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Uranium Isotopic Analyses

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Quantitative Evaluation of Mixture Components in RFETS Uranium Isotopic Analyses

Development & Verification/Validation of Calculations using an Excel Spreadsheet

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9/8/2005 (Revised 3/22/2006)

Background

Uranium isotopic compositions include four isotopes (masses 234, 235, 236 and 238). Of these isotopes, three are naturally present (masses 234, 235, 238), and the isotope with mass 236 is present due to reactions that occur within nuclear reactors. The RFETS AME project has, therefore, utilized the presence and amount of ^{236}U as the indicator of uranium contamination. Uranium isotopic measurements are conventionally presented as ratios to ^{238}U , and for the purpose of evaluating isotopic compositions at RFETS, we will utilize ratios of atoms. Note that some compositions and ratios in the literature utilize the alternative units of mass rather than atoms which results in slight shifts in the resulting ratios.

Uranium processing within the nuclear and defense industrial complex has been developed to produce a range of different compositions with differing amounts of isotopic components. Primarily these compositions involve enrichment or depletion in ^{235}U . The three nominal end-member compositions for uranium present at Rocky Flats are natural, highly-enriched, and depleted (Table 1 and Figure 1). In general, highly enriched uranium was carefully physically controlled, recycled and subject to accounting. Depleted uranium was less valuable and substantial amounts were discharged to waste treatment systems, with relatively high levels discharged to the Solar Ponds. Natural uranium was not directly processed at Rocky Flats, but is present in relatively high concentrations in the geologic units that underlie the facility. For further information on uranium materials, processing, waste handling, contamination releases and environmental characterization, see the RFETS Actinide Migration Evaluation Pathway Analysis Summary Report (2002) and supporting Technical Appendices.

Requirements & Design

The purpose of this effort is to quantitatively evaluate the amount (as fractional %) of each end-member present in each field sample from RFETS that was analyzed for uranium isotopic composition. Accuracy of the calculations is targeted to be within 2% for the fraction of natural and depleted uranium.

The compositions presented in Table 1 are accepted as the end-member compositions and ratios. Analytical results in ratio format will be used to calculate the amount of each end-member. Ranges of measured isotopic ratios and plots of sample data from RFETS were used to define the range of compositions that the calculations needed to cover (Figure 2). These results showed that only a small amount of the potential range of compositions was present in the data, focused on compositions very near to the natural and depleted uranium end-members (<1% highly enriched uranium). Synthetic mixtures of the end-members can be defined by adding atomic contributions for each fraction and then calculating the resulting isotopic ratios.

A key aspect of geometric evaluation of the amounts of end-members in a three-component (triangular) mixing system is the linearity of mixing, in this case on the

mixture as a function of isotopic ratios. Natural to depleted mixture trends were demonstrated to be linear through calculation of atom mixtures and consequent isotopic ratios (Figure 3). In contrast, the mixture of natural (or depleted) uranium with highly enriched uranium is highly non-linear (Figure 4) overall, but was found to be very close to linear, but not a direct relationship, across the range of 0-10% highly-enriched uranium in the mixture.

Because, for these RFETS samples, the amount of highly enriched uranium end-member was found to be very small (<1% from Figure 1) and less than the accuracy target for determination of natural and depleted uranium fractions, calculation of the fractional highly-enriched uranium amount can be effectively separated from calculation of the fractional amounts of natural and depleted uranium end-members. The strategy for calculation of highly enriched uranium end-member is (1) to project the sample point to the natural-enriched mix trend along a parallel to the natural-depleted trend [all points on the line of projection have equal fractional amounts of highly enriched uranium], (2) calculate the fractional distance of the projected sample point along the natural to enriched mixture line, and (3) correct the fractional distance into fractional enriched end-member using the nonlinear fit relationship from Figure 4.

The strategy adopted for calculating fraction of depleted end-member was to transform the coordinate system of the mixture triangle to place natural uranium at the origin, depleted uranium along the horizontal axis, and highly enriched uranium along the vertical axis. In detail, this strategy utilizes a shift of the compositions downward, rotation of the triangle in a counter-clockwise direction, and then skewing the triangle to the right to create a right-triangle form. While the most accurate triangular transformation would have equal intersection angles with the horizontal axis, the extremely tall, narrow, form of this triangle (Figure 1) and concentration of data near the base (Figure 2) makes the right triangle effectively equivalent and makes direct measurements simpler. The fractional amount of natural uranium is then calculated by difference. An alternative approach is to project each sample point to the natural-depleted trend line along a line parallel to the depleted-enriched trend (the equivalent approach used for determining the highly enriched end-member fraction), and this alternative was used to validate the depleted and natural uranium fractional amount results. Note that is latter approach would require a slightly more complex evaluation of limits at the extremes of natural and depleted uranium fractions if it was used as the primary method of evaluation (see discussion below).

Verification & Validation

The calculations of end-member fractions were verified and validated quantitatively by calculations utilizing isotopic ratios of synthetic trends. Synthetic trend sets of mixtures (e.g. those plotted on Figure 2) were calculated for mixing between atomic compositions of the end-member components given in Table 1. Atom ratios for each mixture were calculated, and then the fraction of each end-member was calculated using the approaches defined above. The results of these end-member fraction calculations (Table 2) are consistent for most systems with the synthetically specified fractions within the range 0-1% of highly enriched uranium and within 2% accuracy for the calculated fraction across all mixtures of depleted and natural uranium. Inaccuracy

at 1% to 10% amount of highly-enriched uranium fraction is primarily due to the simple approach of determination of natural uranium by difference.

A second quantitative validation was achieved by comparing the two methods defined above for calculation of depleted uranium fraction. Because some of the sample points are just outside the bounds of the defined field for mixing between the end-member compositions, the alternative projection approach (Figure 5) gives depleted uranium fractions that are up to 10% negative at the 0% fraction limit and up to 20% lower fractions at the 100% fraction limit. This is due to uncertainties in the analytical results (which are not evaluated by these calculations) and real ranges in the compositions of depleted and highly enriched uranium “end-member” materials produced by DOE and used at RFETS (see particularly the apparent range in $^{236}\text{U}/^{238}\text{U}$ around the nominal depleted uranium end-member composition plotted in Figure 2 indicated by the sub-horizontal cluster of samples).

Lastly, the calculations of end-member fractions from isotopic ratio data for all actual samples from RFETS were qualitatively evaluated by plotting samples on axes structured following Figure 2, with sample point (bubble) sizes scaled to the fraction of an end-member (Figure 6). Inspection of these three diagrams reveals that the gradient of bubble sizes is thoroughly consistent with trends expected across the compositional ranges.

Use & Results

Six pieces of data are required for each sample to apply all functionality of the spreadsheet calculations and plots. For fraction of end-member calculations, data must be entered for the ratios $^{235}\text{U}/^{238}\text{U}$ and $^{236}\text{U}/^{238}\text{U}$. Geographic plots require entry of Northing and Easting location data for each sample. Additional data that is shown on spreadsheet plots is uranium concentration (ug/L) and the ratio $^{234}\text{U}/^{238}\text{U}$. Data for each sample is entered into a row in the U_Data_Summary worksheet.

Geographic plots for the data from RFETS water samples analyzed for isotopic composition are presented in Figure 7(a-h). The highest fractions of highly enriched uranium (between 0.2% and 1.0%) are found in the area of the Solar Ponds and for one sample in the center of the Industrial Area. Other samples from the Walnut Creek drainage, the Industrial Area and the Original Landfill area are found to contain 0.05% to 0.2% fraction of highly enriched uranium. Depleted uranium is more broadly distributed within the Industrial Area and at fractions between 0.5% and 100%. In addition, samples with substantial fractions (>0.5%) of depleted uranium are found across the Walnut Creek and Woman Creek drainages. Below (south) of the 903 Pad site a sample location gave consistently high depleted uranium fractions and concentrations of 270-300 ug/L. Similarly, in the area of the Original Landfill one sample location consistently gave >90% depleted uranium fraction and concentrations between 350 and 750 ug/L.

Most high concentrations of uranium in water samples correspond to areas with the highest fractions of depleted uranium. The highest concentration sample (3000 ug/L) has calculated fractions of 82% and 0.6% depleted and highly enriched uranium, respectively. Samples with concentrations between 250 and 1000 ug/L are distributed between fractions of depleted uranium of ~100% (4 samples), ~50% (4 samples) and 0% (5 samples). In these samples, highly enriched uranium fraction is approximately

0.5% in the 5 samples with ~50% depleted uranium fraction and lower than 0.13% in samples with ~100% depleted uranium fraction. One of the samples with 0% depleted uranium fraction contained 0.03% highly enriched uranium fraction, while another is the natural uranium sample with high total concentration from the Rock Creek drainage at the northern edge of the site. Similar distributions are found at lower concentrations, with higher numbers of samples not containing depleted or highly enriched uranium.

Conclusions

A spreadsheet based computational approach has been developed and tested for determining and graphically displaying the fractional distribution of uranium end-members (natural, depleted and highly enriched) from isotopic analytical results for water samples. The approach was specifically verified and validated for the range of compositions observed for Rocky Flats (0-1% highly enriched uranium fraction, 0-100% depleted uranium fraction, and 0-100% natural uranium fraction).

Consistent with previous documentation of uranium operations and contamination (Kaiser-Hill LLC, 2002), only very small amounts of highly enriched uranium are found in a small number of water samples focused in the former Solar Ponds complex and central Industrial Area. Depleted uranium is more widely distributed and samples contain a relatively complete set of mixtures with natural uranium (Figure 5). However, one third of the samples are found to contain no depleted or highly enriched uranium component and three quarters of the samples are found to contain more than 90% natural uranium – substantial fractions given that the focus of these analyses was on evaluating potentially contaminated waters.

References

Kaiser-Hill LLC (2002) Actinide Migration Evaluation Pathway Analysis Summary Report, *Kaiser-Hill Company, US Department of Energy Rocky Flats Environmental Technology Site, Golden, CO*, 33p. (& Technical Appendices)

Table 1. Isotopic compositions and ratios for three nominal end-members of Uranium

Atom %	²³⁴ U	²³⁵ U	²³⁶ U	²³⁸ U
Natural (U)	0.0055	0.720	0.0000	99.280
Depleted (DU)	0.0009	0.190	0.00359	99.807
Highly enriched (HEU)	1.064	93.113	0.4975	5.321

	²³⁴ U/ ²³⁸ U	²³⁵ U/ ²³⁸ U	²³⁶ U/ ²³⁸ U
Natural (U)	0.000055	0.007253	0.000
Depleted (DU)	0.0000095	0.00190	0.000036
Highly enriched (HEU)	0.2	17.5	0.09352

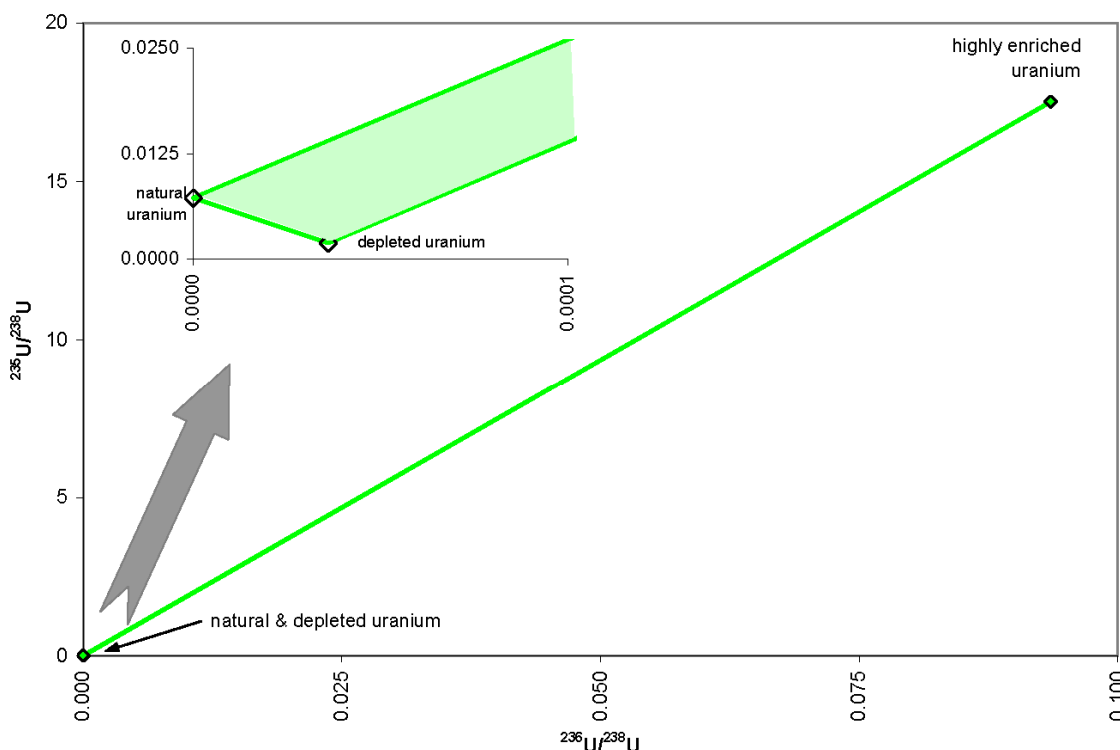


Figure 1. Triangular area of mixing (green shaded field) between natural, depleted and highly enriched uranium within the isotopic ratio plot for ²³⁶U, ²³⁵U relative to ²³⁸U. The triangular area of mixing has a very short base between natural and depleted compositions, with nearly equivalent sides to the highly enriched composition. Samples from RFETS are found in the lower left of the mixing area, very close to natural and depleted end-member compositions (Figure 2).

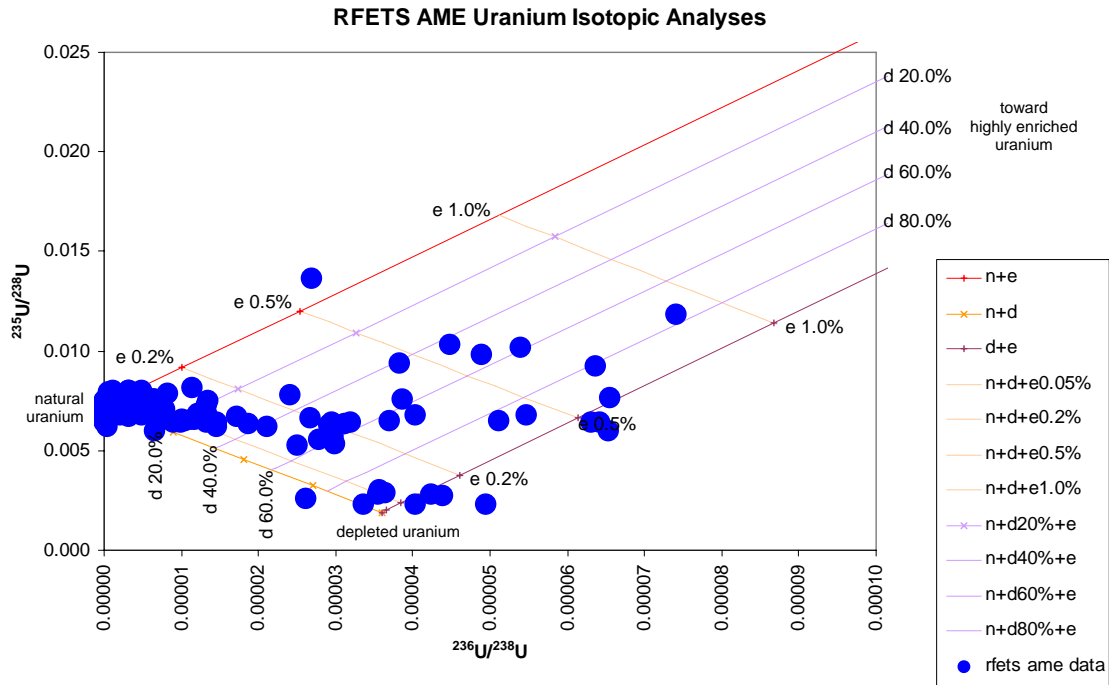


Figure 2. Sample set of uranium isotopic analyses from RFETS in ratio format. The lines contain the area of mixing between the end-member compositions (Table 1 and Figure 1), with natural uranium composition at the left apex, depleted uranium at the lower, central apex, and highly enriched uranium well to the right, upper corner, of the frame. Calculated trends for synthetic intermediate mixtures with specified fixed amounts of highly enriched (0.05, 0.2, 0.5, and 1.0%) or depleted (20, 40, 60, and 80%) uranium are shown with intermediate lines. Comparison of the RFETS sample data with these mixing lines shows that the maximum fraction of highly enriched uranium end-member is less than 1.0%.

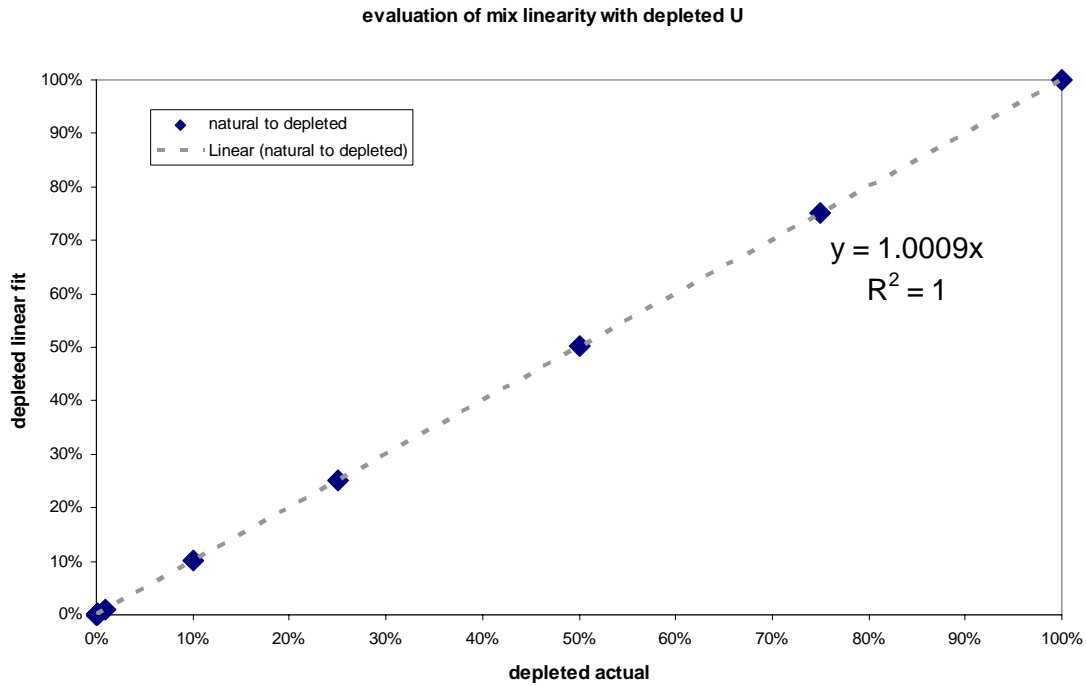


Figure 3. Evaluation of mixing trend from natural to depleted uranium compositions (Table 2, second set). The figure shows the accuracy of a linear fit across the entire mixing range from natural to depleted uranium by comparing results for specific fractional mixes (diamond symbols) with a linear fit (dashed gray line), and the calculated equation of the fit line and measure of goodness of fit (R^2). Therefore, the shifts found on atom ratio plots (e.g. Figures 1 & 2) are linear between these end-member compositions.

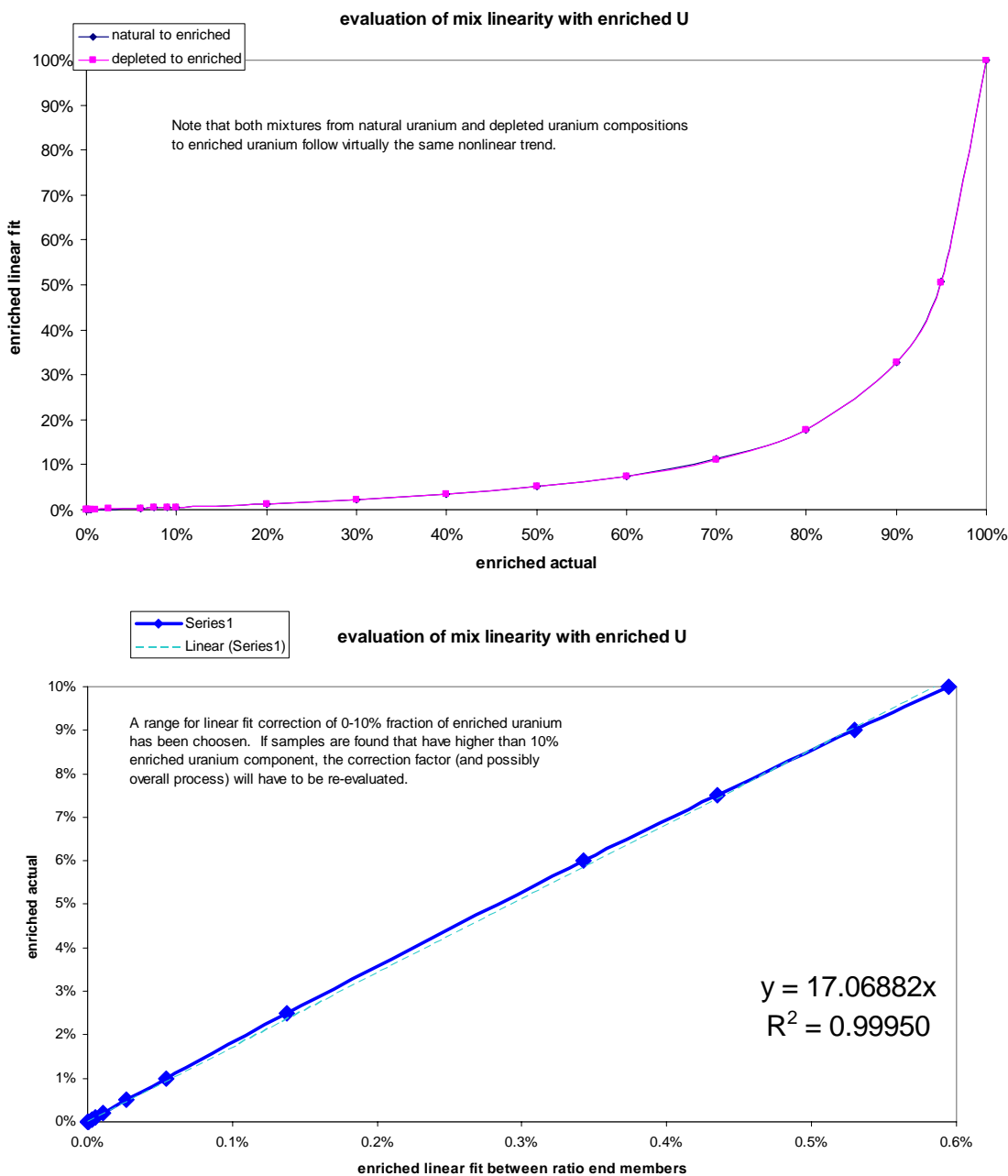


Figure 4. Evaluation of mixing trend from 100% natural (or 100% depleted) to 100% highly enriched uranium compositions. Estimated fraction of highly enriched uranium is calculated from isotopic ratio compositions, and compared on the plots to synthetic mix compositions. Because the compositions are mixtures of multiple isotopes, the shifts found on the atom ratio plot (e.g. Figure 1) are nonlinear. The upper plot demonstrates that mixing from natural and depleted end-members are very similar. The lower figure shows the accuracy of a linear fit between 0% and 10% highly enriched uranium is very good, with a factor of 17.07.

Table 2. Evaluation calculations for fractions of end-members for synthetic mixture compositions.

	synthetic ratios		% fractions calculated		
	$^{236}\text{U}/^{238}\text{U}$	$^{235}\text{U}/^{238}\text{U}$	depleted	enriched	natural
QC checks					
Set 1: natural plus					
e0.00%	0.000000	0.007253			
e0.01%	0.000001	0.007348	0.00%	0.0092%	99.9902%
e0.05%	0.000003	0.007727	0.00%	0.0461%	99.9510%
e0.10%	0.000005	0.008201	0.01%	0.0923%	99.9019%
e0.20%	0.000010	0.009151	0.01%	0.1848%	99.8035%
e0.50%	0.000025	0.012010	0.03%	0.4633%	99.5074%
e6.00%	0.000322	0.067489	0.37%	5.8664%	93.7634%
e7.50%	0.000409	0.083698	0.47%	7.4450%	92.0852%
e9.00%	0.000498	0.100410	0.57%	9.0725%	90.3550%
e10.00%	0.000559	0.111842	0.64%	10.1859%	89.1713%
e20.00%	0.001249	0.240832	1.44%	22.7482%	75.8162%
e30.00%	0.002121	0.403891	2.44%	38.6284%	58.9338%
e40.00%	0.003258	0.616569	3.74%	59.3412%	36.9139%
e50.00%	0.004803	0.905582	5.52%	87.4881%	6.9907%
e60.00%	0.007024	1.321013	8.07%	127.9468%	-36.0212%
e70.00%	0.010488	1.969027	12.06%	191.0567%	-103.1139%
e80.00%	0.016646	3.120875	19.14%	303.2439%	-222.3715%
e90.00%	0.030637	5.737875	35.22%	558.1039%	-493.3347%
e100.00%	0.093520	17.500000	107.51%	1703.6147%	-1711.1262%
Set 2: natural plus					
d0.00%	0.000000	0.007253			
d0.01%	0.000000	0.007252	0.01%	0.0000%	99.9899%
d0.05%	0.000000	0.007250	0.05%	0.0000%	99.9497%
d0.10%	0.000000	0.007248	0.10%	0.0000%	99.8995%
d0.20%	0.000000	0.007242	0.20%	0.0000%	99.7989%
d1.00%	0.000000	0.007199	1.01%	0.0000%	98.9947%
d10.00%	0.000004	0.006715	10.05%	0.0000%	89.9523%
d25.00%	0.000009	0.005909	25.10%	0.0000%	74.9006%
d50.00%	0.000018	0.004569	50.13%	0.0000%	49.8677%
d75.00%	0.000027	0.003233	75.10%	0.0000%	24.9009%
d100.00%	0.000036	0.001900	100.00%	0.0000%	0.0000%

Notes:

Orange highlighted cells indicate calculations beyond the range of the linear fit developed in Figure 4. Pink highlighted cells indicate calculation results that are negative and beyond the specified accuracy target of 2%.

Yellow highlighted cells indicate calculation results for fraction natural uranium that are less than or equal to 0%, but within the 2% accuracy target specified.

Green highlighted cells indicate calculation results where the fraction of natural and depleted uranium is within the specified accuracy target of 2% and the fraction of highly-enriched uranium within the linear fit range.

Table 2 (continued). Evaluation calculations for fractions of end-members for synthetic mixture compositions.

	synthetic ratios		% fractions calculated		
	$^{236}\text{U}/^{238}\text{U}$	$^{235}\text{U}/^{238}\text{U}$	depleted	enriched	natural
Set 3: depleted plus					
e0.00%	0.000036	0.001900	100.00%	0.0000%	0.0000%
e0.010%	0.000037	0.001994	100.00%	0.0092%	-0.0092%
e0.050%	0.000039	0.002371	100.00%	0.0459%	-0.0461%
e0.100%	0.000041	0.002843	100.00%	0.0918%	-0.0922%
e0.200%	0.000046	0.003788	100.00%	0.1838%	-0.1846%
e0.500%	0.000061	0.006634	100.00%	0.4609%	-0.4629%
e1.00%	0.000087	0.011412	100.00%	0.9261%	-0.9302%
e2.50%	0.000165	0.026027	100.01%	2.3490%	-2.3594%
e5.000%	0.000300	0.051352	100.02%	4.8146%	-4.8359%
e6.000%	0.000356	0.061838	100.03%	5.8355%	-5.8612%
e7.500%	0.000442	0.077966	100.03%	7.4058%	-7.4385%
e9.000%	0.000531	0.094596	100.04%	9.0249%	-9.0647%
e10.000%	0.000592	0.105972	100.04%	10.1324%	-10.1771%
e20.000%	0.001278	0.234333	100.10%	22.6297%	-22.7295%
e30.000%	0.002145	0.396612	100.17%	38.4291%	-38.5986%
e40.000%	0.003276	0.608297	100.26%	59.0388%	-59.2991%
e50.000%	0.004813	0.896003	100.38%	87.0498%	-87.4337%
e60.000%	0.007023	1.309643	100.56%	127.3219%	-127.8832%
e70.000%	0.010471	1.955071	100.84%	190.1607%	-190.1607%
e80.000%	0.016603	3.102947	101.33%	301.9179%	-303.2491%
e90.000%	0.030552	5.713893	102.45%	556.1196%	-558.5716%
e100.000%	0.093520	17.500000	107.51%	1703.6147%	-1711.1262%
Set 4: natural plus 20% depleted plus					
e0.000%	0.000007	0.006178	20.08%	0.0000%	79.9152%
e0.010%	0.000008	0.006272	20.09%	0.0092%	79.9035%
e0.05%	0.000010	0.006651	20.10%	0.0461%	79.8567%
e0.100%	0.000012	0.007124	20.11%	0.0922%	79.7982%
e0.200%	0.000017	0.008071	20.13%	0.1846%	79.6809%
e0.500%	0.000033	0.010925	20.21%	0.4628%	79.3278%
e6.000%	0.000329	0.066282	21.66%	5.8598%	72.4785%
e7.500%	0.000416	0.082454	22.09%	7.4365%	70.4776%
e9.000%	0.000505	0.099127	22.52%	9.0620%	68.4146%
e10.000%	0.000566	0.110532	22.82%	10.1739%	67.0034%

Notes:

Orange highlighted cells indicate calculations beyond the range of the linear fit developed in Figure 4. Pink highlighted cells indicate calculation results that are negative and beyond the specified accuracy target of 2%.

Yellow highlighted cells indicate calculation results for fraction natural uranium that are less than or equal to 0%, but within the 2% accuracy target specified.

Green highlighted cells indicate calculation results where the fraction of natural and depleted uranium is within the specified accuracy target of 2% and the fraction of highly-enriched uranium within the linear fit range.

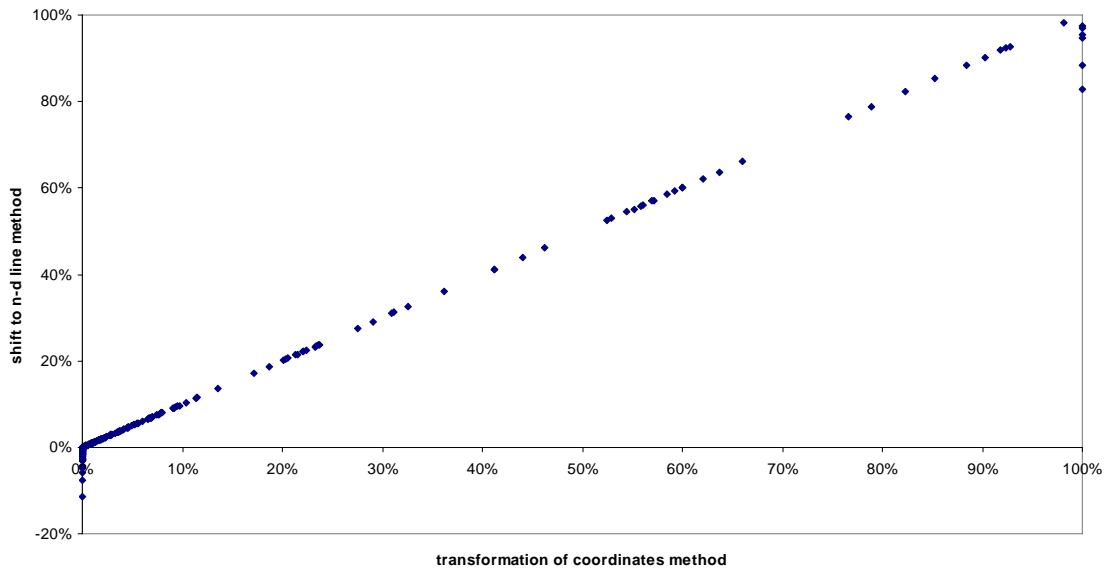


Figure 5. Comparison in RFETS sample data for fraction calculated of depleted uranium end-member in mixtures with natural uranium end-member for the two independent approaches to geometric transformation as described in text. The two methods give effectively identical results, except for samples at extremes of the mix area, where the transformation calculations have limits of 0-100% fractions enforced and the shift to natural-depleted mix line calculations do not have such real, physical limits enforced.

Quantitative Evaluation of Mixture Components in RFETS Uranium Isotopic Analyses
D.R. Janecky

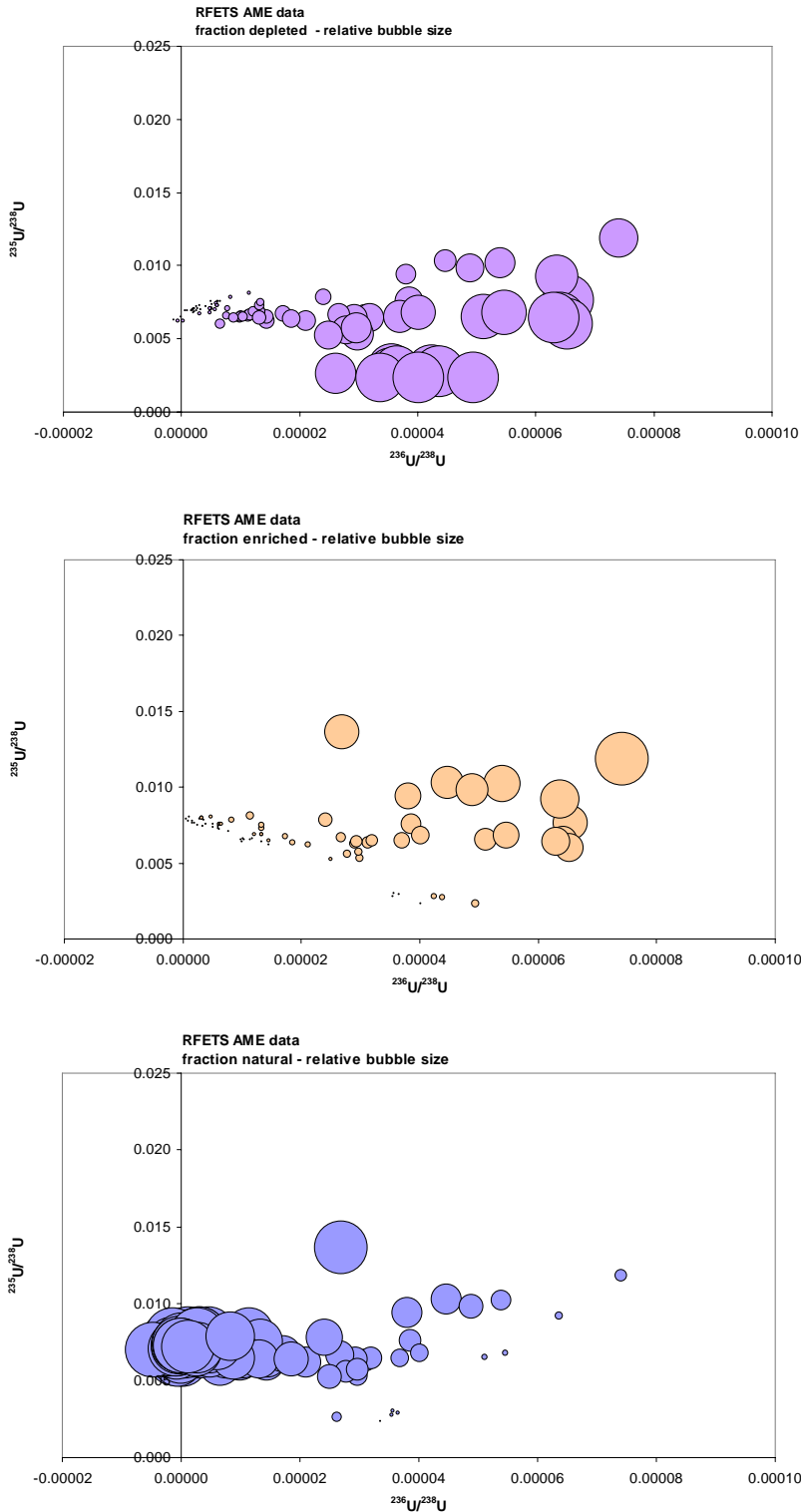


Figure 6. Qualitative evaluation of consistency in fraction evaluation results within the plot framework of Figure 2. Bubble size is proportional to the amount of each respective component calculated.

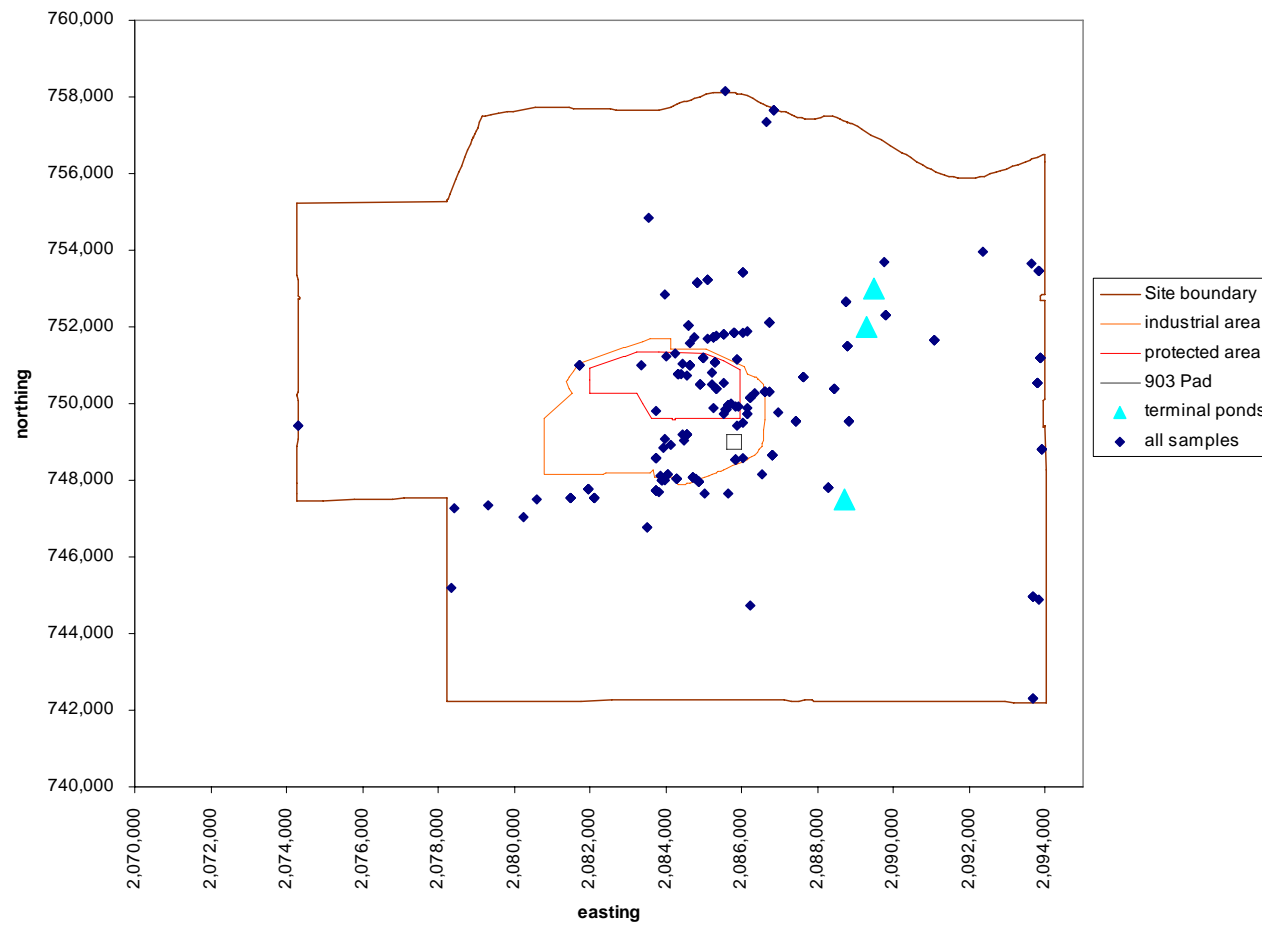


Figure 7a. Distribution of water samples analyzed for uranium isotopic composition by HR-ICP/MS or TIMS.

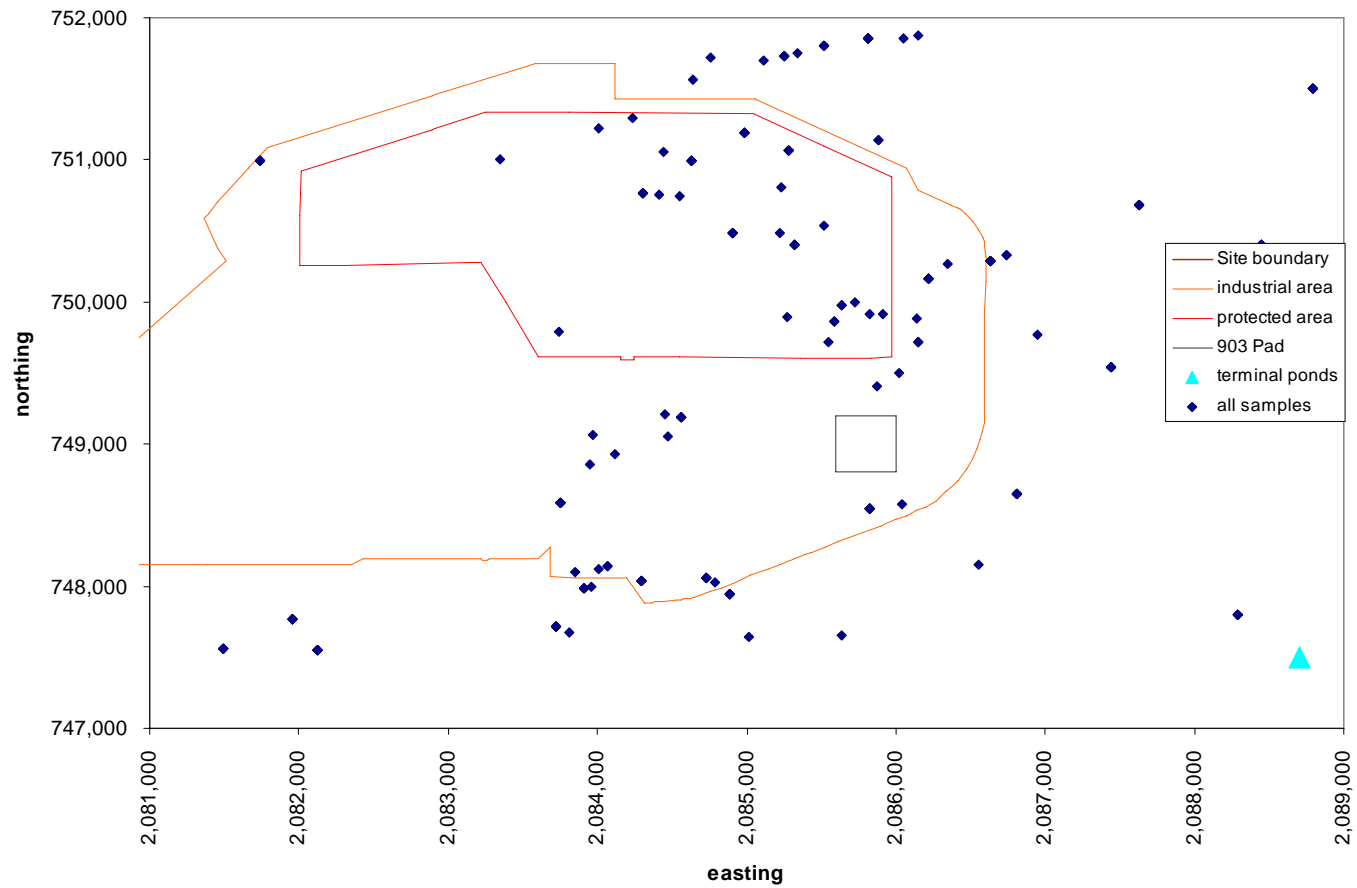


Figure 7b. Distribution of water samples analyzed for uranium isotopic composition by HR-ICP/MS or TIMS, focused on the Industrial Area.

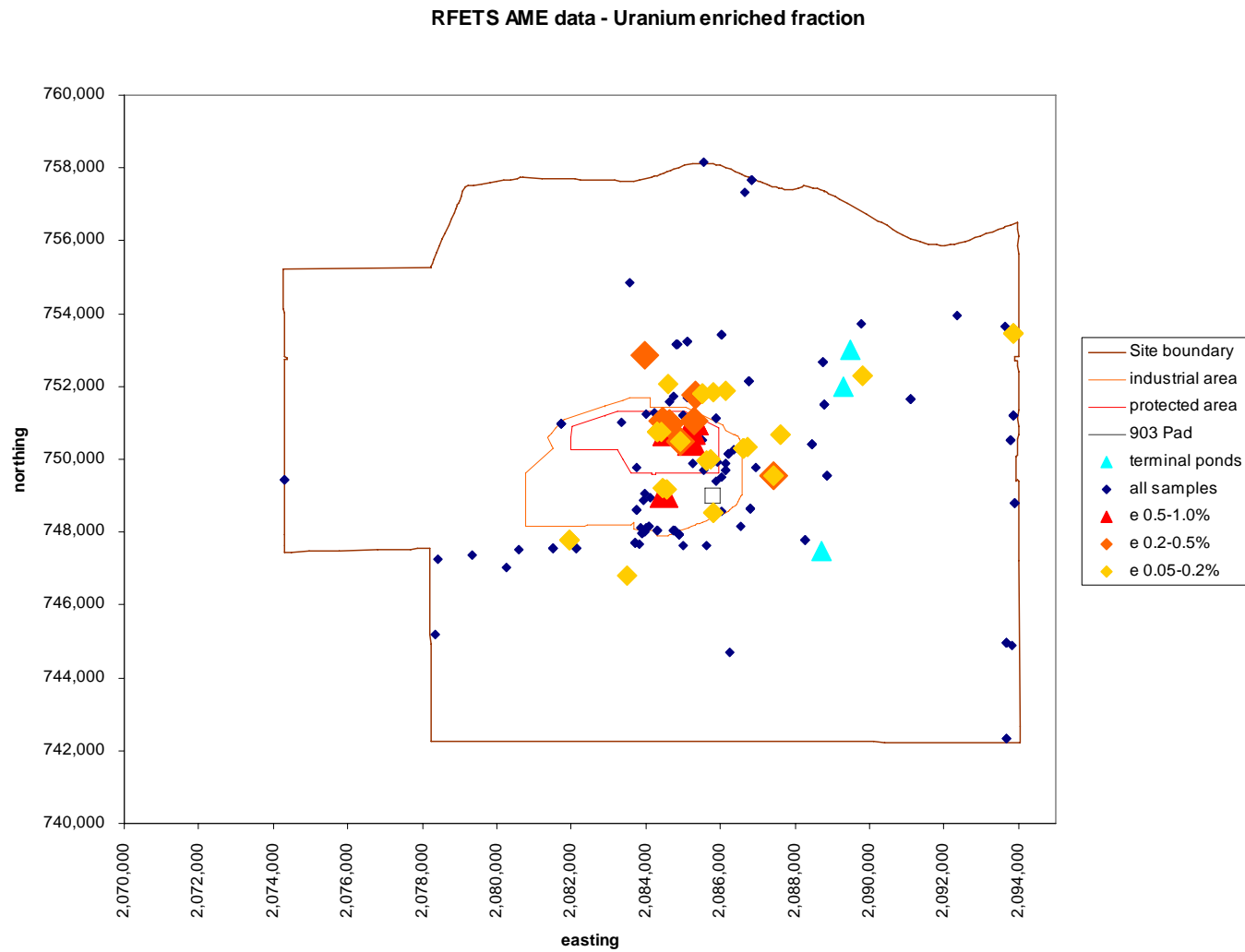


Figure 7c. Distribution of highly enriched uranium fraction geographically as a function of percent ranges.

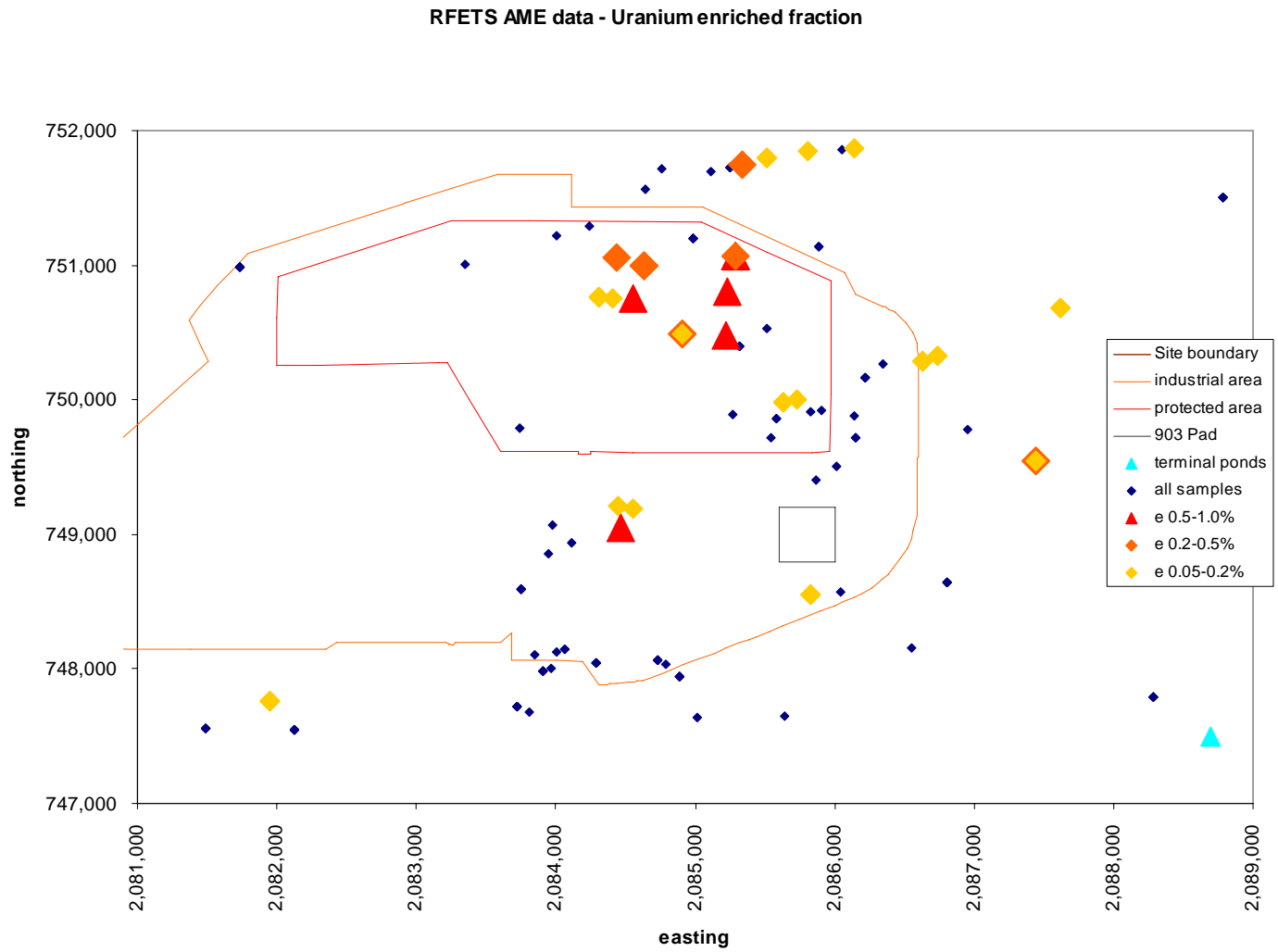


Figure 7d. Distribution of highly enriched uranium fraction geographically within the Industrial Area as a function of percent ranges.

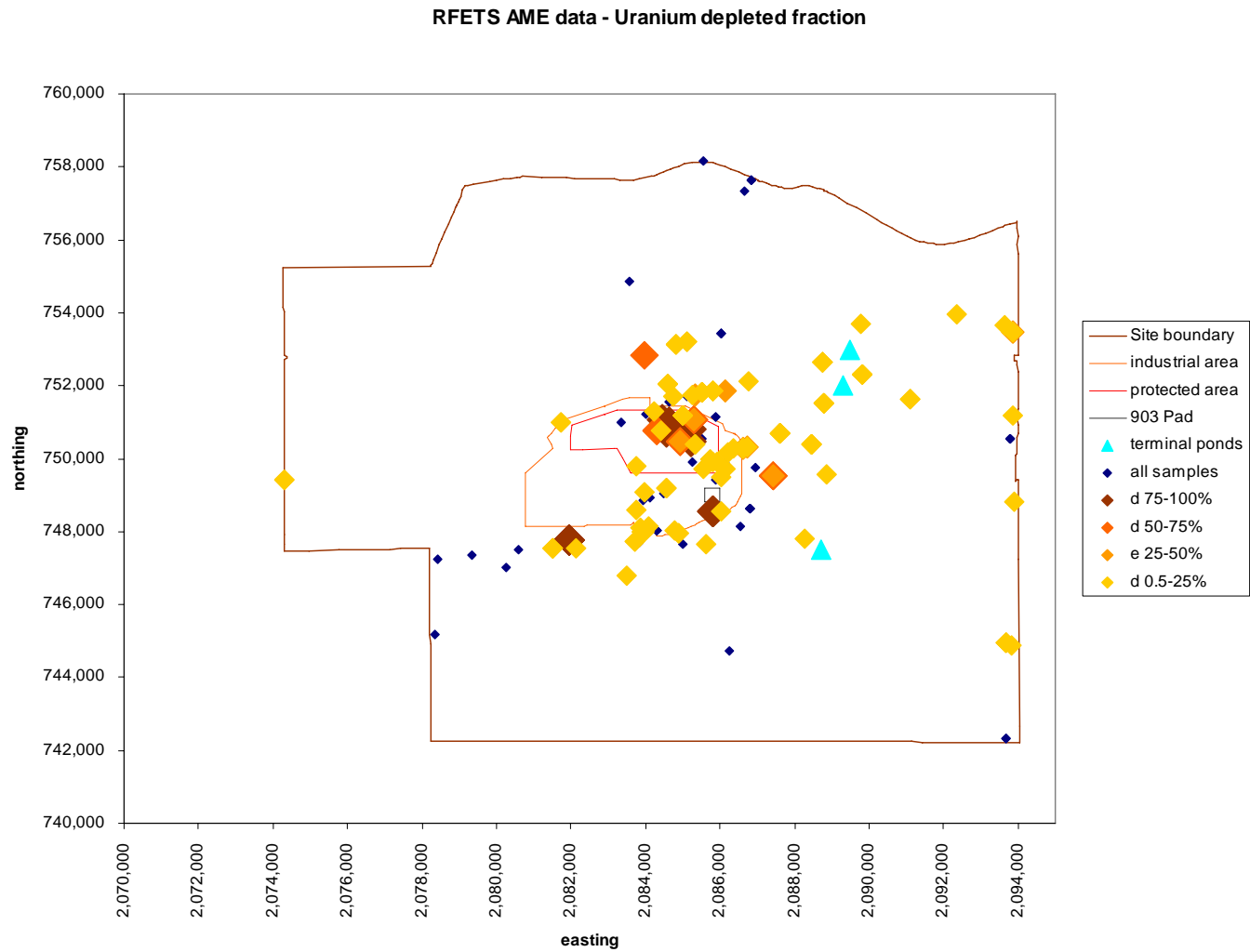


Figure 7e. Distribution of depleted uranium fraction geographically as a function of percent ranges.

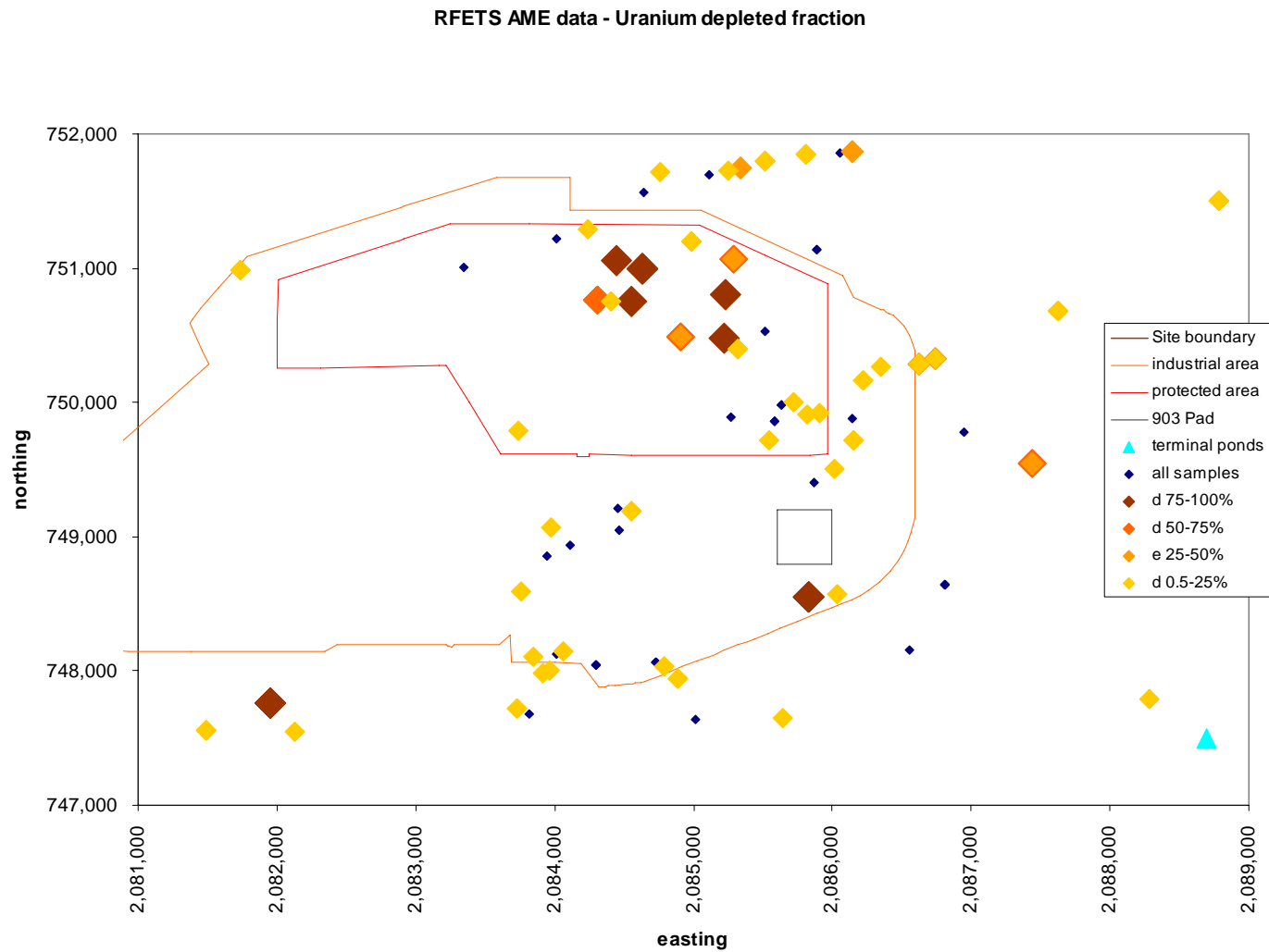


Figure 7f. Distribution of depleted uranium fraction geographically focused on the Industrial Area as a function of percent ranges.

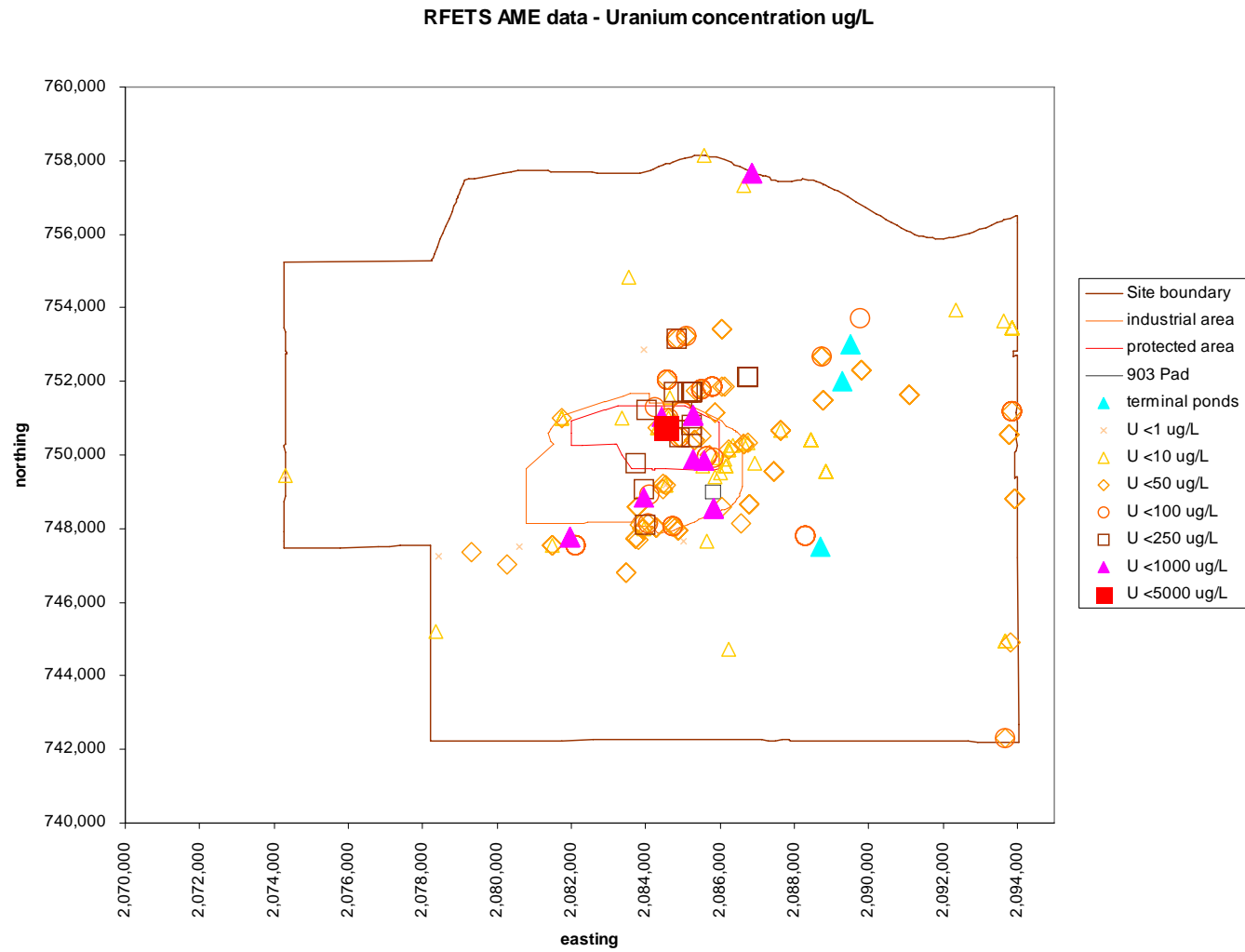


Figure 7g. Distribution of uranium concentrations geographically as a function of ug/L fractions.

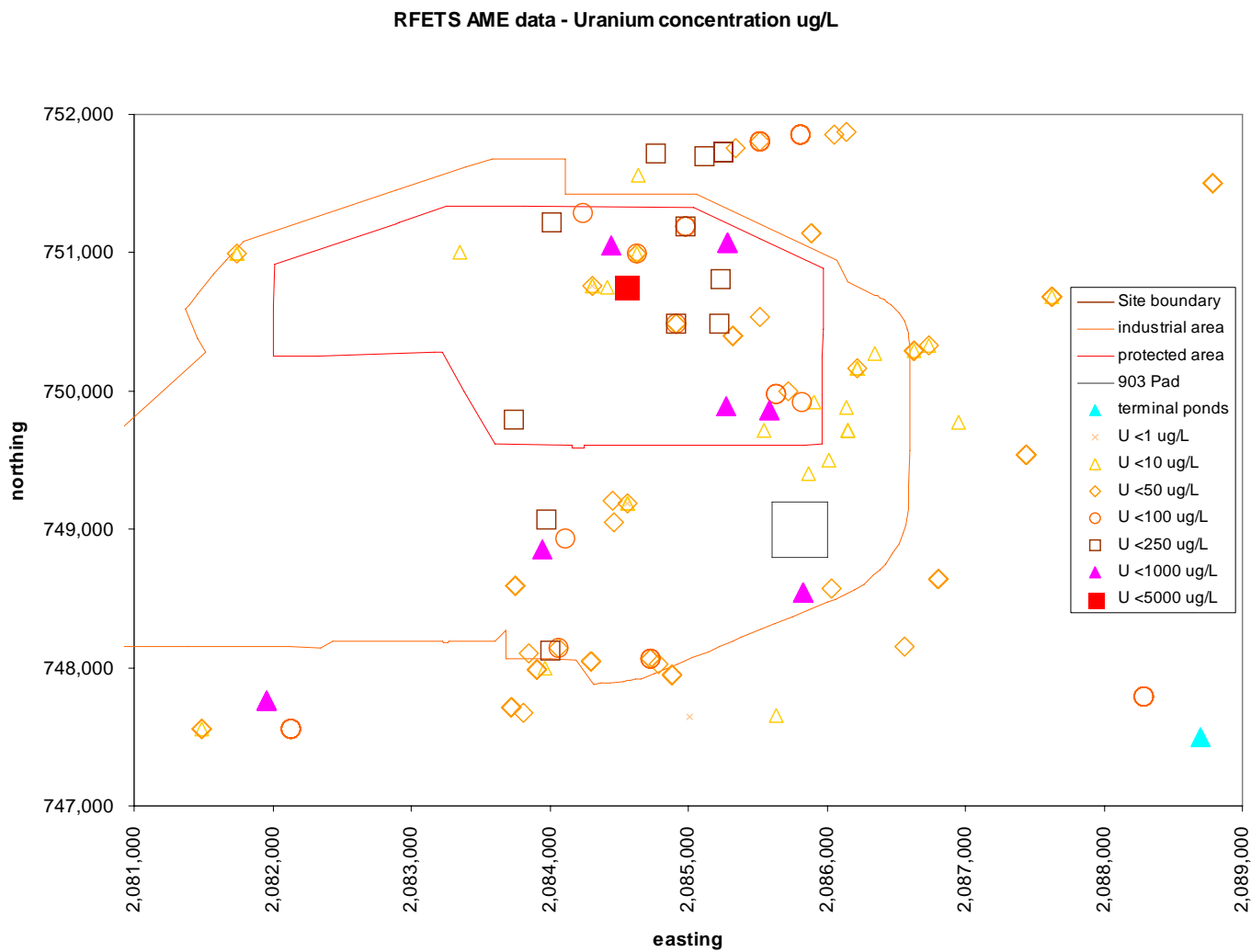


Figure 7h. Distribution of uranium concentrations geographically focused on the Industrial Area as a function of ug/L ranges.

Table 3. Summary of Uranium isotopic data and calculated % fractions of end-members (attached printout on following pages)

Samples with reported ^{236}U concentrations of below detection limits were assigned $^{236}\text{U}/^{238}\text{U}$ ratios of 1×10^{-11} and the cells highlighted in pink.

Table 3. Summary of Uranium isotopic data and calculated % fractions of end-members.

13-133	59793	2082128.1	747552.6	8/1/2000	75.8	6.5970E-05	7.2294E-03	-1.1210E-06	0.0%	0.00%	100.0%
21-97	59793	2082128.1	747552.6	12/2/1999	73.1	7.9290E-05	7.3581E-03	7.1560E-08	0.0%	0.01%	100.0%
8-133	59793	2082128.1	747552.6	2/10/2000	75.2	6.2190E-05	7.0567E-03	1.0000E-11	1.6%	0.00%	98.4%
16-203	59793	2082128.1	747552.6	6/12/2000	68.5	6.1530E-05	7.2663E-03	-5.4230E-07	0.0%	0.00%	100.0%
16-248	59793	2082128.1	747552.6	6/20/2002	57.3	7.0443E-05	7.1308E-03	-8.4988E-07	0.0%	0.00%	100.0%
15-240	60699	2083741.2	749790.1	6/18/2002	121.9	7.6544E-05	7.2783E-03	1.3131E-06	1.8%	0.01%	98.2%
20-40	61093	2081952.0	747764.3	6/28/1999	346.7	9.2360E-05	2.7260E-03	4.3760E-05	100.0%	0.11%	-0.1%
21-98	61093	2081952.0	747764.3	12/7/1999	766.5	1.4280E-05	2.3333E-03	4.9380E-05	100.0%	0.13%	-0.1%
8-134	61093	2081952.0	747764.3	2/17/2000	769.5	1.0950E-05	2.3305E-03	3.3630E-05	92.8%	0.00%	7.2%
16-204	61093	2081952.0	747764.3	5/3/2000	523.5	9.4860E-06	2.3199E-03	4.0220E-05	100.0%	0.06%	-0.1%
15-238	70099	2084761.0	751716.0	6/19/2002	222.0	7.7929E-05	7.2682E-03	4.4644E-07	0.6%	0.00%	99.4%
305	71394	2080265.5	747027.0	2/15/2005	11.8	8.1200E-05	7.2939E-03	1.0000E-11	0.0%	0.00%	100.0%
20-41	75292	2089809.0	752305.0	6/24/1999	29.3	7.8190E-05	7.4290E-03	5.9280E-06	7.7%	0.06%	92.2%
22-99	75292	2089809.0	752305.0	12/2/1999	33.1	7.7310E-05	7.5740E-03	5.0520E-06	5.2%	0.06%	94.8%
8-138	75292	2089809.0	752305.0	2/9/2000	22.3	8.6790E-05	7.3262E-03	5.2000E-06	7.4%	0.05%	92.5%
16-208	75292	2089809.0	752305.0	6/12/2000	37.4	8.4620E-05	7.2244E-03	1.7320E-06	2.9%	0.01%	97.1%
20-42	75992	2086628.0	750290.0	7/27/1999	31.0	7.4960E-05	6.8596E-03	1.3240E-05	23.7%	0.09%	76.2%
22-100	75992	2086628.0	750290.0	12/2/1999	21.9	5.9610E-05	6.6034E-03	1.1670E-05	23.4%	0.06%	76.5%
11-167	75992	2086628.0	750290.0	1/24/2000	20.1	7.1970E-05	6.8831E-03	1.3290E-05	23.6%	0.09%	76.3%
16-209	75992	2086628.0	750290.0	6/12/2000	22.8	6.7480E-05	6.8539E-03	1.2150E-05	22.1%	0.08%	77.8%
14-222	75992	2086628.0	750290.0	6/10/2002	8.9	7.9531E-05	6.7401E-03	1.7245E-05	30.9%	0.11%	69.0%
300	79102	2084441.4	751052.8	6/22/2004	801.0	5.7900E-05	6.4475E-03	6.4200E-05	100.0%	0.47%	-0.5%
16-243	83201	2083947.1	748857.2	6/18/2002	403.0	7.1485E-05	7.2054E-03	6.6298E-08	0.5%	0.00%	99.5%
17-259	86501	2083972.1	749066.9	7/10/2002	122.8	7.3418E-05	7.2698E-03	7.4316E-07	1.0%	0.01%	99.0%
17-258	86701	2084115.2	748933.6	7/10/2002	91.1	6.6595E-05	7.2681E-03	9.1827E-09	0.0%	0.00%	100.0%
16-242	88101	2084009.0	748122.6	6/18/2002	148.1	8.7496E-05	7.2912E-03	7.1425E-08	0.0%	0.00%	100.0%
5-244	90099	2086556.6	748154.1	3/8/2001	10.2	7.3289E-05	7.3231E-03	1.1519E-07	0.0%	0.00%	100.0%
298	99101	2085272.8	749895.3	6/29/2004	397.0	7.7000E-05	7.2516E-03	1.0000E-11	0.0%	0.00%	100.0%
?	99305	2085632.9	5939.5	6/27/2005		7.8100E-05	8.0026E-03	3.2000E-06	0.0%	0.07%	99.9%
17-255	99401	2085586.3	749861.8	7/10/2002	652.7	7.7947E-05	7.3758E-03	-2.4484E-07	0.0%	0.00%	100.0%
N BKGR	B201589	2086648.3	757328.4	4/1/1998	2.1	6.0900E-05	7.3300E-03	-1.8800E-07	0.0%	0.00%	100.0%
ROCK CR	B203189	2083556.5	754848.2	4/1/1998	3.9	9.6300E-05	7.9100E-03	-1.4600E-06	0.0%	0.02%	100.0%
BKGR	B205589	2086855.2	757654.1	4/1/1998	294.1	7.0400E-05	7.1500E-03	-1.4000E-06	0.0%	0.00%	100.0%
BKGR	B205589	2086855.2	757654.1	6/24/1999	286.0	1.7410E-05	7.5774E-03	1.3830E-06	0.0%	0.03%	100.0%
BKGR	B205589	2086855.2	757654.1	2/9/2000	287.1	8.4550E-05	7.3888E-03	-2.2650E-07	0.0%	0.01%	100.0%
PL	B206989	2084835.2	753145.2	12/13/2000	104.5	9.0332E-05	7.3673E-03	-4.5091E-07	0.0%	0.00%	100.0%
14-229	B208189	2085885.1	751138.0	DATE UNKN	27.1	9.1002E-05	7.5186E-03	6.3615E-07	0.0%	0.02%	100.0%
14-229D	B208189	2085885.1	751138.0	3/20/2002	27.4	7.5320E-05	7.2706E-03	-8.4350E-07	0.0%	0.00%	100.0%
SEP	B208689	2085249.9	751727.9	4/1/1998	150.9	7.4600E-05	7.1700E-03	-1.2800E-06	0.0%	0.00%	100.0%
SEP	B208689	2085249.9	751727.9	6/21/1999	146.5	9.7580E-05	7.6026E-03	7.0330E-07	0.0%	0.02%	100.0%
SEP	B208689	2085249.9	751727.9	2/10/2000	144.1	8.7950E-05	7.3402E-03	-1.3290E-06	0.0%	0.00%	100.0%
SEP	B208689	2085249.9	751727.9	4/18/2000	133.6	9.6510E-05	7.3313E-03	1.0630E-06	1.0%	0.01%	99.0%
14-230	B210389	2085116.4	751695.8	3/20/2002	142.7	8.9406E-05	7.1364E-03	-4.0072E-07	0.3%	0.00%	99.7%
SEP	B210489	2085513.2	751801.9	4/1/1998	55.1	6.6500E-05	7.2200E-03	3.9900E-07	0.9%	0.00%	99.1%
20-48	B210489	2085513.2	751801.9	6/22/1999	73.4	8.0940E-05	7.5854E-03	6.2590E-06	6.9%	0.07%	93.0%
9-147	B210489	2085513.2	751801.9	1/31/2000	54.8	7.0240E-05	7.3041E-03	1.3240E-05	20.1%	0.11%	79.8%
18-214	B210489	2085513.2	751801.9	4/18/2000	46.1	7.7010E-05	7.4979E-03	1.3370E-05	18.7%	0.12%	81.2%
20-44	B302089	2083490.5	746786.3	6/28/1999	42.8	8.9420E-05	7.8417E-03	3.4910E-06	0.5%	0.06%	99.4%
9-148	B302089	2083490.5	746786.3	2/14/2000	44.5	8.5640E-05	7.0774E-03	-1.8940E-06	0.0%	0.00%	100.0%
WMN CR	B305389	2086232.0	744718.0	4/1/1998	8.7	6.6100E-05	7.2900E-03	2.4700E-07	0.1%	0.00%	99.9%
W BKGR	B405489	2078357.0	745190.8	4/1/1998	2.4	1.2800E-04	7.1700E-03	-7.2400E-07	0.0%	0.00%	100.0%
514-001	GS01	2093819.9	744894.4	5/1/2002	10.3	7.1052E-05	7.0822E-03	2.0622E-06	4.6%	0.01%	95.4%
514-002	GS03	2093622.1	753639.9	5/1/2002	2.3	6.1873E-05	6.5107E-03	9.9071E-06	21.5%	0.04%	78.5%
514-003	GS04	2085567.9	758144.8	5/1/2002	1.8	6.7613E-05	7.2059E-03	-1.1662E-08	0.4%	0.00%	99.6%
514-004	GS05	2078428.0	747260.2	5/1/2002	0.2	6.0758E-05	6.9733E-03	-4.6507E-06	0.0%	0.00%	100.0%
514-005	GS10	2086741.0	750326.0	5/1/2002	9.6	6.0976E-05	6.5283E-03	1.0415E-05	22.1%	0.04%	77.8%
514-006	GS13	2086144.8	751871.7	5/1/2002	16.0	6.1095E-05	6.4138E-03	1.3277E-05	27.5%	0.06%	72.4%
514-007	GS17	2085638.2	747649.6	5/1/2002	2.7	7.1119E-05	7.1398E-03	1.2547E-06	2.9%	0.00%	97.1%
20-45	P114389	2081738.8	750990.4	7/21/1999	6.4	7.6690E-05	7.0545E-03	4.8160E-06	9.1%	0.03%	90.9%
22-105	P114389	2081738.8	750990.4	12/8/1999	10.1	7.1300E-05	7.0892E-03	-8.4900E-07	0.0%	0.00%	100.0%
11-168	P114389	2081738.8	750990.4	1/26/2000	6.2	8.4880E-05	7.1727E-03	-3.2030E-07	0.2%	0.00%	99.8%
18-215	P114389	2081738.8	750990.4	4/18/2000	8.8	8.5560E-05	7.2236E-03	6.1830E-07	1.2%	0.00%	98.8%
22-106	P207689	2085318.0	750398.0	12/2/1999	31.6	6.8970E-05	7.4053E-03	1.8590E-06	1.6%	0.02%	98.4%
9-149	P207689	2085318.0	750398.0	2/1/2000	29.6	7.6270E-05	7.3178E-03	-3.9860E-07	0.0%	0.00%	100.0%
18-216	P207689	2085318.0	750398.0	5/4/2000	31.8	7.9090E-05	7.1412E-03	1.8820E-06	3.8%	0.01%	96.2%
SEP	P209189	2084309.0	750762.0	4/1/1998	13.2	3.9500E-05	5.3300E-03	2.9800E-05	62.0%	0.14%	37.8%
22-107	P209189	2084309.0	750762.0	12/15/1999	8.8	4.7650E-05	5.5832E-03	2.7860E-05	56.9%	0.13%	42.9%
9-150	P209189	2084309.0	750762.0	2/1/2000	0.5	4.0210E-05	5.2600E-03	2.4970E-05	55.1%	0.09%	44.8%
18-217	P209189	2084309.0	750762.0	5/4/2000	9.9	4.6230E-05	5.7373E-03	2.9700E-05	58.5%	0.16%	41.3%
SEP	P209489	2084634.0	750991.0	4/1/1998	43.4	5.9100E-05	6.5300E-03	5.1200E-05	85.2%	0.37%	14.4%
16-8	P209489	2084634.0	750991.0	6/23/1999	45.5	5.7980E-05	6.7987E-03	5.4670E-05	88.4%	0.42%	11.2%
9-151	P209489	2084634.0	750991.0	1/31/2000	9.9	6.8200E-05	5.9941E-03	6.5310E-05	100.0%	0.46%	-0.5%
18-220	P209489	2084634.0	750991.0	5/4/2000	56.2	6.0730E-05	6.4262E-03	6.3040E-05	100.0%	0.46%	-0.5%
SEP	P209589	2085286.0	751071.0	4/1/1998	414.1	1.0300E-04	1.0300E-02	4.4700E-05	44.0%	0.53%	55.5%
22-108	P209589	2085286.0	751071.0	12/6/1999	475.9	1.0130E-04	1.0191E-02	5.3990E-05	59.2%	0.60%	40.2%
9-152	P209589	2085286.0	751071.0	2/28/2000	320.4	1.1200E-04	9.8251E-03	4.8900E-05	54.4%	0.53%	45.1%
18-218	P209589	2085286.0	751071.0	5/9/2000	321.8	7.2970E-05	9.4194E-03	3.8160E-05	41.1%	0.43%	58.4%
14-228	P209689	2085514.0	750533.0	3/25/2002	47.5	7.9728E-05	7.6218E-03	1.1923E-07	0.0%	0.02%	100.0%
SEP	P209889	2084984.0	751194.0	4/1/1998	102.3	7.8500E-05	7.3200E-03	1.1600E-06	1.2%	0.01%	98.7%

Table 3. Summary of Uranium isotopic data and calculated % fractions of end-members.

22-109	P209889	2084984.0	751194.0	12/20/1999	90.0	7.8660E-05	7.2241E-03	2.1330E-06	3.5%	0.02%	96.4%
9-153	P209889	2084984.0	751194.0	2/1/2000	91.2	7.6130E-05	7.0919E-03	2.1350E-06	4.6%	0.01%	95.4%
18-219	P209889	2084984.0	751194.0	5/9/2000	96.1	8.4110E-05	7.3643E-03	2.4850E-07	0.0%	0.01%	100.0%
303	P210089	2084639.3	751563.5	11/9/2004	9.0	8.7100E-05	7.2674E-03	7.0000E-09	0.0%	0.00%	100.0%
15-231	P210189	2084410.8	750752.2	3/26/2002	3.6	8.2604E-05	7.5554E-03	6.5292E-06	7.6%	0.07%	92.3%
299	P219189	2084010.0	751222.0	6/23/2004	117.0	7.5500E-05	7.2622E-03	1.0000E-11	0.0%	0.00%	100.0%
15-239	SPP DIS GALLERY	2085339.4	751751.4	6/18/2002	40.5	7.7564E-05	7.8281E-03	2.4123E-05	32.6%	0.23%	67.2%
514-009	STP	2086347.4	750266.9	DATE UNKN	9.9	6.4486E-05	6.4305E-03	8.8854E-06	20.6%	0.03%	79.4%
5-243A	SW097	2083957.0	752846.6	1/31/2001	0.3	7.0628E-05	6.7934E-03	4.0221E-05	66.0%	0.30%	33.7%
5-243B	SW13494	2083963.7	748000.2	4/1/2001	8.4	6.6626E-05	7.2401E-03	6.0990E-06	9.5%	0.05%	90.4%
514-008	SW18	2083350.6	751006.4	5/1/2002	5.2	8.0665E-05	7.4064E-03	-1.1208E-07	0.0%	0.01%	100.0%
									-	-	-
									-	-	-
99305	99305	2085632.9	749978.0	8/8/2005	53.7	7.8100E-05	8.0026E-03	3.2000E-06	0.0%	0.07%	99.9%
99305	99305	2085632.9	749978.0	8/8/2005	54.5	7.7600E-05	7.9051E-03	2.8000E-06	0.0%	0.06%	99.9%
99405	99405	2085584.8	749862.7	8/8/2005	395.7	7.4800E-05	7.2464E-03	1.0000E-11	0.1%	0.00%	99.9%
91203	91203	2085909.8	749919.0	8/8/2005	3.9	7.6200E-05	6.9444E-03	2.6000E-06	6.6%	0.00%	93.4%
91305	91305	2085822.7	749915.2	8/9/2005	54.1	7.6100E-05	7.0972E-03	2.8000E-06	5.6%	0.01%	94.4%
89104	89104	2085012.6	747640.6	8/18/2005	378.0	7.6800E-05	7.2307E-03	1.0000E-11	0.2%	0.00%	99.8%
GS10	GS10	2086741.0	750326.0	8/11/2005	13.2	5.7500E-05	6.3613E-03	1.8600E-05	36.2%	0.10%	63.7%
SW056	SW056	2085544.3	749713.2	8/2/2005	9.5	7.6400E-05	7.2098E-03	1.3000E-06	2.4%	0.01%	97.6%
SW141	SW141	2085725.0	749997.0	8/2/2005	16.3	7.6600E-05	7.8493E-03	8.3000E-06	7.9%	0.10%	92.0%