uranium, whether it be natural or anthropogenic (depleted or enriched). In addition, both the U-233 and U-236 isotopes indicate uranium from an anthropogenic source, with U-236 typically used as a marker because U-233 is generally present in very low concentrations and is difficult to detect. Detection of U-236 and accurate assessment of uranium mass ratios requires the use of a suitable analytical method, such as High Resolution ICP/MS or TIMS. Both of these methods were employed at RFETS to distinguish between natural and anthropogenic uranium, at specific locations, in surface water and groundwater.

Uranium can exist in the III, IV, V, and VI oxidation states, but is most commonly found in the environment as U(IV) and U(VI). Of these, U(IV) is relatively insoluble and immobile due to its low aqueous solubility and tendency to strongly sorb onto soil media (Langmuir, 1997; Cleveland et al., 1976; and Honeyman and Santschi, 1997), whereas U(VI) is relatively more soluble and is therefore more capable of being transported in solution. The solubility of U(VI) accounts for its wide distribution in fresh water (Clark et al., 1997).

U(VI) is generally least mobile in reducing (anaerobic) environments and is most mobile in oxidizing (aerobic) environments. In addition, U(VI) is particularly mobile in high ionic strength solutions, where cations (such as Ca^{2+}, Mg^{2+}, or K^{2+}) will displace the uranyl ions away from soil exchange sites and force the U(VI) into solution. A USGS study of the groundwater uranium plume under the Solar Ponds found that, under the pH and redox conditions studied, there are no obvious solubility controls on the mobility of uranium. That suggests any transport retardation that may occur in that area is due to sorption processes.

Carbonate chemistry is important in U(VI) speciation. Carbonate complexes are soluble, and therefore promote U(VI) mobility. In the presence of carbonate and organic complexing agents, sorption has been shown to be substantially reduced, thereby increasing the mobility of the uranium (Hsi and Langmuir, 1985). The Upper Hydrostratigraphic Unit at RFETS is primarily a calcium-bicarbonate water, and therefore ample quantities of carbonate should generally be available to complex with uranium and promote its mobility in groundwater (EG&G, 1995a).

Sorption onto iron and manganese oxides represents an important process for extracting uranium out of solution. These can represent relatively irreversible sinks for uranium to be bound in soils.

4.2 RESULTS FROM RFETS STUDIES

4.2.1 Soil

Mapping of uranium isotopes in surface soil indicates the presence of isolated areas with elevated uranium activity. These areas are associated with locations historically used for disposal of wastes that contained uranium. The pattern of isolated areas with elevated activity is a function of the uranium's solubility in the RFETS environment. Instead of remaining on the ground surface as an insoluble species and being transported by soil erosion, the uranium, as U(VI), leaches downward in the soil.

4.2.2 Surface Water

In surface water, since RFCA monitoring was first implemented on October 1, 1996, water quality at all the POC monitoring locations has been compliant with the applicable RFCA standard for uranium. The standard is 10 pCi/L in Walnut Creek and 11 pCi/L in Woman Creek. For perspective, the final uranium MCL for drinking water is 30 μg /L, or nearly three times more uranium than the RFCA surface water standard allows for at the POCs.

A LANL study in the early 1990s (Efurd et al., 1993a), using TIMS, determined that the largest source of radioactivity in the terminal ponds was naturally-occurring uranium and its decay product, radium. However, all of the ponds, except Pond C-1, contained mixtures of naturally-occurring and depleted uranium, with concentrations ranging from 0.2 to 15.8 pCi/L. No depleted uranium was detected in Pond C-1. These findings are consistent with a separate LANL analysis in 2002, using ICP/MS, where depleted uranium was detected in North and South Walnut Creeks. However, it is noted that, historically, the 30-day moving average total uranium concentration has always been less than 4 pCi/L at all of the Walnut Creek surface water POC locations. That compares to the 10 pCi/L RFCA standard for Walnut Creek and is within the range observed for natural uranium in surface water in the RFETS area.

4.2.3 Groundwater

Results of ICP/MS studies of isotopic uranium ratios indicate that uranium in groundwater at RFETS, in the majority of wells sampled, is associated with natural material. The areas with relatively high concentrations of anthropogenic uranium in groundwater are near known source areas.

The Solar Ponds area is the main region of the Site that has a distinctive plume of uranium in groundwater. High Resolution ICP/MS analyses indicate, not surprisingly, the presence of both depleted and highly enriched uranium in the groundwater in the Solar Ponds area. A new groundwater collection and treatment system was constructed in 1999 to capture groundwater and provide treatment for uranium, as well as nitrate.

Some of the highest of uranium activity observed in groundwater at the Site is associated with natural uranium. Groundwater samples with uranium activities above RFCA Tier I Action Levels have been collected at the extreme north and south sides of the Buffer Zone, far removed from the Industrial Area. Those wells, notably in Rock Creek (on the RFETS northern boundary) and Smart Ditch (in the southeast corner of the Site), have natural isotope ratios and do not contain U-236, thereby indicating the uranium is natural. Those wells are good examples of locations where high uranium concentrations are caused by natural sources, not by anthropogenic material from the Site.

4.3 IMPLICATIONS FOR SITE CLOSURE

For surface water, control measures for uranium sources, to achieve surface water quality at the POCs, do not appear to be warranted based on historic surface water data. Surface water quality at RFETS has been compliant with the RFCA uranium standards since RFCA monitoring was implemented in 1996. Total uranium concentrations at the POCs, based on a 30-day moving average, have historically been less than one-half of the allowable standards: 10 pCi/L for Walnut Creek and 11 pCi/L for Woman Creek. For perspective, the RFCA standards allow only one half (approximately) of the uranium that is permissible in drinking water, as established by the EPA.

In groundwater, the area with the largest uranium plume is the site of the former Solar Evaporation Ponds. A collection trench and treatment system to capture groundwater and remove uranium was installed downgradient from the Solar Evaporation Ponds in 1999. Similarly, a collection trench and treatment system was constructed in 1999 downgradient from the East Trenches, another uranium source area.

The largest source of uranium in the terminal ponds is associated with natural uranium, and its decay product, radium. Depleted uranium has been detected in the RFETS ponds, except for Pond C-1. However, as noted above, the low level of depleted uranium in the ponds does not adversely impact compliance with the RFCA surface water quality standards. Total uranium concentrations at the surface water POCs are well below the RFCA standards, and are within the range of natural uranium concentrations for the area.

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