

Evaluation of Uranium Enrichment/Depletion and Recycled Uranium Residuals in Soils and Groundwater at the Harshaw FUSRAP Site - 9336

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ABSTRACT

This paper reports on investigations of uranium isotope ratios in environmental media at the Former Harshaw Chemical Site measured using three analysis techniques - alpha spectroscopy, gamma spectroscopy and inductively-coupled plasma - mass spectrometry (ICP-MS). Results of these analyses revealed that conclusive determinations of uranium enrichment or depletion in site media could only be determined via ICP-MS at the slight deviations from natural ratios that were present.

A second, related, topic addressed in this paper is the identification of residuals of recycled uranium (RU) and trace residuals of associated radiological contaminants in site soils and groundwater. This investigation relied on the use of a variety of analytical techniques to analyze for plutonium, neptunium, technetium, americium isotopes and uranium-236 in site media. The presence of several of these contaminants together in some soil samples established the presence of RU and traces of its contaminants in site soils. Detections of the latter contaminants in groundwater were few and sporadic, with only single contaminants detected in isolated samples. Thus, impacts on groundwater were inconclusive.

INTRODUCTION

The Former Harshaw Chemical Site in Cleveland, Ohio, is being investigated under the Formerly Utilized Sites Remedial Action Program (FUSRAP) and comprises buildings, soil, sediments, and groundwater contaminated with uranium and other radiological and non-radiological contaminants from the production of uranium products under Manhattan Engineering District (MED) and Atomic Energy Commission (AEC) programs in the 1940s and 1950s. Records from the operational period and afterwards indicate that the main feeds processed under these programs involved both natural uranium and uranium slightly depleted in the isotope U-235. Natural uranium feeds included high purity UO_2 and uranium ore concentrates from various suppliers of primary feed materials. Depleted uranium feed was associated with RU oxide received from the Department of Energy (DOE) Hanford complex in southeast Washington state [1]. Roughly 5,000 metric tons of uranium in all was processed, of which around 1,900 tons, or about 40%, was RU.

Other records indicate that research quantities of slightly enriched uranium were also processed at the plant. Quantities in the 10s-of-pounds range were processed during the period of MED/AEC operations, but it is not known if records are complete. Several 1946 records reference shipments of “slightly enriched” UF_6 and UO_3 to the Harshaw Chemical Company (HCC) for disposal/processing purposes.

Another record from 1944 references HCC research to evaluate the recovery of UF_6 from spent carbon traps potentially originating from uranium enrichment processes. Records do not indicate that HCC processed large quantities of slightly enriched material, but they do indicate the HCC uranium refining process may have been utilized for the disposal or recycling of slightly enriched material. Because natural, depleted, and enriched uranium were processed, the actual uranium isotopic composition in environmental media is of interest for a number of purposes, including risk assessment, cleanup, and waste management.

In addition, RU from Hanford is known to have contained a number of radiological contaminants of potential concern. A 2003 DOE report [2] identified plutonium, neptunium, and technetium in RU shipped from Hanford to Harshaw for purification prior to final delivery to the Oak Ridge Gaseous Diffusion Plant. The RU would have also contained small quantities of the isotope uranium-236 from nuclear processes carried out at Hanford.

Determining the presence or absence of enriched uranium at the site is important for the future management of potential remediation wastes and for the associated analyses in the upcoming feasibility study. MED/AEC work was carried out mainly in a fenced area of the plant known as "Plant C," in Building G-1 and surrounding areas. A map of the site is shown in Figure 1.

ENRICHED URANIUM INVESTIGATIONS

The possible presence of enriched uranium contamination at the Former Harshaw Chemical Site was investigated to address historical reports that small quantities of slightly enriched material were processed during early years of operation (1942-1945). Enrichment determinations based on both alpha and gamma spectroscopy made in initial phases of investigation indicated the possible presence of slightly enriched uranium in site soils. However, the results were not definitive because uncertainties in the U-235:U-238 ratios measured by either method may have exceeded the slight deviations from natural ratios expected from the known feeds. Others have reported difficulties in making determinations of slight enrichment or depletion using alpha spectroscopy [3]. The possible presence of RU further complicates the latter determination because RU could introduce the isotope U-236, which interferes with U-235 abundance in alpha spectroscopy measurements, artificially increasing apparent enrichment.

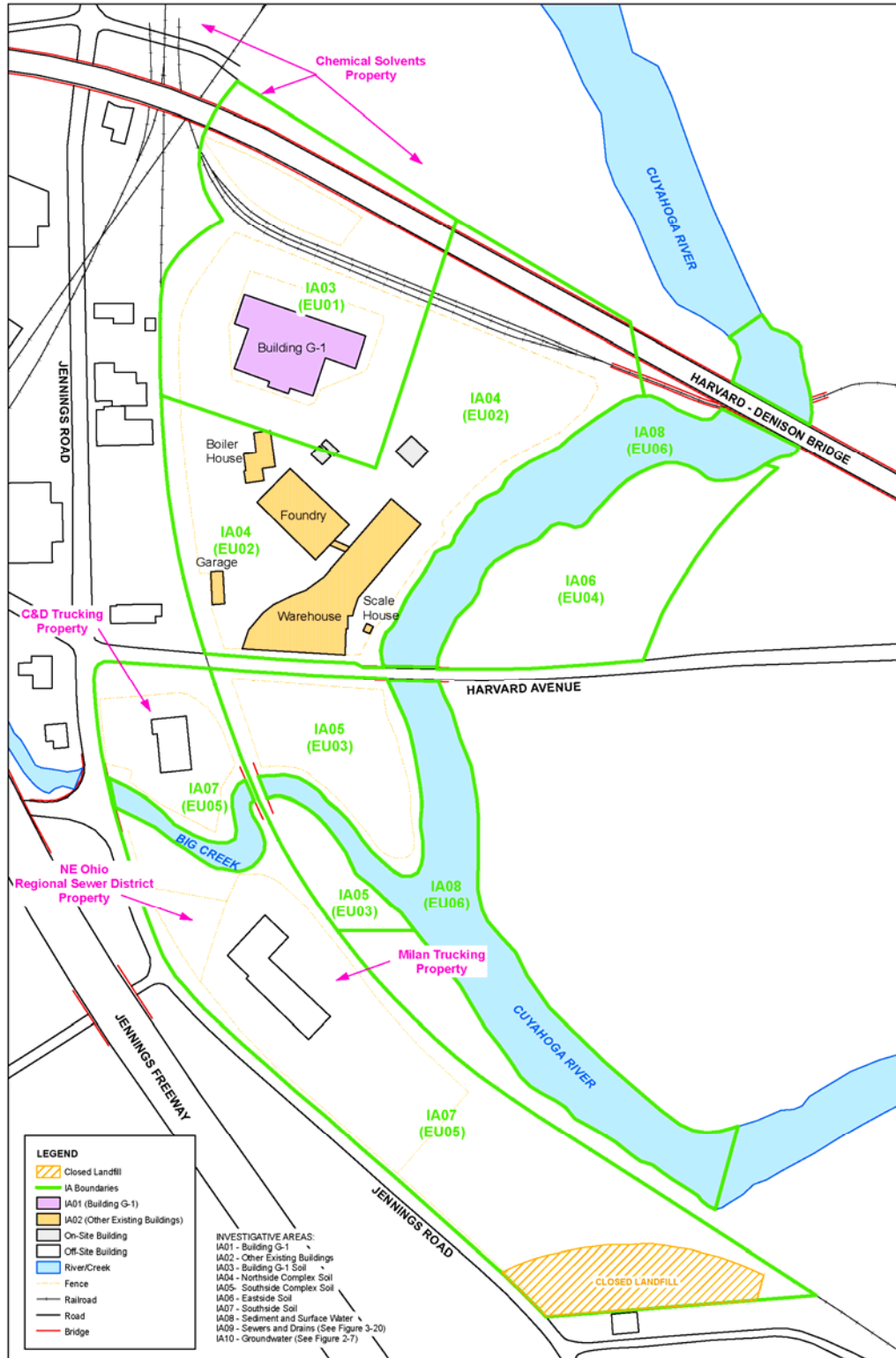


Fig. 1. Map of the Former Harshaw Chemical Company FUSRAP site.

Thus, determinations of whether uranium residuals are slightly depleted or slightly enriched are difficult, especially when RU residuals are present and isotope abundances have been affected by uranium processing. In addition, gamma spectroscopy is insensitive to U-234, and thus does not produce results for U-234:U-238 ratios, while both alpha and gamma spectroscopy have generally lack the precision needed to make measurements of slight deviations from natural ratios.

Analysis of uranium isotope ratios using ICP-MS was identified as a means to address the shortcomings of the other two methods. ICP-MS is capable of making very precise measurements of isotopic ratios and is much less susceptible to interference from other sample constituents than is either alpha spectroscopy or gamma spectroscopy. In addition, ICP-MS can make measurements of all uranium isotopes in a single analysis. U-233 and U-236 abundances can also be determined and used in interpreting deviations from normal isotope ratios and for identifying the presence of RU.

The U-235:U-238 activity ratio in natural uranium is approximately 0.044. Ratios significantly greater than 0.044 are indicative of enriched uranium, while ratios significantly less than 0.044 represent depleted uranium. On a mass basis the ratio in natural uranium is approximately 0.0071:1, or 0.71% uranium-235.

The U-234:U-238 activity ratio in natural uranium is equal to 1, i.e., the two isotopes have equivalent activities under secular equilibrium. On a mass basis, natural uranium has a U-234:U-238 ratio of approximately 5.4E-5:1.

Uranium isotope measurements for the three methods were made in two off-site laboratories. Alpha and gamma spectroscopy measurements were made at General Engineering Laboratories (GEL) in Charleston, South Carolina, and ICP-MS measurements were made at Test America (TA) in St. Louis, Missouri.

A summary of the gamma- and alpha-spectroscopy results of uranium isotope measurements in site and background soils at the Former Harshaw Chemical Site is given in Table I. Results from ICP-MS analysis are discussed separately below.

Table I. Uranium Isotope Activity Ratios in Soils by Gamma and Alpha Spectroscopy

Method and Sample Location	U-235:U-238 ^a	U-234:U-238 ^a
Gamma Spec – Background Soils	0.071 ± 0.013 (N=5)	ND ^b
Gamma Spec – On-Site Soils	0.068 ± 0.005 (N=137)	ND
Alpha Spec – Background Soils	0.093 ± 0.036 (N=2)	0.947 ± 0.033 (N=12)
Alpha Spec – On-Site Soils	0.088 ± 0.011 (N=134)	0.997 ± 0.020 (N=183)
Natural Uranium	0.044	1.0

^aMean activity ratio ± 95% confidence interval computed using N results.

^bND reflects that Gamma spectroscopy is insensitive to U-234.

A total of 137 on-site and 5 off-site background soil samples were included in the gamma spectroscopy results. U-235:U-238 ratios for each sample data pair were calculated and compared to the expected ratio of 0.044:1 using sample-specific uncertainty calculated from the counting statistics of the analysis. Twenty six (18%) individual sample ratios, including uncertainty, exceeded 0.044 in on-site soils, while none of the background samples did. The range of U235:U238 ratios in on-site soils (0.034 to 0.242) and background soils (0.060 to 0.97) reveals a good deal of variability in the measurements, which may account for apparent enrichment in some on-site samples.

In the aggregate, the mean U235:U238 ratio for all on-site soils was 0.068 ± 0.005 [95% confidence interval (CI)] compared to a background value of 0.071 ± 0.013 . Thus, the means of both on-site and background soil samples exceeded the expected U235:U238 ratio of 0.044, indicating a high bias in the measurements. While this bias may be the result of interferences in the measured gamma ray peaks or inaccuracies in spectral background subtraction, no specific cause has been identified.

Uranium isotope ratio measurements by alpha spectroscopy are also shown in Table I. The mean U-234:U-238 activity ratio in site soils was 0.997, or effectively equal to the value for natural uranium of 1.0, while the background mean of 0.947 was similarly close to natural uranium. Conversely, the mean U-235:U-238 activity ratio in site soils was 0.088, or double the expected value of 0.044, and was similarly elevated in background soil at 0.093. Of these two conflicting results – uranium-234 would not be a natural levels when uranium-235 is somewhat enriched – the U-234:U-238 ratio would seem more reliable. Many of the U-235 results were near detection limits and might have been systematically biased high. In addition, contributions to U-235 abundance from U-236 in RU would tend to bias these ratios high.

After reviewing gamma and alpha spectroscopy results from the initial phase of the investigation and finding that results were not conclusive due to uncertainties in the underlying isotope abundance measurements, ICP-MS was used to determine uranium isotopic ratio measurements with higher precision and accuracy than is typically possible with gamma or alpha spectroscopy.

ICP-MS Soil Results

Results of uranium isotope ratio measurements by ICP-MS in soil, groundwater, and surface water (in sewer lines) samples are shown in Table II. Samples were analyzed for the isotopes uranium-233, uranium-234, uranium-235, uranium-236, and uranium-238. Results originally reported in terms of mass have been converted to activity for comparison to results from gamma and alpha spectroscopy.

Ten background soil samples were taken from the Cleveland Metroparks property near the site. None of the background samples had detectable levels of U-234, but all had detectable levels of U-235 and U-238. U-235:U-238 activity ratios in background soils had an average value of 0.0496 ± 0.0007 in the 10 samples. Compared to an expected value of 0.044 for natural uranium, this result indicates a high bias in the measurements **Table II. Uranium Isotope Activity Ratios by ICP-MS for Soils, Groundwater and Surface Water**

Medium and Sample Source	U-235:U-238 ^a	U-234:U-238 ^a
Soil - Background	0.0496 ± 0.0007 (N=10)	ND ^b
Soil – On-site	0.0410 ± 0.0009 (N=30)	0.874 ± 0.026 (N=14)
Groundwater - Background	0.0410 (N=1)	ND
Groundwater – On-site	0.0403 ± 0.0016 (N=10)	0.928 ± 0.033 (N=4)
Surface Water – On-site ^c	0.0405 ± 0.0010 (N=3)	0.887 (N=1)
Natural uranium	0.044	1.0

^aMean activity ratio \pm 95% confidence interval computed using N results.

^bND indicates that no samples had values of both isotopes above detection limits.

^cSurface water taken from the site sewer system; no background samples were taken.

in background samples of about 8.5%. This bias may or may not also be present in on-site soil samples where uranium levels are considerably higher and perhaps free of bias that might be present nearer detection limits.

A total of 30 site soils were analyzed, all of which had measurable U-235:U-238 activity ratios, with a mean of 0.0410 ± 0.0009 , or slightly depleted relative to either the measured background value, or natural uranium. In addition, nine of the 30 samples had detectable levels of U-236, a marker of recycled uranium, which is good evidence of the presence of recycled uranium, as this isotope is not present in natural uranium.

The mean U-238 concentration in the 30 samples was 333 mg/kg compared to 3.0 mg/kg in the 10 background samples. The measured U-235:U-238 ratio is consistent with the presence in site soils of residuals of RU from Hanford, which is known to have been slightly depleted. On a mass basis, this U-235:U-238 ratio converts to 0.64%, which is similar to the reported value of 0.67% for Hanford RU processed at Harshaw [1, 4]. Natural uranium has 0.71% U-235.

Fourteen of the 30 soil samples had measurable U-234:U-238 ratios. Ratios had a mean value of 0.874, similarly indicating slightly depleted levels compared to natural uranium. U-234 was not detectable in background samples due to its low mass concentration owing to its relatively short half-life, so comparisons to background ratios are not possible.

Figure 2 shows U-235:U-238 ratios in soil as a function of U-238 levels, which range from background levels (2-3 mg/kg) up to 1460 mg/kg. This data suggests a slight increase in depletion with increasing U-238 levels. Near background concentrations isotope ratios are near natural levels and become more depleted at higher uranium levels, which might be the result of spills of slightly depleted RU feed materials.

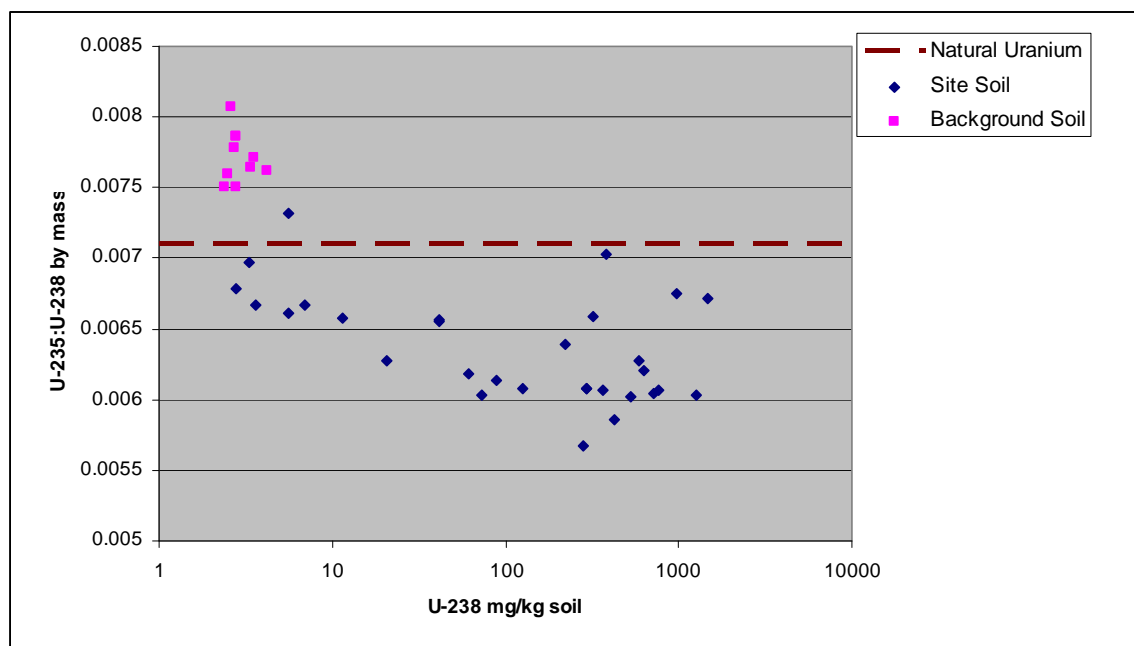


Fig. 2. U-235:U-238 mass ratios as a function of U-238 concentration in soil.

Identification of RU can be further evaluated by examining the relative amount of U-236 in soils, an isotope not present in natural uranium. Figure 3 shows a plot of U-236:U-238 as a function of U-238 soil concentration on a mass basis for nine soils samples that had detectable U-236. The average U-236:U-238 ratio was 0.000028, or 0.0028% U-236 by mass. The maximum ratio was 0.0056%, which occurred at the maximum U-238 concentration of 1,460 mg/kg.

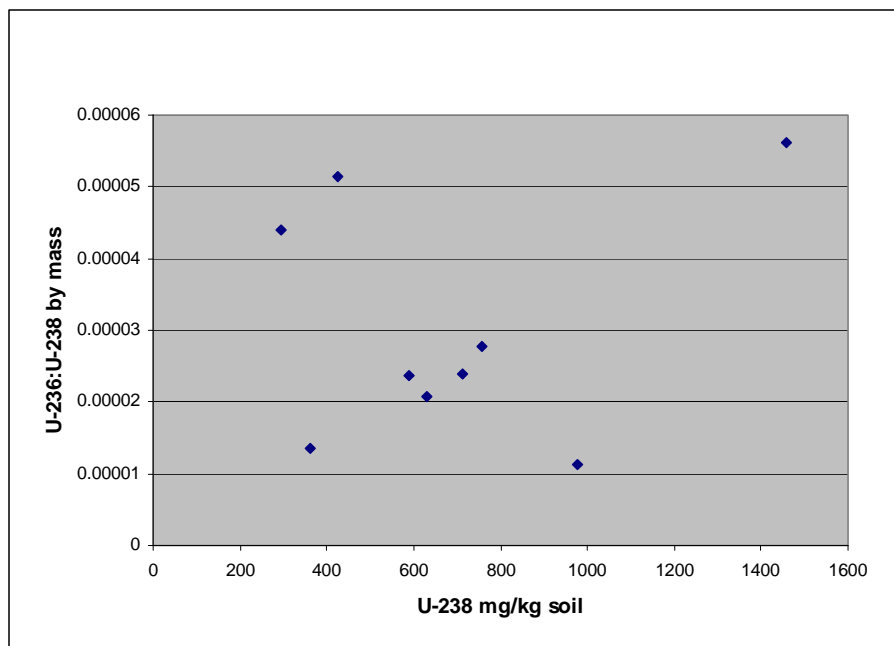


Fig. 3. U-236:U-238 mass ratios as a function of U-238 concentration in soil.

Figure 3 shows no definitive trend in U-236:U-238 with increasing U-238 concentration. This result might be due to the fact that U-236 was only detectable in soils with high U-238 concentration, i.e., above about 300 mg/kg. However, several locations with U-238 above this level had low or undetectable U-236 levels, indicating separate contamination by natural uranium from other feed materials.

Individual samples, likewise, exhibit no particular correlation between the level of U-236 and U-235:U-238 ratios in the same samples (data not shown). That is, samples with the most depleted ratios did not have the highest levels of U-236. This result might be due to the low levels of U-236 involved and to uncertainties in individual measurements of U-236 and U-235:U-238 ratios. Thus, both U-236:U-238 and U-235:U-238 ratios are considered in the aggregate as evidence of the presence of depleted recycled uranium.

Documented values of U-236:U-238 in RU sent to HCC are not available. Reported values from a later period were from 0.016 to 0.071% by mass in Hanford RU produced during the 1980s [1], which is higher by 6-25 times than the average 0.0028% detected in Harshaw site soils. Such lower levels of U-236 in soils might be due to the fact that Hanford RU in the 1940s was slightly depleted in U-235, and thus also in U-236, while that produced in the 1970s-1980s was slightly enriched in U-235 (average = 0.86%) and, thus, even more so in U-236 [1]. It is also possible that U-236 levels are diluted by the co-presence of natural uranium.

ICP-MS Groundwater and Surface Water Results

Uranium isotope ratios measured in groundwater are shown in Table II. ICP-MS measurements were made on 17 site groundwater samples. Unfiltered groundwater samples were analyzed so as to include both dissolved and particulate phases. Of these measurements, 10 had measurable levels of U-235 and U-238 and four had measurable levels of U-234 and U-238. No sample had a measurable U-236 level.

The mean U-235:U-238 activity ratio in site groundwater samples 0.0403, was nearly the same as that in site soils (0.0410), while the average U-234:U-238 activity ratio of 0.93 was close to the value of 0.87 in on-site soils (Table II).

Four background groundwater wells were sampled, and uranium isotopes analyzed by ICP-MS. Detectable levels of U-238 were found in all four samples, but only one well had detectable U-235, just above the detection limit, and none had detectable U-234. The U-235:U-238 activity ratio determined in this sample was 0.0410, very nearly the same value as for site groundwater. However, this background well, which was near the site border might have been impacted by the site, as indicated by a somewhat elevated level of U-238 compared to the other background wells.

Figure 4, below, shows the same U-235:U238 ratios in groundwater in terms of mass as determined by ICP-MS. Measured ratios are all below the value of 0.71 in natural uranium.

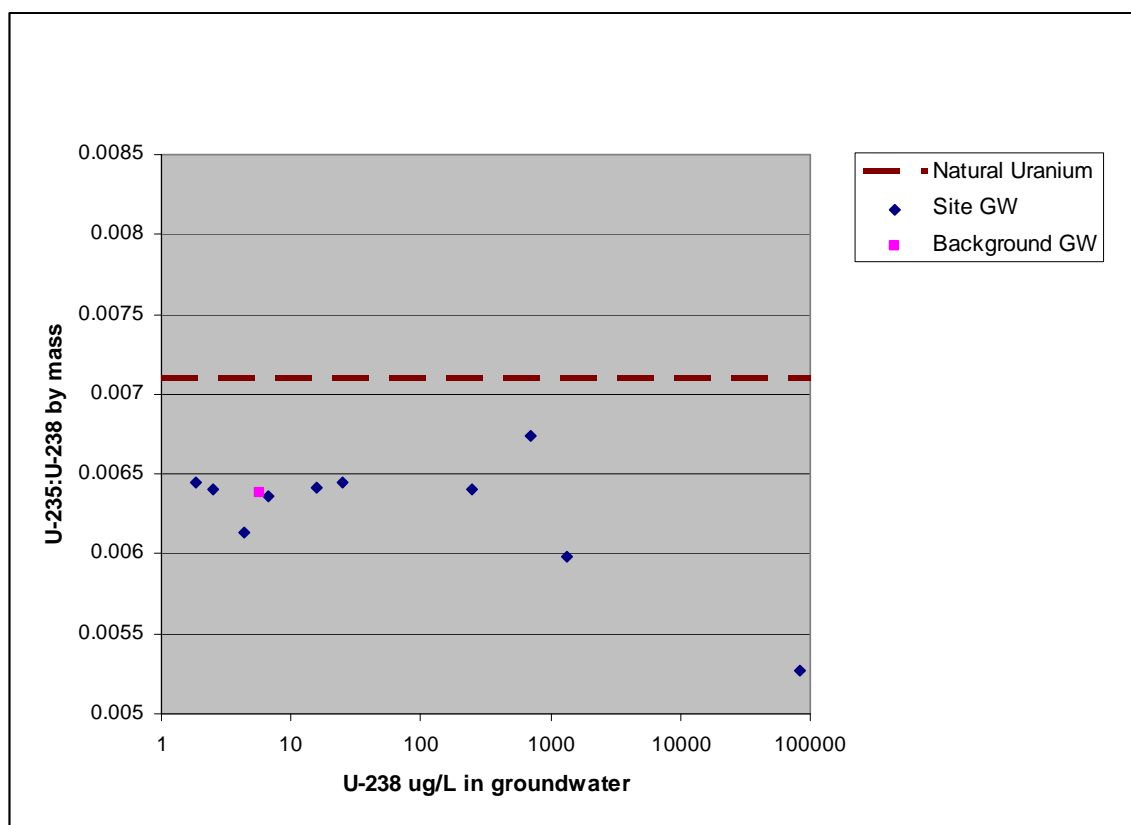


Fig. 4. U-235:U-238 mass ratios in groundwater as a function of U-238 concentration.

While these results suggests that U-235:U-238 ratios in site groundwater are not depleted (nor enriched) relative to background, the site values do indicate depletion relative to standard ratios for both U-234:U-238 and U-235:U-238. Given the more robust measurements in on-site groundwater, both in terms of numbers of detects and overall isotope levels, it might be concluded that on-site groundwater has the same U-234:U-238 and U-235:U-238 ratios as site soils, and that these ratios are slightly depleted on average relative to natural uranium. A lone background groundwater sample would not be sufficient to firmly establish a measured background ratio.

Four surface water samples taken from site storm sewers were analyzed, which produced an average U-235:U-238 activity ratio of 0.0405, essentially the same as site groundwater and background groundwater, and a single U-234:U-238 activity ratio result of 0.0887, which was similarly consistent with groundwater

values. These results could be explained from the possible infiltration of groundwater into the storm sewers.

ICP-MS Results Summary

In summary, uranium isotope ratios in site soils, groundwater, and surface water measured by ICP-MS indicate the presence of residuals of slightly depleted uranium from processing of RU from Hanford. Measured U-235:U-238 ratios are similar to documented ratios in Hanford feed. Conversely, there is no evidence of the presence of enriched uranium in any of the same samples. No site samples exceeded U-235:U-238 ratios measured in background soils, while background groundwater results were insufficient for such comparisons relative to site groundwater samples. However, results for site soils, groundwater, and surface water results indicated slightly depleted levels of U-234 and U-235 compared to standard values for natural uranium. Areas of presumed natural uranium contamination were also detected in soil, as indicated by reduced or undetectable levels of U-236 relative to U-238 levels. It is likely that there are areas of mixed natural and depleted uranium contamination in the vicinity of Building G-1, since large quantities of feeds of each type were handled there.

Of the three analysis methods used, only ICP-MS produced measurements of both U-234:U-238 and U-235:U-238 ratios that had high precision, measured both ratios simultaneously, and produced results that were both internally consistent and consistent with site records. This being the case, the ICP-MS measurements of uranium isotope measurements are considered to be definitive measurements, while gamma and alpha spectroscopy methods are considered secondary in this regard. However, given the greater site coverage of samples analyzed by the latter two methods and the fact that their respective measurements bound the range of isotope ratios, they provide good evidence that the more limited ICP-MS data can be extended across the site.

RECYCLED URANIUM AND CONTAMINANTS

Recent reports of historical production and shipment of RU since the 1950s indicate that HCC had received small quantities of radiological contaminants in RU from Hanford [2, 5]. In light of these reports, plutonium isotopes, Np-237, and Tc-99 were investigated in site soils, surface water, and groundwater.

Records indicate that RU was sent to HCC to be purified prior to further processing at the K-25 Gaseous Diffusion Plant in Oak Ridge, Tennessee. Contaminants would have been separated from RU feed, likely in the form of UO_3 , in the refinery plant in Building G-1, which was built to process uranium ore concentrates in 1952-1953. Any releases of the RU feed and associated contaminants to site soils and groundwater, thus, would most likely have occurred during handling in the vicinity of Building G-1 and adjacent rail yard.

The estimated concentration of radiological contaminants in recycled UO_3 feed ranged from low-ppb levels for plutonium isotopes to low-ppm levels for Tc-99 [2, 6]. Levels in site soils, then, would be expected to be even lower levels and might be below analytical detection limits. Levels in groundwater and surface water (inside sewer lines) resulting from soil leaching, would be even lower. Plutonium and neptunium as oxides would be expected to be primarily adsorbed to soils, while technetium is known to be environmentally mobile under oxidizing conditions (as Tc VII), and would be more likely to appear in groundwater than the other contaminants, especially given its greater level in the feed material.

An investigation of RU residuals was designed that employed archived samples selected from areas where elevated uranium contamination had been previously found, plus additional biased and systematic

samples based on process knowledge. Sampling was done in soil, groundwater, and in sewer lines. Background soil and groundwater samples were also collected.

RU Constituents Results and Discussion

Archived or new samples collected from site soils, groundwater, and storm sewer water and sediments were analyzed at a commercial laboratory (Test America) by various methods, including gamma and alpha spectroscopy and ICP-MS for determining uranium isotopes associated with RU. Detection of RU contaminants in soil and groundwater were infrequent. In soil samples, the collocation of up to three contaminants was taken as evidence of the presence of RU residuals, while similar evidence was not present in groundwater, as discussed in the following paragraphs.

Contaminants of interest in RU included Tc-99, Eu-152, Eu-154, Np-237, Pu-238, Pu-239, Pu-230, and Am-241. In addition, the isotopes U-233 and U-236 were of interest as markers of RU. The results of sample analysis for soil and groundwater are given in Table III. In addition to results in these media, one of two storm sewer sediment samples near Building G-1 had coincident detections of Pu-239/240 and U-236. In four storm sewer water samples analyzed there was only a single detection of one contaminant (Np-237).

Table III. Frequency of Detection of RU Constituents in Site Soil and Groundwater Samples

Constituent ^a	Soil			Groundwater	
	Frequency of Detection ^b	Frequency of two co-located hits ^c	Frequency of three co-located hits ^d	Frequency of Detection	Frequency of two co-located hits
Am-241	0/39	0/39	0/39	1/18	0/18
Eu-152/154	0/39	0/39	0/39	0/18	0/18
Np-237	1/22 ^e	0/22	0/22	6/18	0/18
Pu-238	0/30	0/30	0/30	0/18	0/18
Pu-239/240	6/30	6/30 (U-236)	2/30 (Tc-99)	0/18	0/18
Tc-99	2/30	2/30 (Pu-239)	2/30 (U-236)	2/20	1/20 (U-236)
U-236	9/30	6/30 (Pu-239)	2/30 (Tc-99)	2/20	1/20 (Tc-99)

^aU-238 was detected in all samples in addition to the listed constituents.

^bFrequency does not include off-site blanks, which had no detections of RU constituents.

^cThe most frequently detected co-contaminant is listed in parentheses.

^dThe listed co-contaminant is in addition to those listed in columns to the left.

^eNp-237 was not included in all analyses.

As shown in Table III, the co-location of up to three RU constituents in several soil samples is good evidence of the presence of RU and its associated radiological contaminants. It is highly unlikely that the observed co-located hits are due to pure chance from individual false positives.

Further evidence of the presence of RU is the appearance of the isotope U-236 in several locations. U-236 is not found in natural uranium and, as noted above, was not detected in background soil or groundwater samples. While the U-236 detections are intermittent and low level, they are generally collocated with similarly low-level detects of Tc-99 and Pu-239/240. Together, these results indicate fairly conclusively that residuals of RU are present in the Former Harshaw Chemical Site soils.

At the same time, the very infrequent or non-detection of Am-241, Eu-152, Eu-154, Np-237, and Pu-238 in this sampling effort suggests that these contaminants, while theoretically present in RU, are not present at detectable levels in site soils or groundwater.

U-236 was found in only one of 21 groundwater samples at barely detectable levels. A few sporadic detections of similarly low levels of RU contaminants were found in groundwater, including isolated detections of Tc-99, Pu-239/240, and Am-241. In contrast to soil detections, however, these constituents were not collocated in groundwater samples, with the exception of a single coincident detection of Tc-99 and U-236. Thus, while trace levels of RU constituents appear sporadically in groundwater samples, the evidence for RU contamination is not as strong as that for soils, where constituents tended to be collocated. It is possible that Tc-99 and U-236 might be present, while the few other groundwater hits were false positive results.

Summary of RU Analysis

Residuals of RU in soils were found mainly in the vicinity of Building G-1 and the adjacent rail yard located to the north, as confirmed by the coincident detection of up to three constituents. No RU residuals were confirmed in any other investigation area. The locations of the detections and the ratios of constituents found are consistent with contamination from handling RU feed materials near Building G-1 prior to processing. Soil contamination around Building G-1 appears to have different areas contaminated with RU feed and ore concentrate feed, based on the appearance of U-236. Groundwater is possibly impacted by Tc-99 in addition to uranium from RU, as indicated by a single coincident detection of Tc-99 and U-236.

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