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Utilizing Isotopic Uranium Ratios in Ground-water Evaluations at NFSS

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ABSTRACT

The U.S. Army Corps of Engineers (USACE) Buffalo District is currently evaluating environmental contamination at the Niagara Falls Storage Site (NFSS) under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) as part of its Formerly Utilized Sites Remedial Action Program (FUSRAP). The NFSS is located in the Town of Lewiston in western New York and has been used to store uranium-contaminated materials since 1944. Most of the radioactive materials are currently contained in an on-site structure, but past contamination remains in soil and ground water. As a naturally occurring radionuclide, uranium is present in all ground water. Because contamination levels at the site are quite low, it can be difficult to distinguish zones that have been impacted by the past releases from those at the high end of the natural background range. The differences in the isotopic ratio of uranium-234 (U-234) to uranium-238 (U-238) between natural ground-water systems and affected areas are being used in an innovative way to better define the nature and extent of ground-water contamination at NFSS. In natural ground water, the ratio of U-234 to U-238 exceeds 1 due to the alpha particle recoil effect, in which U-234 is preferentially mobilized to ground water from adjacent rock or soil. This process is very slow, and it can be hundreds to thousands of years before a measurable impact is seen in the isotopic ratio. Thus, as a result of the recoil effect, the ratio of U-234 to U-238 will be higher in natural ground water than in contaminated ground water. This means that if site releases were the source of the uranium being measured in ground water at NFSS, the ratio of U-234 to U-238 would be expected to be very close to 1 (the same ratio that exists in wastes and soil at the site), because not enough time has elapsed for the alpha particle recoil effect to have significantly altered that ratio. From an evaluation of site and regional ground-water data, an isotopic ratio of 1.2 has been identified as a site-specific signature to help distinguish natural ground water (e.g., at the high end of the background range) from zones impacted by past releases. This information is crucial for focusing the ongoing CERCLA evaluation and decision making process. This signature value is not applied as a bright line, e.g., to define samples with ratios of U-234 to U-238 above 1.2 as representing background and those with ratios below 1.2 as being affected by site releases. Rather, this ratio serves as a weight of evidence for use in conjunction with other site information, including historical activities, to form science-based decisions regarding contaminated ground water. This novel approach for developing a ground-water signature from the isotopic uranium ratio has proven to be a very useful tool for NFSS, and it is now being considered for broader application.

INTRODUCTION

All soils and rock exhibit differing levels of radioactivity, largely associated with the varying levels of naturally-occurring potassium, uranium, thorium, and radium. Oftentimes the radioactive properties of soils and rock, along with the ratios at which they exist, are used for geologic mapping purposes. For example, the ratio of potassium to thorium may provide an indication of igneous and metamorphic mineralization. In soil and radioactive wastes containing natural uranium, the ratio of uranium-234 (U-234) to uranium-238 (U-238) is essentially 1. As a naturally occurring radionuclide, uranium is present in all ground water. However, in ground water, the ratio increases with time because of the alpha particle recoil effect. When a U-238 atom undergoes radioactive decay by emitting an alpha particle to form thorium-234 (Th-234), its nucleus recoils in the opposite direction to conserve momentum. If this U-238 atom is within rock or soil that is in contact with ground water, the relatively short-lived daughter product Th-234 can have sufficient recoil energy to shortly enter the aqueous phase and then precipitate from solution. This Th-234 atom transforms fairly quickly to U-234 through two successive beta-particle decays (see Figure 1 and Table I); this atom is generally soluble. Thus, the alpha particle recoil effect could preferentially mobilize U-234 into adjacent ground water when it would otherwise have sorbed to solid material [1].

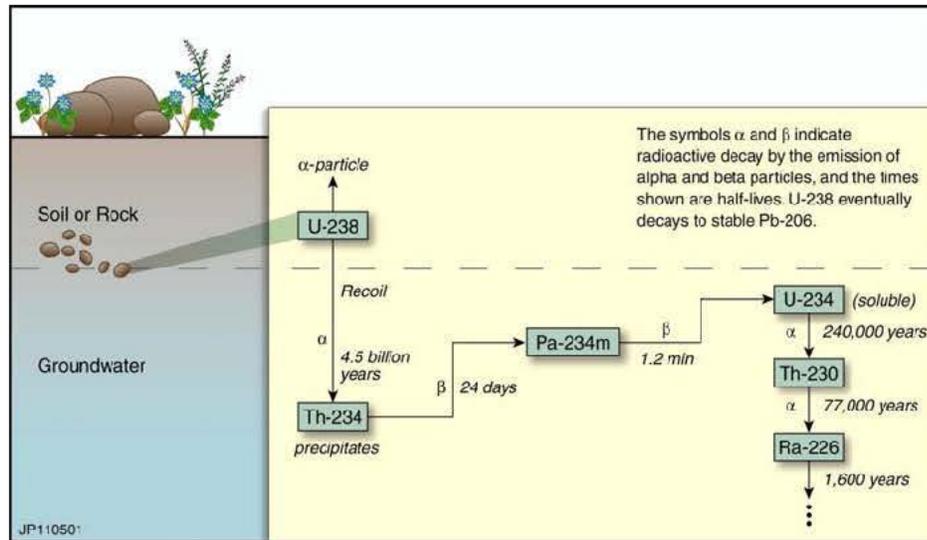


Fig. 1. Radioactive Decay of U-238 and U-234 [2].

Although only a very small fraction of U-238 decays produce this effect because it is limited by the presence of a phase interface and recoil geometry, over considerable time, ground-water concentrations of U-234 can become much higher than those of U-238. Since uranium is a contaminant at many milling, processing, storage, and disposal sites, this phenomenon makes the isotopic uranium ratio a good candidate for a signature to indicate contaminated versus uncontaminated ground water when the distinction is unclear.

The ratio of U-234 to U-238 in natural ground water typically ranges from 1 to 3 but can reach 10 or more (a value of 28 has been reported [3]). High ratios usually reflect very old ground water in deep formations. In evaluating ground-water data at sites contaminated with natural uranium, the U-234 to U-238 ratio would be expected to be near 1 if site materials were the source of the uranium being measured (additional evaluations are warranted if depleted or enriched uranium is present). This ratio of near unity occurs because not enough residence time would have elapsed to allow the alpha particle recoil effect to increase the ratio. In contrast, background ground water would be expected to exhibit a higher ratio, especially for deeper hydrogeologic units. Determining site-specific U-234 to U-238 ratios and applying them as signature indicators of contaminated versus uncontaminated areas can provide a very practical, scientific basis for framing the boundaries of contaminant plumes for cleanup decisions.

Naturally occurring uranium in soil typically ranges from about 1 to 5 picocuries per gram (pCi/g) [4], although values can be much higher depending on local geological conditions. This natural radioactive element exists primarily as three isotopes: U-234, uranium-235 (U-235), and U-238. The relative activity concentrations of these isotopes in soil and rock (i.e., U-234:U-235:U-238) are in the approximate ratio of 1.0:0.046:1.0. The main interest for this analysis is the ratio of the two most prominent isotopes, U-234 and U-238, since they occur in the same radioactive decay series. Basic properties of the first four radionuclides in that series are summarized in Table I (including primary decay modes). The two short-lived decay products of U-238 are italicized in this table.

Table I. Properties of Naturally Occurring Uranium Isotopes

Radionuclide	Half Life	Specific Activity (Ci/g)	Decay Mode	Natural Abundance
Uranium-238	4.5 billion yr	0.00000034	α	>99%
<i>Thorium-234</i>	<i>24 days</i>	<i>23,000</i>	β	
<i>Protactinium-234m</i>	<i>1.2 min</i>	<i>690,000,000</i>	β	
Uranium-234	240,000 yr	0.0063	α	0.0055%

Concentrations of naturally occurring uranium vary more in ground water than in soil, due to location-specific geological, hydrological, and geochemical factors. Those factors include the uranium concentration in the host rock,

the nature of the contact between the uranium minerals and ground water, the presence of oxygen and complexing agents, and chemical reactions with dissolved ions [3]. Naturally occurring uranium concentrations in ground water have been reported to range from <1 to 650 pCi/L [5], with values generally falling between 1 and 10 pCi/L. Recent data indicate that natural concentrations could be much higher, with a value above 6,000 pCi/L reported for a well in South Carolina [6]. While the activity concentrations of U-234 and U-238 in soil and rock are essentially equal, in ground water this ratio usually exceeds 1 and is often near 2 [7].

The U-234 to U-238 ratio is affected by site-specific hydrogeological and geochemical characteristics, and different ratios would be expected in different water-bearing zones, with a higher ratio in deeper zones hydraulically separated from recent recharge. This phenomenon makes the isotopic uranium ratio a good candidate for defining a ground-water signature at sites contaminated with anthropogenically-derived uranium, i.e., to indicate contaminated versus uncontaminated areas when the distinction is otherwise unclear. This approach can be particularly useful to distinguish between results at or only slightly above the high end of a background range, and indicative of those site contributions or natural variability. While the utility of this concept is most apparent for sites that processed and stored natural uranium, it may be useful for sites that handled enriched or depleted uranium, although some modifications would be needed to assess expected uranium ratios associated with site-related contamination.

CASE STUDY – NIAGARA FALLS STORAGE SITE, LEWISTON, NY

The U.S. Army Corps of Engineers (USACE) Buffalo District is evaluating conditions at the Niagara Falls Storage Site (NFSS) under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) as part of its Formerly Utilized Sites Remedial Action Program (FUSRAP). The NFSS is located in the Town of Lewiston in western New York and has been used for the storage of uranium-contaminated materials since 1944. In the 1980s, the U.S. Department of Energy (USDOE) conducted a number of interim remedial actions under FUSRAP to stabilize radioactive materials at the site, most of which are enclosed in the on-site interim waste containment structure (IWCS). The Energy and Water Appropriations Act for Fiscal Year 1998 transferred responsibility for administering and executing FUSRAP from the USDOE to the USACE in October 1997; the USACE has maintained control of the site since then. However, responsibility for long-term stewardship of the site will revert back to the USDOE two years after remedial actions are completed, in accordance with a memorandum of understanding between these two agencies.

Ground water at NFSS contains elevated (i.e., above background) levels of a number of radioactive and chemical contaminants (including uranium) in several isolated plumes. It is challenging to determine the full aerial extent of uranium contamination in these plumes because of its natural presence in host soils. Developing a site-specific signature for uranium contamination versus naturally occurring uranium will help to better define the nature and extent of contamination at the site, which in turn will provide useful information for the development and evaluation of remedial action alternatives under the ongoing CERCLA process. Because the site will eventually be transferred back to the USDOE, a clear definition of the extent of ground-water contamination is of interest to both federal agencies, i.e., to the USACE for framing remedial actions and to the USDOE for framing long-term surveillance and maintenance actions.

Evaluation of Background Ground-water Data and the Development of a Site-Specific Ratio

The NFSS has been used to store uranium-contaminated materials in their natural isotopic ratios for over a half of a century; the contaminated soils and residues at the site (contained within the IWCS) have a U-234 to U-238 ratio of essentially 1. If these materials were the source of the uranium measured in ground water at the site, the U-234 to U-238 ratio from ground-water data would be expected to be close to 1 because too little time has elapsed for alpha particle recoil to have measurably affected the U-234 to U-238 ratio. In contrast, background ground water would be expected to exhibit a somewhat higher ratio, especially for deeper units storing older ground water.

Evaluating the background concentration of uranium in ground water at NFSS is central to both determining the nature and extent of site contamination and supporting assessments of human health and ecological risks. Ground-water samples were collected from nearby areas not expected to have been impacted by site releases, and the measured concentrations were then used to identify local background conditions. In evaluating these data, a variety of information was reviewed to provide a weight-of-evidence indication for this background determination. This information included historical activities, physical and geochemical properties of the soil and rock, the nature of the

measurement (including its depth, the screened lithologic unit, and data validation/verification context), and an understanding of what levels are clearly considered reference background concentrations for the region.

Uranium isotopic concentrations sampled from nearby wells were tentatively identified as representing background ground water. Isotopic uranium concentrations resulting from radiochemical analysis using alpha spectroscopy with a four hour count time are reported in Table II below. Two values are generally reported for each isotope in each well, one for the total concentration and one for the dissolved concentration. The dissolved value represents the concentration of the sample after it was passed through a 0.45 micron filter to remove entrained solids, while the value for total uranium reflects the unfiltered sample. These values are very similar, meaning that most of the uranium in these samples is in solution. Table II lists the uranium concentration data and associated isotopic ratios for background ground-water wells in the order of increased well depth (in meters above mean sea level (meters AMSL)).

Table II. Background Uranium Ground-water Concentrations and Isotopic Ratios by Lithology^a

Well Location	Well Depth (meters AMSL)	Screened Lithology	Uranium Concentration (pCi/L)				Total U-234:238	Dissolved U-234:238
			Total		Dissolved			
			U-234	U-238	U-234	U-238		
GW-PZ23S	95.75	UCT	4.65	3.65	-	-	1.27	-
GW-PZ18S	94.49	UCT	7.32	5.79	7.23	5.93	1.26	1.22
GW-PZ21S	94.45	UCT	13.7	11.9	10.9	9.04	1.15	1.21
GW-PZ25S	94.30	UCT	20.8	19.7	20.9	19.8	1.06	1.06
GW-GW2A	93.25	GLC	8.19	4.36	7.06	4.54	1.88	1.56
GW-SP2M	93.23	ASG	4	3.13	8.28	6.32	1.28	1.31
GW-SP5M	93.06	ASG	5.29	4.27	6.5	4.8	1.24	1.35
GW-PZ21M	92.52	GLC	8.73	3.4	8.94	3.35	2.57	2.67
GW-MW17	92.44	ASG	4.1	3.15	4.44	3.12	1.3	1.42
GW-W12	92.11	ASG	5.62	3.92	5.86	4.15	1.43	1.41
GW-SP1M	90.96	ASG	1.83	1.43	2.21	1.67	1.28	1.32
GW-GW2B	89.86	BRT	1.41	1.15	1.42	1.04	1.23	1.37
GW-PZ18M	89.29	GLC	1.56	0.616	1.05	0.55	2.53	1.91
GW-SP9M	89.25	ASG/BRT	1.54	1.1	1.63	1.07	1.4	1.52
GW-W11	88.64	QFM	0.674	0.322	0.687	0.42	2.09	1.64
GW-W12D	88.03	QFM	1.14	0.604	1.23	0.695	1.89	1.77
GW-W3R	87.24	QFM	0.539	0.247	0.628	0.295	2.18	2.13
GW-W13D	86.63	QFM	0.608	-	0.409	0.229	-	1.79
GW-SP14D	86.51	QFM	0.324	0.116	0.279	0.17	2.79	1.64
GW-W14D	85.74	QFM	-	-	0.557	0.195	-	2.86
GW-PZ21D	85.52	QFM	0.39	-	0.196	0.109	-	1.80
GW-SP9D	84.88	QFM	1.29	0.954	1.09	0.729	1.35	1.50
GW-PZ7M	84.85	QFM	0.452	0.3	0.68	0.469	1.51	1.45
GW-PZ8D	84.62	QFM	0.605	0.425	0.964	0.245	1.42	3.93

^a Shown are total and dissolved concentrations of U-234 and U-238 from wells initially identified as background for site ground water. Also shown are U-234 to U-238 ratios; the site-specific cutoff for background was identified as 1.2. A hyphen indicates the concentration was either not detected or available. Two wells potentially impacted by the site in this data set are bolded (PZ-21S and PZ-25S). Lithology acronyms used in this table are defined in Figure 2.

The data sorted by well depth show that the ratios of the U-234 to U-238 concentrations are typically higher in the bedrock unit (shaded cells in Table II) than the overburden, as would be expected from the alpha particle recoil effect for naturally occurring uranium in older (deeper) ground water.

Although initially considered as part of the background data set, the ratios from piezometers PZ-21S and PZ-25S (bolded in Table II) screened in the upper zone were significantly lower than other locations, and thus may not represent background, but be impacted by site releases. These two piezometers are located along an old rail line in an area where the soil was heavily disturbed in the past. Two factors suggest that these measurements might reflect site-related contamination: (1) the rail line could have been used to transport radioactive materials for the site, and (2) the higher porosity of disturbed soils compared to undisturbed soils could enhance leaching of potential soil contamination to ground water, such that the impact might be seen within several decades.

In addition, the U-234 and U-238 measurements for all other locations are less than 9 pCi/L, whereas the concentrations from PZ-21S and PZ-25S are noticeably higher than the others. In addition, most wells show U-234 to U-238 ratios ranging from 1.23 to 2.79 for total concentrations, and from 1.22 to 3.93 for dissolved concentrations. The ratio of U-234 to U-238 in PZ-25S is 1.06 for both total and dissolved uranium. These results show a clear distinction between the U-234 and U-238 concentrations reported for PZ-25S and those for the other wells identified as background. The much higher uranium concentrations coupled with the low ratio of U-234 to U-238 strongly suggest that the uranium reported for PZ-25S is largely attributable to past site releases.

The evidence for PZ-21S also supports a similar conclusion but is somewhat less compelling. The uranium concentrations in this piezometer are about 50% higher than the next highest value, and the concentration ratios of U-234 to U-238 are 1.15 for total uranium and 1.21 for dissolved uranium. While these ratios are smaller than those for the other wells, they do not provide conclusive evidence that the uranium from this piezometer largely reflects site releases. It appears that PZ-21S might have been impacted to a small extent by site releases, but not like PZ-25S. That is, uranium in ground water at that location could represent a combination of naturally occurring levels with a relatively small contribution from NFSS releases.

The concentration ratios in PZ-18S are only slightly higher than those in PZ-21S, which might indicate that these data also reflect a combination of naturally occurring uranium with a small contribution from NFSS materials. However, the uranium concentrations themselves are much lower and are comparable to the concentrations reported for several other background wells, such as GW-2A and PZ-21M. The isotopic ratios for GW-2A and PZ-21M exceed 1.5 for both total and dissolved concentrations, which would indicate that these wells are measuring naturally occurring rather than site-related uranium. These data together indicate that PZ-18S may be slightly impacted by site releases (similar to but less than PZ-21S), but are not sufficient to support deleting this well from the background data set.

The evidence that PZ-25S has been impacted by site releases is strong, considering the historical context and physical observations, and the higher-than-expected uranium concentrations and lower U-234 to U-238 ratios compared with wells considered representative of local background. This conclusion also applies for PZ-21S, although the evidence is not as compelling.

Although uranium concentrations in PZ-21S are higher than those in other wells, the U-234 to U-238 ratios are not so much lower than the ratios for other wells to make a definitive conclusion about background. This piezometer likely reflects naturally occurring uranium with a relatively small contribution from the site. While the isotopic ratios for PZ-18S are somewhat similar to those for PZ-21S, the measured concentrations are much lower, which indicates that this piezometer could reflect the higher end of natural uranium in ground water.

On the basis of these data and physical considerations, a decision was made to remove the PZ-21S and PZ-25S data from the data set being used to calculate background uranium concentrations in ground water. The resulting values for U-234 and U-238 better represent background conditions for the upper water-bearing zone at this site. These findings are being summarized for inclusion in the CERCLA Remedial Investigation Report (RIR) being prepared for this site, and they are also being incorporated in the ongoing human health and ecological risk assessments to support site cleanup decisions.

Once ground water locations PZ-21S and PZ-25S were eliminated from the background dataset for this evaluation, a closer look at the geochemical impacts of various lithologic strata on the isotopic uranium ratio was conducted. Figure 2 illustrates the lithology at NFSS, which consists of a thin Fill, an Upper Clay Till (UCT), a Glacio-Lacustrine Clay (GLC), an Alluvial Sand and Gravel (ASG), and a Basal Red Till (BRT). The bedrock at NFSS is a reddish-brown zone of the Queenston shale formation (QFM).

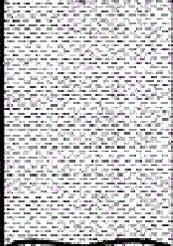
Fill		
UCT		Upper Clay Till: Brown or reddish-brown clay with significant amounts of silt or sand and interspersed lenses of sand and gravel.
GLC		Glacio-Lacustrine Clay: Homogeneous gray clay with occasional laminations of red-brown silt and minor amounts of sand and gravel.
MST		Middle Silt Till: Gray to gray-brown silt with little sand and gravel.
GLC		Glacio-Lacustrine Clay: Homogeneous gray clay with occasional laminations of red-brown silt and minor amounts of sand and gravel.
ASG		Alluvial Sand and Gravel: Stratified coarse sands, non-stratified coarse silt and sand or interlayered silt, sand and clay.
BRT		Basal Red Till: Reddish-brown silt and coarse to fine sand.
QFM		Queenston Formation: Reddish-brown fissile shale.

Fig. 2. NFSS Lithology [8].

It is apparent from Table II that the uranium ratio signature for ground-water wells screened in the overburden is typically less than those from wells screened in bedrock (shaded in Table II), with the exception of those screened in the GLC unit (italicized in Table II). Since the bedrock wells will exhibit a higher uranium ratio due to the older ground water in this deeper geologic formation, it is not readily apparent why the uranium ratio is also elevated for wells in the GLC unit. A likely two-fold explanation is that there is an increased probability for the recoil effect to

occur since a greater number of cations (U-238 atoms) will sorb onto the charged clay particles due to a higher partition coefficient (Kd) driven by greater electrostatic potentials (i.e., negative charge) on the clay. In addition, the slow movement of ground water through this clay unit provides enough residence time for the alpha recoil effect to enrich ground water in U-234. These two conditions increase the probability that the Th-234 atom generated by the U-238 decay will reach the ground water in the clay unit. Because of this, the alpha particle recoil effect in the GLC is more pronounced than in the rock units, resulting in the observed higher U-234:U-238 ratio.

Use of Isotopic Ratio to Evaluate Ground-water Plumes at NFSS

Cleanup decisions for ground water can have substantial cost implications, so the ability to distinguish between ambient levels and those reflecting site contamination is crucial. Ground-water zones that are obviously contaminated are easy to define, but for areas where measurements fall at or slightly above the high end of the background range, it can be difficult to determine whether the data for a given analyte reflect site contributions or natural variability. Thus, a scientific approach for defining practical thresholds to determine plume boundaries would be useful to guide cleanup decisions.

The isotopic uranium ratio signature at NFSS proved to be a useful tool in evaluating ground-water contamination at the site and in the preparation of ground-water contaminant plume maps, which provide a pictorial representation of current ground-water conditions at the site. Typically, background screening values or maximum contaminant levels developed by the U.S. Environmental Protection Agency for drinking water sources are used to define boundaries of uranium plumes. Background screening values were used to define plumes at the NFSS. However, some of the plume boundaries were modified based upon consideration of other factors, such as the uranium ratio signature, as discussed below.

Ground-water plume maps for dissolved concentrations of U-234, U-238, and U-235 were initially developed by comparing the measured concentrations of these three uranium isotopes against the screening values of 8.94, 6.32, and 0.51 pCi/L for dissolved U-234, U-238, and U-235, respectively. These screening values represent the 95% UTL of the background data for these three radionuclides in ground water at the site. Background data for U-234 and U-238 are shown in Table II; similar information was developed for U-235, but it is not included in the table.

In evaluating these initial plumes, it was noted that for some locations a plume would be indicated for one or two of these isotopes (based on comparison to the screening values noted above), but not all three. In these situations, the data were re-evaluated to determine if indeed a plume should be indicated in this area of the site. For example, at one location in the northernmost bounds of the site a plume was indicated for U-235, but no similar plumes were identified for U-234 and U-238. Since enriched uranium is not expected in natural ground water at this site, U-235 should be collocated with U-234 and U-238. In addition, documentation indicated that there was no storage or burial of contaminated waste in this area. The question therefore arose as to whether the dissolved U-235 plume was indeed in excess of background conditions or simply an artifact of the background statistics, since the concentrations of the other two uranium isotopes were not elevated at this location.

To clarify this condition, the dissolved U-234:U-238 ratio signature at this location was calculated to be 1.44. Since the well producing this U-235 “plume” was screened in the UCT unit, the isotopic uranium ratio signature of 1.44 would indicate that the dissolved uranium detected in ground water at this location was likely due to natural background sources, i.e., 1.44 is above the background uranium ratio signature of 1.2. Since uncertainty in the reported result can influence this ratio, it was determined that the isotopic uranium ratio would be 1.2 (indicative of background), even if half of the uncertainty associated with the dissolved U-234 result was subtracted and half of the uncertainty associated with the dissolved U-238 result was added to the measured concentrations. The influence of the data uncertainty can be greatly reduced, thereby increasing the confidence in the isotopic uranium ratio signature, by increasing the alpha count time during analysis.

Current and historical information on site operations, scientific knowledge on the migration of uranium isotopes to ground water, and the application of the background isotopic uranium ratio signature would indicate that the dissolved U-235 “plume” at this location is a result of the background statistics and not site contamination. This is further supported by the background concentrations of U-234 and U-238. This weight of evidence indicates a very low level of confidence associated with this U-235 “plume”, which will be articulated in the RIR.

A similar condition was applied to a preliminary dissolved U-235 ground-water plume located to the east of the IWCS, where a ground-water plume was identified for U-235, but no collocated plumes were evident for the other two uranium isotopes. The isotopic uranium plumes indicate locations where the concentrations of U-234, U-238, and U-235 exceed the background screening values of 8.94, 6.32, and 0.51 pCi/L. As noted above, a ground-water plume for U-235 would be expected to be collocated with plumes for the other two uranium isotopes. This condition flagged the area as a location requiring more detailed analysis.

The U-235 ground-water “plume”, east of the IWCS, was the result of elevated concentrations in two wells, although the concentrations of U-234 and U-238 in both wells were below their respective background screening values. The isotopic uranium ratios in these two wells were calculated to be 1.07 and 1.33; both wells were screened in the UCT unit. The well having the lower ratio (1.07) would be indicative of having been impacted by releases from wastes formerly stored at this site since the value is less than the background signature of 1.2. Conversely, the well having the higher ratio (1.33) was likely not impacted by site releases, in that this ratio is above 1.2. In addition, the dissolved U-235 concentration of 0.684 pCi/L in this second well was only slightly greater than the background screening value of 0.51 pCi/L. This supports the contention that the uranium being measured in the second well is likely associated with natural sources and is not related to site releases.

In contrast, the U-235 concentration in the well having the lower ratio of 1.07 was close to three times the background screening value of 0.51 pCi/L. The isotopic uranium ratio and the measured concentration of U-235 at this location both suggest that this well has been impacted by uranium releases from the site. However, since uncertainty in the reported concentrations of U-234 and U-238 can influence the isotopic ratio, it was determined that the ratio of U-234 to U-238 for this well would be 1.18, if 25% of the uncertainty from the dissolved U-234 concentration was added to and 25% of the uncertainty from the dissolved U-238 concentration subtracted from the measured values. As previously noted, the influence of the data uncertainty can be greatly reduced by increasing the alpha count time during analysis.

Based on these analyses, it was concluded that the U-235 concentration of 0.684 pCi/L in the first well was elevated based solely on the manner in which the data were obtained and analyzed. That is, this well does not likely represent site-related contamination. In contrast, the second well, with a U-235 concentration of 1.33 pCi/L, could have been impacted by site releases. However, the confidence associated with this conclusion is low, as reflected in the uncertainty evaluations associated with the isotopic uranium ratio at this location and the lack of elevated concentrations of U-234 and U-238. This information is also being incorporated into the RIR being prepared for the site.

CONCLUSIONS AND RECOMMENDATIONS

The development of a site-specific isotopic uranium ratio to signify background conditions for the NFSS proved to be a valuable resource in evaluating the measured concentrations of uranium in ground water. This signature ratio helps differentiate between natural uranium in ground water at the high end of background and areas impacted by site releases. This site-specific ratio of 1.2 for U-234 to U-238 has provided the technical team with an additional tool to better define the nature and extent of uranium contamination in ground water. This ratio, when used in conjunction with other site-related information such as previous site activities, hydrogeochemistry, and collocation of contaminants, allowed the technical team to more accurately assess the measured data in a timely manner, and should reduce the likelihood of identifying uncontaminated areas as possibly requiring remediation. This will help ensure remediation resources are properly applied to address site-related releases.

In considering the use of this concept at other sites, it must be remembered that the isotopic uranium ratio signature is site specific. The value of 1.2 given here is only relevant to NFSS and the use of this approach may not be applicable at other sites. It is possible that for some sites the data may yield a large enough uncertainty that renders this application useless, because there is not an obvious distinction between the ratio associated with site-related contamination and background. The data must be of sufficient quality and quantity for use in this application. It is recommended that the alpha count time for the detection of uranium isotopes be sufficient to yield low uncertainty and count be of sufficient quantity to be statistically significant. The higher the quality of the data, the greater the probability that there will be a clear distinction between naturally occurring uranium ratios and those associated with site releases. Finally, the uranium ratio signature is a tool that can support site evaluations, but should not be the sole source for making technically sound decisions.

REFERENCES

1. Arndt, M.F., and L. West, 2005, "A Study of the Factors Affecting the Gross Alpha Measurement, and a Radiochemical Analysis of some Groundwater Samples from the State of Wisconsin Exhibiting an Elevated Gross Alpha Activity," http://www.slh.wisc.edu/ehd/radiochem/dnr_reports/dnrfinal.pdf, accessed January.
2. MacDonell, et al., 2005. "Using Isotopic Uranium Ratios as a Signature for Contaminated versus Uncontaminated Groundwater," 2005 International Conference of Environmental Remediation and Radioactive Waste Management. Glasgow, Scotland, September.
3. Hess, C.T., et al., 1985, "The Occurrence of Radioactivity in Public Water Supplies in the United States," *Health Physics*, 48:553-586.
4. Myrick, T.E., B.A. Berven, and F.F. Haywood, 1983, "Determination of Concentrations of Selected Radionuclides in Surface Soil in the U.S.," *Health Physics*, 45:631-642.
5. Cothorn, C.R., and W.L. Lappenbusch, 1983, "Occurrence of Uranium in Drinking Water in the U.S.," *Health Physics*, 45:89-99.
6. Hughes, L.D., et al., 2005, "Anomalously High Levels of Uranium and Other Naturally Occurring Radionuclides in Private Wells in the Piedmont Region of South Carolina," *Health Physics*, 88:248-252.
7. Kitto, M.E., and M.S. Kim, 2005, "Naturally Occurring Radionuclides in Community Water Supplies of New York State," *Health Physics*, 88:253-260.
8. HydroGeoLogic, Inc., 2002, "Draft NFSS Groundwater Flow Model and Calibration Technical Memo", April.