# DISTINGUISHING ANTHROPOGENIC URANIUM AT THE ROCKY FLATS ENVIRONMENTAL TECHNOLOGY SITE, GOLDEN, COLORADO

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## ABSTRACT

Background uranium (U) in ground water at the Rocky Flats Environmental Technology Site (Site) may be derived from several geologic sources. The Site has ground water with detectable concentrations of dissolved U in many background sampling wells. Both enriched and depleted U was processed at various times at the Site and some material was disposed of in the environment. A major challenge for the Site has been to determine whether the uranium detected in ground water was due to natural or man-made processes. Historic Alpha Spectroscopy results for the uranium isotopes were not accurate enough to determine natural isotopic ratios. Background concentrations appear to be related to lithologic units. With impending cleanup deadlines in 2006 there is no time to waste in disputing the background data. Most ground water becomes surface water before leaving the site and must meet an ambient standard of 10 pCi/l. High Resolution Inductively Coupled Plasma Mass Spectrometry (HR-ICP/MS) methods were used to investigate U isotopic ratios in ground water for the Site. The Solar Ponds area of the site has U concentrations up to 2550 pCi/l but the full impact of downgradient contamination is blurred by potential for natural concentrations in the stream alluvium. A sampling plan developed by the State regulator in cooperation with the Site has been used to distinguish background uranium from contamination at all areas of the site exceeding the surface water standard of 10 pCi/l U in ground water. The plan evaluates the sample results against the natural abundance ratio of U235 /U238 and a significant level of the U236/U238 ratio. The U236 isotope is an activation product present in anthropogenic U. The HR-ICP/MS data has allowed differentiation of U contamination at the Solar Ponds. These results have shown additional areas where buried depleted U has impacted ground water and one additional area with an enriched U signature. Selected samples with low levels of U236 were analyzed with Thermal Ionization Mass Spectrometry (TIMS) to help define the extent of impacted ground and surface waters. The U236/U238 ratio serves as a contaminant tracer even where the U235/U238 ratio appears normal.

## INTRODUCTION

The Rocky Flats Environmental Technology Site (RFETS or Site) is located 16 miles northwest of downtown Denver, Colorado. Local rocks and mineral deposits are enriched in uranium causing difficulty in the determination of a background value for uranium in ground water. Alluvial deposits from the Ralston Buttes Uranium District make up the main aquifer on site. Uranium bearing claystone also underlies the Site. Rocky Flats was a nuclear weapons manufacturing facility from 1952 until 1989, producing metal components from plutonium, uranium, beryllium, and stainless steel. Some areas of the Site have uranium-contaminated ground water, however traditional statistical methods of determining background concentration were complicated by large variability in those concentrations. Isotopic ratios that could

distinguish contamination from background could not be accurately calculated with existing alpha spectroscopy data. The Colorado Department of Public Health and Environment (CDPHE) found utility in a pilot study using mass ratios analyzed by High Resolution Inductively Coupled Plasma Mass transuranic radionuclides, research and development programs in metallurgy, machining, chemistry and physics.

Both enriched and depleted uranium were used on the Site. Both liquid and solid uranium waste streams were generated on the Site. Liquid process wastes were piped to treatment buildings and to the Solar Evaporation Ponds. Most solid U waste was either re-used or drummed and disposed of properly. However, in the 1950s - early 1960s drums containing depleted uranium were buried in a trench (T1) until removed in 1997. Uranium has been a component of other spills or found in trenches at the Site (Mound, T3-T4), which have also been remediated.

In December of 2000 the U.S. Congress passed a bill designating the site as a Wild Life Refuge. All buildings onsite will be removed to below three feet of ground surface. Surface contamination will be removed to action levels calculated safe for a Wildlife Refuge Worker. Removal of subsurface contamination depends on the potential for that contamination to be brought to the Spectrometry (HR-ICP/MS) and proposed using the method site-wide to resolve the ground water issues with uranium. The results of that study are presented here.

## SITE HISTORY AND REMEDIATION STATUS

Rocky Flats Plant produced nuclear weapons components from 1953 – 1989. The industrial processes involved extensive metallurgical extraction, precipitation and forming of parts involving uranium and plutonium. Other plant activities included chemical recovery and purification of recyclable surface. Neither surface nor subsurface contamination will be allowed to impact surface water above standards. Remediation work is on track or ahead of schedule and the site anticipates achieving closure by the end of 2006.

## GEOLOGIC SETTING

The Site is located on the western edge of the Denver Basin. The Idaho Springs-Ralston sheer zone a few miles west of the Site truncates and deforms the sedimentary formations of the basin. Sedimentary Cretaceous formations, Pierre Shale to Arapahoe, underlie the site on the slightly eastward dipping rim of the Denver Basin, Fig. 1.

The Ralston Buttes District, an area of about 7 square miles in Precambrian metamorphic country rocks is located west of the sheer zone. Precambrian structures were reactivated and the sheer zone faulting occurred during the Laramide. Several mafic monzonite intrusions were emplaced along the fault zone about 62 m.y.b.p. A set of northwest trending breccia-reef faults served as a conduit for Uranium migration. Mineralization is associated with hydrothermal solutions from these intrusions (1).

The Ralston Buttes District is dissected by Coal Creek canyon. The 10 square mile RFETS is situated on the Rocky Flats Alluvium, an alluvial fan deposit of early Quaternary age, emanating from the mouth of Coal Creek Canyon. The Rocky Flats Alluvium and other Quaternary

alluviums overlie the Cretaceous bedrock. This pediment of alluvium-capped sediments is dissected by several drainages originating within the site boundaries. Recent colluvial deposits composed of reworked Cretaceous sediments and Quaternary alluviums are found on the slopes of these drainages and valley fill alluvial deposits surround the current streams.

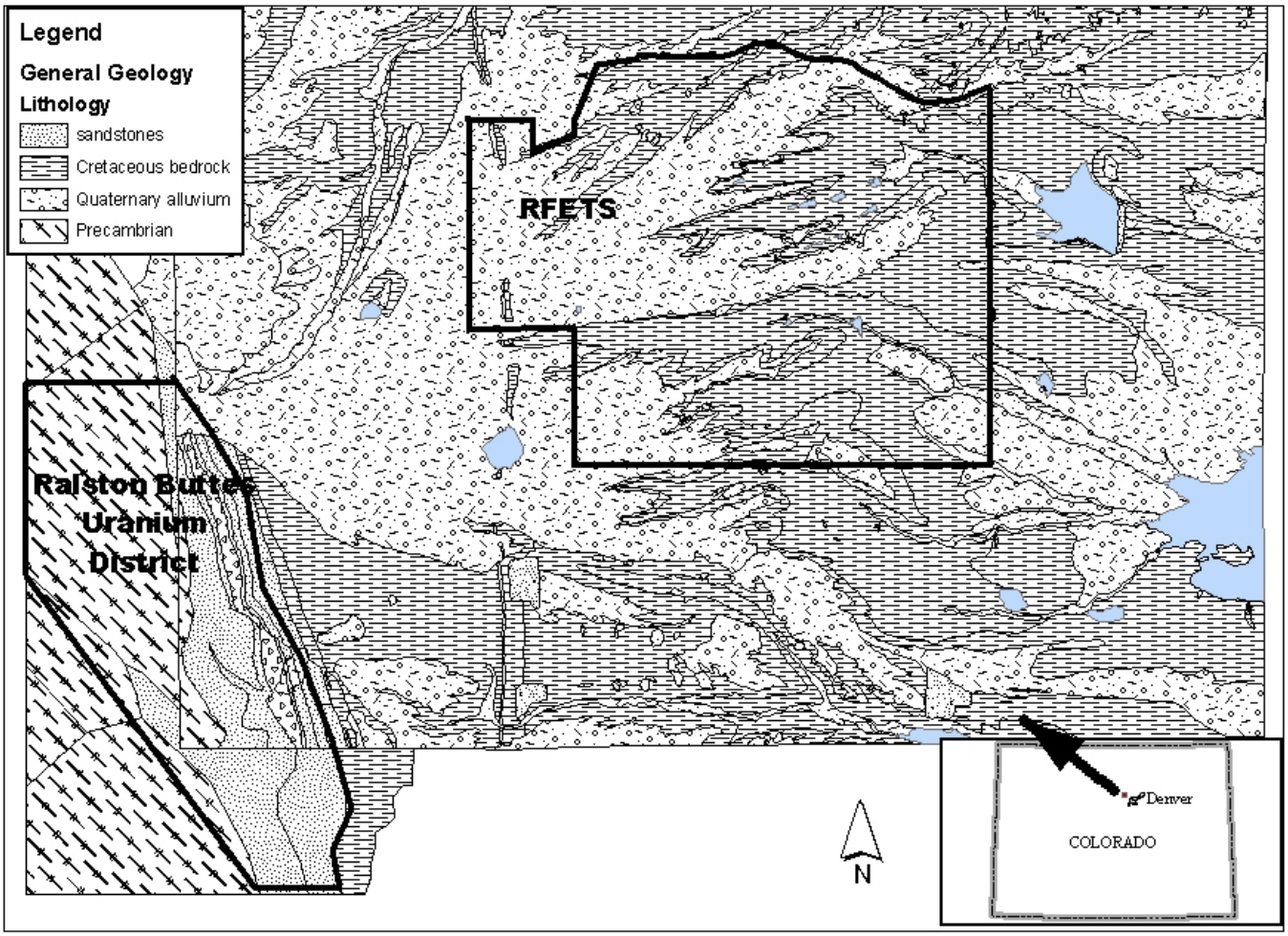


Fig. 1 RFETS and Ralston Buttes area geology

## SITE HYDROGEOLOGY

Coal Creek now exits the mouth of the canyon below the alluvial deposits and flows northeast outside the RFETS boundaries some ditches that divert water from Coal Creek cross the site. The hydrogeology of the site is divided into upper and lower flow systems called hydrostratigrahpic units (HSU). The upper HSU is comprised of 5 - 100 feet of Rocky Flats Alluvium, weathered bedrock, colluvial, and valley fill deposits. The lower HSU is 600-1000 feet of primarily claystone in the Laramie and Arapahoe formations. The Laramie-Fox Hills Aquifer outcrops in nearly vertical section on the western side of the site but assumes the regional dip of 2-3 degrees eastward within a short distance either by bending or fracturing. No site contaminants have migrated through the thick claystones and this aquifer is considered unimpacted by site activity. The ground water flow of concern for transport of contaminants is in the upper HSU. Ground

water is not a viable resource on Site and very few areas have saturated thickness or lateral extent to supply a typical domestic well. Because most ground water becomes surface water before leaving the Site, ground water is classified for the protection of surface water.

Walnut Creek and Woman Creek originate onsite. At one time these streams contributed water to public drinking water supplies but are currently diverted. Until the EPA established a Maximum Contaminant Limit (MCL) of 30 ug/l (about 27 pCi/l) in 2000, there was no promulgated drinking water standard for uranium that could be applied to the streams. Establishing a surface water background concentration with upgradient sampling was not possible because both streams originate on site. Therefore, in 1990 the State established ambient surface water concentrations for these streams at the eastern Site boundary of 10 pCi/l total uranium for Woman Creek, 11 pCi/l for Walnut Creek.

**Uranium in Groundwater**

Historically, the approach to establishing a background value for uranium in ground water began with the collection of water from upgradient wells beyond the known extent of contamination. Those samples were analyzed for U isotopes 234, 235, and 238 by alpha spectrometry. However, when examined by formation, significant differences appear in data from the Background Geochemical Characterization Report (2):

Table I Total uranium in ground water by formation

| Geologic Unit | Sample Size | Mean pCi/l | 99/99 UTL pCi/l |
|---|---|---|---|
| Rocky Flats Alluvium | 78 | 0.4 | 1.67 |
| Colluvium | 30 | 59.38 | 412.0 |
| Valley Fill Alluvium | 60 | 3.97 | 20.19 |
| Weathered Bedrock | 39 | 12.33 | 93.38 |

The lack of dissolved uranium in the Rocky Flats Alluvium, the formation derived from the Ralston Buttes mineralized zone, is striking. Higher values are found in the weathered Cretaceous claystones of the Arapahoe and Laramie formations. Higher still are the results from the Colluvium, a lithology consisting of reworked bedrock and alluvium. Background U at RFETS apparently is more related to fine grained Cretaceous sediments than the Ralston Buttes District mineralization. The values for the Valley Fill Alluvium are of most concern, being closely associated with surface water and close to the ambient standard of 10 pCi/L total U established by the State. A method to distinguish anthropomorphic U was necessary because the areas believed contaminated were in the more permeable alluvial formations.

**Alpha Spectroscopy Background Data**

An approach designed to utilize Alpha Spectrometry for the purpose of calculating isotopic ratios of uranium produced results that were inconclusive. The objective was to determine whether the differences in uranium isotope abundances can reliably indicate the presence of natural, enriched, or depleted Uranium sources. Isotopic ratios can be used for activity or mass. Alpha Spectroscopy is an activity measurement and therefore activity ratios should be used with that

data, especially in a situation where secular equilibrium may be in question because the conversion to mass assumes equilibrium. The U234/U238 activity ratio should be about one, however physical and chemical differentiation processes may result in a natural radiochemical disequilibrium in this ratio. The Alpha Spectrometry data was found to lack sufficient accuracy and precision to calculate meaningful isotopic ratios.

## SOLAR PONDS RESOLUTION

The Solar Evaporation ponds are the largest area of U contamination on site. While there was not much question the uranium underneath the Solar Ponds was contamination, the ground water in the drainage below appeared to be related to the plume as well, even though a ground water intercept system had been constructed between the two areas in 1971 and upgraded to an Interceptor Trench System (ITS) in the 1980s. There are several other areas on site where uranium solutions may have leaked from process waste lines or solid uranium was buried. Ground water U concentrations frequently exceed the State standard. The U234/U238 activity ratios calculated with alpha spec data were inconsistent. Upgradient areas thought to be background had ratios suggesting contamination and some areas of known contamination areas had ratios of about one. In 1995 the Rocky Flats Cleanup Agreement (RFCA) was negotiated between Kaiser-Hill, DOE, EPA and CDPHE. The Agreement allowed the parties to develop site-specific approaches to problem solving with the goal of accelerated site closure. Impending decisions on the Solar Ponds Plume were the driver for the site to find a more accurate analysis method to settle the background issue.

The U235/U238 ratio is essentially constant in nature both in rock and water (4). However, low activity levels of U235 preclude accurate measurement of this isotope with alpha spectroscopy. With mass measurements it is U234 that is difficult to measure accurately. This leaves the U235/U238 mass ratio as the most reliable for determination of disequilibrium in the isotopes of uranium. The Site sought and found a mass method for isotopic analysis to resolve these data quality issues through Los Alamos National Laboratory (LANL).

In late 1997, the site conducted an evaluation of a limited number of wells in the Solar Pond and North Walnut Creek areas and several wells considered to be background. Ground water samples were analyzed by the high-resolution inductively coupled plasma/ mass spectroscopy (HR-ICP/MS) method developed at LANL. The method had expected errors between 1-5% of the mass of each isotope. The evaluation also analyzed for U236, an artificial activation isotope found in industrially processed uranium but not in nature.

All uranium in North Walnut Creek appeared to be natural, something that might be too good to be true. The CDPHE review of the method and results allowed the selection of a remedial alternative to proceed although questions remained about the actual detectable level for U236. An impermeable barrier system was installed in 1999 to improve the ground water collection ability of the ITS at the Solar Ponds. The barrier system feeds into a sawdust and iron filings treatment cell, which precipitates uranium onto the treatment media. The nitrate plume associated with the Solar Ponds is converted to nitrogen gas in this reducing environment. Replacement of the previous evaporation treatment of the uranium and nitrate contaminated water produced cost savings passed on to other site cleanup projects (5).

## MASS ISOTOPIC STUDY

The CDPHE proposed a sampling and analysis plan using the method for the many other areas on site where uranium in ground water is above the surface water standard and needs resolution for site closure. The Site and CDPHE worked together to choose the wells of interest and develop the sampling and analysis plan. The State allowed the Site to fund the analysis at LANL with part of the oversight grant. The Site funded sample collection by Site contractors. As many wells as possible were chosen from the current sampling program to reduce costs.

### Hypothesis

To formulate a hypothesis test for this project, the initial work done by RFETS and LANL was carefully examined. The actual percent isotopic abundances of the U234, U235, and U238 found in the background wells at RFETS were observed to be well within the literature values for these naturally occurring U isotopes (3). However, the U234 abundances from the background well population were indistinguishable from the U234 abundances in the potentially impacted wells. This observation is believed to be directly related to the fact that U234 is a radioisotopic daughter product of U238 in secular equilibrium with its parent, regardless of the source of U238 (anthropogenic, or natural). This decreases the utility of using the U234/U238 ratio for the purpose of identifying wells impacted by RFETS activities. This observation supports the use of the U235/U238 ratio as an indicator of anthropogenic U sources for contamination.

U236 is an artificial isotope created through fission reaction with U235 in a nuclear reactor. Because RFETS often used reactor uranium as feed stock it would contain U236 as an impurity. U236 would probably occur in both enriched and depleted uranium used at RFETS. U-236 has a half-life of $2.34 \times 10^7$yr and decays by alpha particle emission. Data for U236 was accumulated by LANL using the HR-ICP/MS. The detection of this man-made uranium isotope was not available using Alpha Spectrometry, and adds an additional dimension to this work. There are three possible radioisotopic parents for U236 (Pa236, Np236, and Pu240), all of which are also man-made radionuclides. The background data set contained a single positive detection of U236 (well B305389) at about 2 picograms/liter, and one North Walnut Creek well (B210489) had a positive detection of U236 at about 22 picograms/liter. The U235/U238 ratio shows these two wells to be, otherwise, in the background population. Understanding these two U236 detections was critical to using detections of U236 for this project. The HR-ICP/MS approach employed by LANL measures the 236 mass with a resolution of 1 amu. Because of this resolution, an isobaric interference is not believed to be operating on these measurements (isobar= element, or molecule with the same atomic weight). Instead, these U236 detections are believed to be manifestations of uncertainty about the U236 detection at these extremely low concentrations.

The original hypothesis was that in contaminated wells the U235/U238 ratio would be outside the mean plus 3 standard deviations of the natural ratio and that the U236/U238 ratio would confirm the contamination. A significant level for the U236/U238 ratio would be established by analysis of the data.

**Sampling Methods**

First phase of the study collected 4 samples each from 47 wells plus repeat samples of the Solar Ponds study wells. Samples were collected down gradient of suspected sources, in drainages, and where U>10 pCi/l. A second phase of sampling collected 47 additional single samples from wells and surface water locations. Samples were field filtered with a 0.45-micron filter and collected in 8-ounce polyethylene or fluorocarbon containers. Samples were preserved with dilute HNO3 to pH < 2. Field quality control samples were collected at a frequency consistent with the Site ground water program. Samples were sealed after collection. A chain of custody form was maintained for each batch of samples. Samples passed a radiation screening on site before being transported to the laboratory.

## RESULTS AND DISCUSSION

Data from 10 wells known to be outside of any site activities were used to examine the natural variability of the site background. For the U235/U238 ratio we have chosen 3 standard deviations around the background mean were chosen to include 99% of the data. This analysis is represented as the horizontal origin of the graph and plus or minus 3 standard deviation lines bracketing the origin in Fig. 2. The significant level of the U236/U238 ratio, established by data evaluation, is represented by the vertical line at 2E-6. Data points in the upper right quadrant represent a signature enriched in U235, those in the lower right quadrant represent uranium depleted of U235.

To date 95 locations have been analyzed by this method, 30 locations show evidence of impact. The U235/U238 ratio is outside 3 standard deviations of background in only 7 locations, 4 showing enrichment of U235, 3 showing depletion relative to U235, all have definitive ratios of U236/U238 (greater than 2E-5). These locations are all wells, relatively near the suspected sources. Three of the 4 enriched locations are in the Solar Ponds plume. The other is near the now demolished criticality laboratory and a contaminant source was not found during the demolition and environmental sampling. The depleted wells represent one Solar Ponds location and confirmed contaminant sources at two buried waste locations.

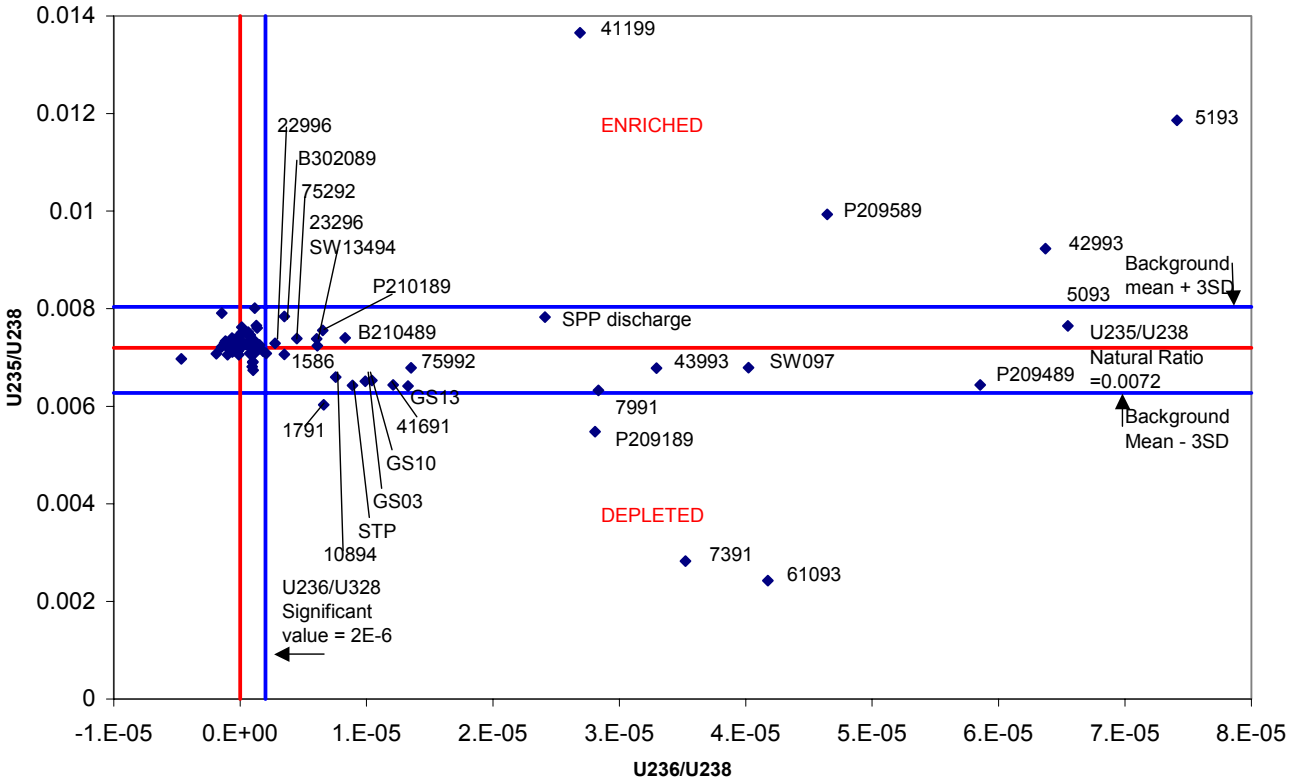


Fig. 2 Three isotope diagram of HR-ICP/MS results

Six locations have U235/U238 ratios within the M3SD but U236/U238 ratios in the same range as the source area data. Four of these locations are in the Solar Ponds. The other locations are the Solar Ponds treatment system discharge gallery and SW097 a seep at the Present Landfill, where U was improperly disposed. This suggests that the U236/U238 ratio is a better predictor of contamination than the U235/U238 ratio, and that the hypothesis should be that contamination is indicated when either ratio is outside a normal range.

A third grouping of data raised questions about the lower limit of significance the U236/U238 ratio. In these results the U235/U238 ratio is within the MS3D range of background and the U236/U238 ratio is between 2E-6 and 2E-5. The physical locations include wells and surface water stations in the Solar Ponds and downgradient in both North and South Walnut Creek to the site eastern boundary. The concentration at the eastern boundary locations does not violate the surface water standard, averaging about 2 pCi/l for ground water and 1.6 pCi/l for surface water. Five non Solar Ponds locations are also in this group, the sewage treatment plant effluent (also below surface water standard), a well downgradient of Trench 1(a now remediated disposal site), and a well and surface water location associated with Building 881 (a former lab). The last location, well B302089, is located in a drainage south of Woman Creek and was considered a background location. In order to resolve these levels a method with better accuracy and precision was suggested by LANL.

**TIMS Confirmation Samples**

The consistent low detections of U236 combined with CDPHE's initial questions about the practical quantitation limit required further evaluation. In order to determine if the U236/U238 ratio was significant HR-ICP/MS results were checked by thermal ionization mass spectrometry. The resulting data in Fig. 3 shows the U236/U238 ratio is significant down to about 2E-6. However one in 3 samples analyzed by HR-ICP/MS from the above mentioned background well has a U236/U238 ratio of 3.49E-6 while the other results for this well are negative or in the 1E-7 range. Rather than repeated analysis with HR-ICP/MS, results below 1E-5 should be confirmed with TIMS if there is a compliance issue.

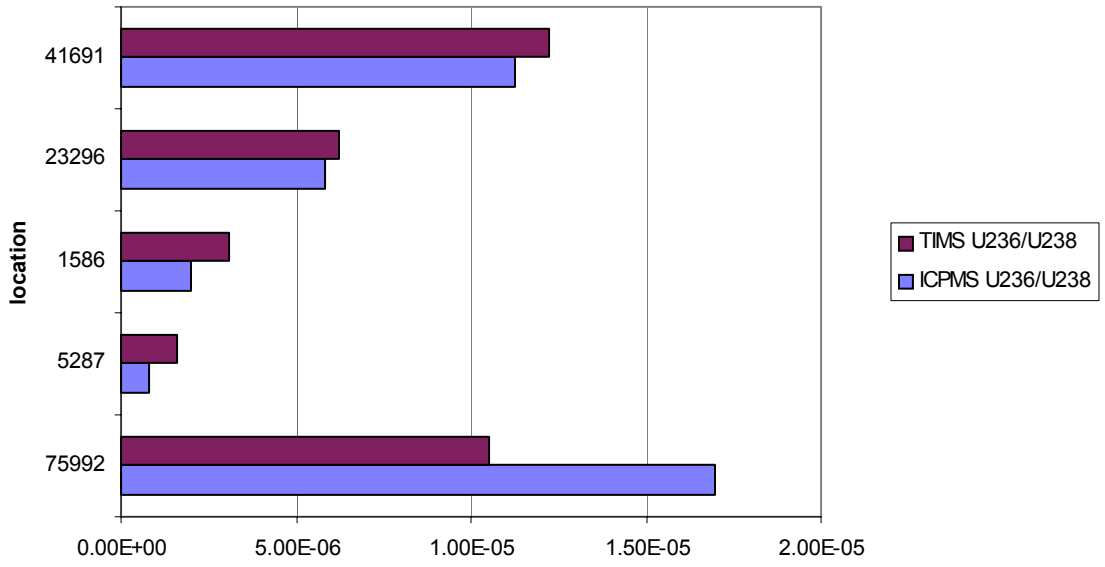


Fig. 3 Comparison of U236/U238 in TIMS and HR-ICP/MS duplicate samples

**Data Consistency**

Statistics for wells with multiple samples illustrate the consistent results this method produced. For wells within M2SD of the U235/U238 background ratio, one sample is probably sufficient. If the result is closer to either 3rd standard deviation line a confirmatory sample is probably warranted. If the 236/238 ratio is below 1E-5 confirming the U236 with TIMS analysis is helpful. Contaminated ground water is mixing with natural U along flow pathways away from sources, which may account for some of the U235/U238 within the natural range in wells that have low ratios of U236/U238.

Table II Mean and standard deviation of ratio data

| Sampling Location | U236/U238 AVG | 236/238 SD | U235/U238 AVG | U235/U238 SD |
|---|---|---|---|---|
| 1586 | 2.52E-05 | 2.41E-05 | 7.33E-03 | 1.84E-03 |
| 3586 | 2.38E-05 | 1.78E-05 | 6.70E-03 | 3.53E-03 |
| 4087 | 2.19E-05 | 3.56E-06 | 4.02E-03 | 2.47E-03 |
| 5287 | 1.27E-05 | 1.30E-05 | 3.24E-03 | 1.10E-03 |
| 5387 | 1.28E-05 | 7.49E-06 | 2.71E-03 | 1.25E-03 |
| 10294 | 1.01E-05 | 3.75E-06 | 1.98E-03 | 1.03E-03 |
| 10394 | 8.55E-06 | 3.86E-06 | 1.74E-03 | 7.61E-04 |
| 10594 | 8.51E-06 | 3.79E-06 | 1.51E-03 | 6.75E-04 |
| 10694 | 5.69E-06 | 2.59E-06 | 1.14E-03 | 4.66E-04 |
| 22996 | 5.15E-06 | 2.58E-06 | 9.49E-04 | 4.69E-04 |
| 23296 | 3.96E-06 | 1.43E-06 | 7.41E-04 | 3.00E-04 |
| 36391 | 2.66E-06 | 1.27E-06 | 5.03E-04 | 2.22E-04 |
| 37791 | 2.33E-06 | 1.25E-06 | 4.41E-04 | 2.32E-04 |
| 37991 | 1.88E-06 | 7.25E-07 | 3.50E-04 | 1.44E-04 |
| 41591 | 1.28E-06 | 5.76E-07 | 2.42E-04 | 1.03E-04 |
| 41691 | 1.14E-06 | 5.16E-07 | 2.14E-04 | 9.58E-05 |
| 43993 | 8.80E-07 | 3.91E-07 | 1.64E-04 | 7.51E-05 |
| 52894 | 5.95E-07 | 2.54E-07 | 1.12E-04 | 4.64E-05 |
| 53194 | 4.13E-07 | 1.72E-07 | 7.77E-05 | 3.26E-05 |
| 59393 | 3.59E-07 | 1.87E-07 | 6.71E-05 | 3.51E-05 |
| 59793 | 2.77E-07 | 1.06E-07 | 5.18E-05 | 1.98E-05 |
| 61093 | 2.32E-07 | 1.10E-07 | 4.35E-05 | 2.04E-05 |
| 75292 | 1.81E-07 | 8.67E-08 | 3.39E-05 | 1.62E-05 |
| 75992 | 1.43E-07 | 6.14E-08 | 2.68E-05 | 1.15E-05 |
| 193 | 1.18E-07 | 5.42E-08 | 2.21E-05 | 1.01E-05 |
| 491 | 9.42E-08 | 4.34E-08 | 1.76E-05 | 8.11E-06 |
| 897 | 6.88E-08 | 3.59E-08 | 1.29E-05 | 6.72E-06 |
| 997 | 6.06E-08 | 2.65E-08 | 1.13E-05 | 4.95E-06 |
| 386 | 4.79E-08 | 2.00E-08 | 8.97E-06 | 3.74E-06 |
| 3991 | 3.15E-08 | 1.46E-08 | 5.89E-06 | 2.74E-06 |
| 487 | 2.85E-08 | 1.47E-08 | 5.33E-06 | 2.76E-06 |
| P114389 | 2.01E-08 | 4.07E-11 | 3.76E-06 | 7.61E-09 |
| P207689 | 1.34E-08 | 1.16E-08 | 2.51E-06 | 2.16E-06 |
| P209189 | 1.13E-08 | 8.33E-09 | 2.11E-06 | 1.56E-06 |
| P209489 | 1.11E-08 | 2.10E-09 | 2.08E-06 | 3.92E-07 |
| P209589 | 8.21E-09 | 4.30E-09 | 1.54E-06 | 8.04E-07 |
| P209889 | 6.44E-09 | 4.03E-09 | 1.20E-06 | 7.54E-07 |

**Migration of Contamination**

Figure 4 shows the location, concentration and contaminant signature of the data. Wells in the Solar Ponds area show both enriched or depleted signature in the U235/U238 ratio. This would be consistent with the ponds history of leaks and fixes of the pond liners. Migration to North Walnut Creek could have occurred before installation of the interceptor trench system or as underflow of the system. The current ground water barrier collection system truncates the interceptor system. The Discharge Gallery for the new treatment system includes piping from the old trench system and is thought to be picking up contamination after treatment. Concentrations at this location range from 5 to 113 pCi/l. The surface water performance monitoring station, GS13 averages 9 pCi/l (6). Determination of the rate of ground water transport is not possible in this area because of the repeated disturbances and piping. Two wells father down the North Walnut drainage do not show a contaminant signature. This plume is possibly attenuating in the reducing environment in the alluvium of this drainage in the vicinity of Pond A1.

A lobe of the Solar Ponds nitrate and uranium plume was believed to be migrating to the east toward South Walnut Creek however, none of the wells evaluated east of ponds shows a contaminant signature. The first well in the South Walnut Creek drainage has about 7 pCi/l U and no contaminant signature. The next well downstream, 75992, has about 16 pCi/l and a U236/U238 ratio of 1E-5, the nearby surface water station, GS10 has a similar contaminant signature. From that point, all wells downstream in South Walnut Creek and Walnut Creek show low levels of U contamination. The maximum is 26 pCi/l in well 75292 below pond B-4, dropping down to 2 pCi/l at the site boundary. Uranium sources in this area would be primarily Trench 1 where drums containing depleted U were excavated in 1999. Only one well is located near this excavation, 1791 shows 5pCi/l with a depleted U235/U238 signature and 6.6E-6 U236/U238 confirming impact. The Sewage Treatment Plant discharges to South Walnut Creek in Pond B3 several hundred feet downstream of 75992. The one sample collected from the STP also shows a contaminant signature, 7pCi/l with 9E-6 U236/U238. Influent samples for the STP typically range from 2 – 5 pCi/l since 1998. Further investigation is needed with these sources to determine if this contamination is a result of past discharges or if there is an ongoing contribution from ground water.

An enriched signature is found in several wells near the now demolished criticality laboratory. Full assessment of a transport pathway is not possible because a source was not found during demolition and environmental sampling and building drains may have disrupted ground water flow in the area.

Depleted signature wells occur near 4 waste burial sites, the Mound, East Trenches, Ryan's Pit, and the Original Landfill. The Mound Source was remediated in 1997, the concentration is below the standard and a downgradient well shows no impact. One well in the East Trenches area shows 20 pCi/l U, surrounding wells have not been assessed with this method. Ryan's Pit area was remediated in 1996, soil concentrations were not above action levels but the downgradient well contains 128 pCi/l U. No other well exists directly downgradient to track

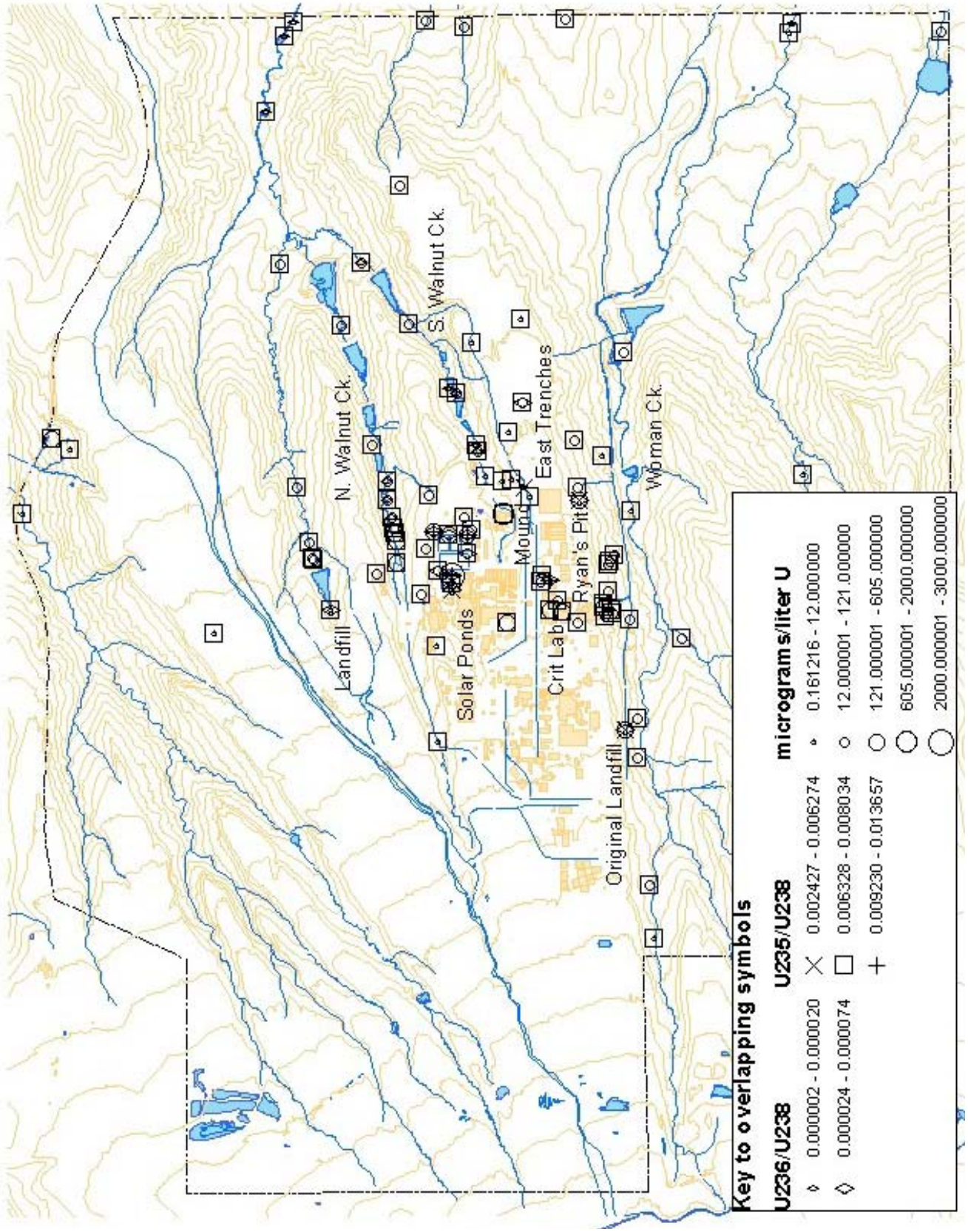


Fig. 4 Location, concentration, and contaminant signature of HR-ICP/MS data

migration. The Original Landfill well is associated with an identified buried source, which will be remediated before closure. This well contains 250 pCi/l U and a nearby downgradient well has not yet been analyzed with this method.

Contaminant migration is difficult to trace along flow paths for the various reasons mentioned above: interception by drains or surface water, lack of appropriately spaced downgradient wells, lack of source term information. However transport by ground or surface waters is evident from the U236/U238 ratios that persist even at low concentrations. Some generalities may be drawn about the impact of various sources on ground water. Sources of solid depleted uranium can leach into ground water at a rate consistent with recharge and depth to ground water. Sources of enriched uranium were more likely liquid and move to ground water without leaving high levels in soil. A source of liquid or dissolved depleted uranium may exist in the South Walnut Creek drainage, possibly linked to the sewage treatment plant. Presently no transport modeling is planned to evaluate loading of uranium to surface water. No uncontrolled source is thought to pose a threat to surface water standards of water leaving the site. On-site surface waters may be impacted in localized areas at levels above the standard but rarely above the MCL.

## CONCLUSIONS

The High Resolution ICP/MS method has proven ability to distinguish uranium contamination from a highly variable natural background. The enriched or depleted signature can be established by the U235/U238 ratio and the U236/U238 ratio serves as confirmation of contamination even when the U235/U238 ratio is within the natural variability of the background. Use of this method has successfully identified areas of uranium contamination to ground and surface waters at RFETS.

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