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**Elevated Concentrations of Primordial Radionuclides in Sediments from the
Reedy River and Surrounding Creeks in Simpsonville, South Carolina**

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10 **Abstract.** A gamma-ray survey and analysis of sixteen riverbed samples from the Reedy
River watershed near Simpsonville, SC were conducted and compared with national and
international studies of primordial radionuclides. The study reported here follows on a
recent discovery of anomalously high uranium concentrations in several private well
waters in the area near Simpsonville, SC. A HPGe spectrometer was used for
15 quantification of gamma emitting radionuclides in the sediments. All sediments contained
radionuclides from the uranium and thorium series as well as ⁴⁰K. Uranium-238
concentrations in sediment samples ranged from 11.1 to 74.2 Bq kg⁻¹. The measured
radionuclide concentrations were compared with data from UNSCEAR and NURE
reports. The river and stream sediment data were augmented by *in situ* NaI(Tl) gamma-
20 ray spectrometer measurements. Comparisons between the *ex-situ* and *in-situ*
measurements indicate equivalently distributed uranium in the surface soils and stream
sediments, the source of which is likely attributed to the monazite belts that are known to
exist in the area.

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5 1. Introduction

Uranium-238, ^{235}U , and ^{232}Th are the parents of the three natural decay series, called the uranium series, the actinium series and the thorium series, respectively. Each of these series consists of many daughter products generated through successive decay of parent radionuclides (Firestone, 1996). Natural uranium is a composite of the isotopes ^{238}U (99.28%), ^{234}U (0.0057%), and ^{235}U (0.72%). While on a mass basis there is far more ^{238}U than ^{235}U in a natural sample, the activity ratio is approximately 21:1. Many factors influence the aqueous concentration and mobility of a given radionuclide. Chemical specific characteristics such as precipitation/dissolution and sorption generally limit the aqueous phase concentration as well as physical characteristics such as particle deposition/suspension and weathering of host minerals. The mobility and groundwater concentration of decay series daughter products is also significantly affected by processes such as decay, sorption, and alpha recoil (Porcelli and Swarzenski, 2003). For example, ^{234}U is a daughter product of ^{238}U and in the host mineral these isotopes exist in secular equilibrium, i.e. $^{238}\text{U}/^{234}\text{U}$ activity ratio is equal to 1. However, the $^{238}\text{U}/^{234}\text{U}$ activity ratio is typically less than one in natural waters due to alpha recoil of the ^{234}U daughter product. Ejection of ^{234}U from the host mineral lowers the $^{238}\text{U}/^{234}\text{U}$ activity ratio in the aqueous phase.

The average abundance of uranium and thorium in the continental crust are 2.7 ppm and 9.6 ppm, respectively (Taylor, 1964). Thorium is typically encountered as Th(IV), which has a low solubility and tends to sorb very strongly to mineral/solid surfaces. The aqueous concentration of thorium is inhibited by formation of sparsely

soluble Th (oxyhydr)oxides (Neck and Kim, 2001). The uranous, U(IV), and uranyl, U(VI), oxidation states of uranium are the most typically encountered in the environment. As the UO_2^{2+} uranyl ion, uranium is relatively mobile in the subsurface and may migrate until reducing conditions are encountered. At this point, reduction of U(VI) may lead to precipitation of uranium minerals such as uraninite, coffinite, or brannerite (Finch and Murakami, 1999). Changes in the bulk solution chemistry surrounding reduced uranium minerals may lead to oxidation and dissolution of the mineral and subsequent increases in the uranium aqueous phase concentration. Many more detailed reviews of actinide geochemistry are available (Allard, 1982; Newton and Sullivan, 1985; Kim et al., 1986; Morse and Choppin, 1991; Fuger, 1992; Murphy and Shock, 1999).

Mertie (1953) and Overstreet et al., (1986) characterized fluvial monazite deposits within the southeastern United States that were commercially mined between 1887 and 1917. Monazite is a rare earth phosphate mineral which typically contains 2 to 10 percent thorium and smaller amounts of uranium. There are two large monazite belts running through the southeastern United States. The principal western belt passes through the area surrounding Simpsonville, SC (Mertie, 1953; Overstreet et al., 1986). Mertie (1953) reported the mean content of ThO_2 and U_3O_8 in the monazite placers as 5.7 and 0.4 percent, respectively. These monazite placers reported by Mertie (1953) and Overstreet et al., (1986) are presumed to be the source of anomalously high levels of uranium and other naturally occurring radionuclides measured in several private wells in the area surrounding Simpsonville, SC (Price, 2006). Total uranium concentrations in the well water ranged from 44.3 to 5570 $\mu\text{g L}^{-1}$ (Hughes et al., 2005). Measurements of the isotopic ratios of the uranium isotopes in water samples from the effected wells indicate that the uranium comes from a natural source (Hughes et al., 2005).

The concentration of uranium and other naturally occurring radionuclides in several river sediments was measured in an effort to better understand the spatial

distribution of the radionuclides in the surrounding areas. These data augment existing measurements of uranium and other naturally occurring radionuclides in water from private wells in the same area (Hughes et al., 2005; SC DHEC 2004). Primordial radionuclide concentrations were determined in river sediments collected from the Reedy River and surrounding creeks near Simpsonville, SC, USA. Radionuclide concentrations were measured *ex-situ* using high-resolution gamma-ray spectroscopy. Results from *ex-situ* measurements were compared with *in-situ* measurements of the surrounding area using a portable NaI:TI detector. No spatial correlation was found between the uranium concentrations in the well waters. Thus it was hypothesized that uranium exists in discrete zones and the wells tap into different fractures.

2. Materials and Methods

Sediments were collected from riverbeds using a Wildco Polar deep lake dredge, or hand scooped on 9/28/2002 and on 3/23/2003. The sediments were dried in an air oven at 110°C then passed through an ASTM #10 (2mm) sieve. Analysis was performed on the <2mm fraction. Approximately 225 g of sediment was packed into 125 mL Nalgene wide mouth (HDPE) containers. The containers were stored for >28 days to attain secular equilibrium between ^{226}Ra and ^{222}Rn and associated daughter products. The sediment samples were analyzed using an Ortec™ POPTOP HPGe spectrometer (GMX-25190-P-S, Oak Ridge, TN). Each 125 mL sample was counted for 18,000 s. An ANALYTICS (Atlanta, GA) calibrated sediment standard was used to determine the efficiency calibration curve.

The gamma emitting radionuclides within the uranium, actinium, and thorium series were used to quantify the parent radionuclide concentration via high-resolution gamma-ray spectroscopic techniques. The concentration of ^{40}K was quantified using the 1460.8 keV (f=10.7%) gamma ray. The concentration of ^{232}Th was inferred by measuring

the concentration of ^{228}Ac using the 911.0 keV (f=27.7%) gamma ray and assuming secular equilibrium between the two radionuclides. The concentration of ^{214}Pb and ^{214}Bi were quantified using the 351.9 keV (f=37.2%) gamma ray and 609.3 keV (f=46.3%) gamma ray, respectively. ^{214}Pb and ^{214}Bi were assumed to be in secular equilibrium with ^{222}Rn . The concentration of ^{214}Pb and ^{214}Bi were in turn used to quantify the concentration of ^{226}Ra in the sediment, based upon secular equilibrium being established between ^{226}Ra and ^{222}Rn . ^{235}U and ^{226}Ra emit gamma rays of energy 185.7 keV (f=57.2%) and 186.2 keV (f=3.6%), respectively. The detector energy resolution is not sufficient to easily separate these peaks. Therefore, the concentration of ^{235}U was calculated by subtracting the fraction of ^{226}Ra using the following equation,

$$[^{235}\text{U}] = \frac{\left(\left(\frac{CR_{187}}{\varepsilon_{peak}} \right) \frac{1}{m_{soil}} - [^{226}\text{Ra}] * f_{226\text{Ra}} \right)}{f_{235\text{U}}} \quad (1)$$

where CR_{187} is the count rate of the peak centered at 187 keV, ε_{peak} is the detector efficiency at that energy, m_{soil} is the mass of sediment in the sample (g), $f_{226\text{Ra}}$ is the gamma ray emission fraction for ^{226}Ra , $f_{235\text{U}}$ is the gamma-ray emission fraction for ^{235}U , $[^{226}\text{Ra}]$ is the concentration of ^{226}Ra in the sediment (Bq/g) based on the average of the ^{214}Bi and ^{214}Pb analyses, and $[^{235}\text{U}]$ is the concentration of ^{235}U in the sediment (Bq/g). The concentration of ^{238}U was calculated by assuming an isotopic activity ratio, $^{238}\text{U}/^{235}\text{U}$, of 21.6.

In-situ gamma measurements were made using a 3" x 3" NaI(Tl) detector (Scionix, Netherlands) with a Nano-spec (Target Instruments, Germany) data acquisition system. One hundred second measurements were made at 47 locations within the

sampling area. The detector was positioned approximately 0.6 m from the side of the truck and approximately 1 m from the ground surface for all measurements. The total equivalent uranium (eU) concentration reported in this work was calculated based upon the activity of ^{214}Bi , and is the combined activity of ^{238}U and ^{234}U . It was assumed that ^{214}Bi was in secular equilibrium with ^{234}U . eU was then calculated assuming 97.68% of the uranium activity in the sample was from ^{234}U and ^{238}U . For the comparison with published datasets, a mass concentration of uranium in soil (usually reported in ppm) was converted to an activity concentration using the factor 12347.5 ± 6.7 Bq of ^{238}U per g of natural uranium as reported by Nir-EI et al. (1999). The calculated ^{238}U activity concentration was then converted to eU as described above.

The sample locations were recorded in terms of degree-minute-second latitudinal and longitudinal position (WGS 84) using a hand-held Global Positioning System (GPS) unit. These coordinates were converted into decimal degrees and used to map the sampling locations onto a Geographic Information System (GIS) map file that contained a digitized rendering of the Simpsonville area topological quadrangle. The ArcMap 8.3 GIS program (ESRI, 1999-2002) was used with a Transverse Mercator projection of 1980 geodetic reference system (GRS80) satellite-determined spheroid information for North America that is compatible with the acquired GPS location information. The contour regions provided to show areas of equal value for *in-situ* gamma data were generated using the spatial interpolation features of ArcMap 8.3, relying upon Inverse Distance Weighted estimations.

Monte Carlo N-Particle Transport Code MCNP 5 (Forster et al., 2004) was used to estimate the detection efficiency of the 3" x 3" NaI(Tl) spectrometer for the *in-situ* geometry. The modeled geometry assumes uranium was uniformly mixed in soil ($\rho = 1.6 \text{ g/cm}^3$) to a depth of 30 cm. This simulated disk source had a radius of 10 m and the detector was positioned 1 m above the disk on the axis and perpendicular to the disk. To

assure good statistics, 10^8 histories were run. The 609 keV gamma-ray from ^{214}Bi was used to calculate the absolute detection efficiency.

3. Results and Discussion

5 3.1 *Ex-situ* measurements of uranium and other radionuclides in stream sediments

The concentration of ^{238}U , ^{232}Th , ^{226}Ra , and ^{40}K for the 16 sediment samples are listed in Table 1. The concentration of uranium and other radionuclides measured in this work are compared with the concentrations measurements of two major surveys. In 1973 the National Uranium Resource Evaluation (NURE) was initiated by the Atomic Energy Commission to locate potential uranium sources within the United States. Price and Ferguson (1980) discuss the regional distribution of uranium and thorium surrounding the sampling area was reported by elevated uranium and thorium as part of the Hydrogeochemical and Stream Sediment Reconnaissance program within NURE. In 2000, the United Nations Scientific Committee on the effects of Atomic Radiation (UNSCEAR) performed aerial gamma-ray surveys on natural radiation exposures. The mean concentration and concentration range from the UNSCEAR survey for the United States are listed in Table 1 for each radionuclide. The range of ^{238}U concentrations measured as part of this work in sediments collected from the Reedy River and surrounding creeks (11.1 to 74.2 Bq kg^{-1}) are consistent with the range of values from the UNSCEAR and NURE surveys. The NURE survey data from the Greenville quadrangle reported uranium concentrations in stream sediment ranging from below detection to 335 ppm with an average value of 17.2 ppm. The average uranium mass concentration of 17.2 ppm corresponds to an average uranium activity concentration of 212.4 Bq kg^{-1} . The average uranium concentration from the NURE survey is higher than the average value measured in the stream sediments collected in this work (37.8 Bq kg^{-1}). Therefore, the uranium concentrations in stream sediments from the Reedy River and

surrounding creeks are slightly lower than other sediments within the Greenville Quadrangle of the NURE survey. The uranium activity concentrations reported in Table 1 are consistent with the reported mean value of 35 Bq kg⁻¹ for the United States from the UNSCEAR survey. Since the UNSCEAR data was collected using aerial gamma-ray surveys, they are assumed to represent an *in-situ* measurement of the entire area. Comparatively, the *ex-situ* stream sediment data shown in Table 1 were collected solely on stream sediments. The average concentrations of ²³²Th, ²²⁶Ra, and ⁴⁰K measured in the stream sediments are consistent with the range of values for the United States from the UNSCEAR survey. Four sediment samples have ²³²Th concentrations higher than the average value of 35 Bq kg⁻¹ from the UNSCEAR survey. However, only two sediments are outside the specified concentration range. All ²²⁶Ra concentrations were within the range of concentrations reported by the UNSCEAR survey. These data indicate that the anomalously high levels of uranium and other radionuclides in groundwater supplying local private drinking water wells are not significantly affecting stream sediments.

The ²³⁸U/²³²Th activity ratio ranges from 0.29 to 3.12 with an average value of 1.28, as computed using the values listed in Table 1. Comparison of these ratios with the average ²³⁸U/²³²Th activity ratio in the upper crust is typically 0.8 (Porcelli and Swarzenski, 2003), indicates a slight enhancement of ²³⁸U, relative to ²³²Th. Extraordinarily high ²³⁸U concentrations relative to ²³²Th were observed by Lozano et al. (2002) when measuring the radionuclide concentrations in sediments downstream from a uranium mine. At a discretely identified accumulation point, Lozano et al. (2002) reported average ²³⁸U and ²³²Th concentrations of 5025 and 23 Bq kg⁻¹, respectively (yielding a ²³⁸U/²³²Th activity ratio of 218).

The enhanced ²³⁸U concentration may be due to the solubility and mobility of U(VI)O₂²⁺. Thorium is relatively immobile in most environments as it is easily removed

from solution by precipitation, co-precipitation, or sorption. The aqueous phase concentration is limited by the formation of sparsely soluble hydroxide solids (Neck and Kim, 2001). Uranium is present in the tetravalent state in many host minerals (Finch and Murakami, 1999). Oxidizing conditions may favor formation of the hexavalent uranyl cation, UO_2^{2+} , which is more soluble and mobile in most environments, relative to U(IV). Once leached or dissolved from the host mineral, partitioning of uranyl between the solid and aqueous phase will be controlled by sorption, particle deposition/suspension, and dissolution/precipitation mechanisms. Therefore, the slight enhancement of uranium relative to thorium in some samples may be due to dissolution and re-adsorption of uranium.

The ^{226}Ra and ^{232}Th activity concentrations are relatively similar, although a slight depletion of ^{226}Ra relative to ^{232}Th was observed in several sediments (Table 1). ^{226}Ra may be produced on the surface of the host rock or ejected into the aqueous phase via alpha recoil. Both processes would result in a depletion of ^{226}Ra relative to ^{232}Th . Additionally, ^{226}Ra is more soluble and mobile in groundwater than ^{232}Th and may leach from the host rock, independent of the point of production.

3.2 *In-situ* uranium measurements

A contour map of eU concentration from 47 *in-situ* radiation measurements using the NaI(Tl) gamma spectrometer is shown in Figure 1. It is important to note that eU concentration in the soil was inferred via radiation emitted from the ^{214}Bi daughter product. An assumption inherent in the calculation is that secular equilibrium is attained between the daughter products, which is not necessarily the case. The most common source of deviation between eU and actual uranium concentration can be attributed to the diffusion of radon gas out of the soil, resulting in a lower ^{214}Bi activity concentration, and thus a lower uranium concentration.

There is no clearly distinct uranium source in the contour map shown in Figure 1 (although the dataset is not large enough to allow for an accurate prediction). The eU concentration ranged from 2 Bq kg⁻¹ to 75 Bq kg⁻¹ across the sampling area. The mean and median eU concentrations were 40 Bq kg⁻¹ and 41 Bq kg⁻¹. Galbraith and Saunders (1983) and Taylor (1964) reported average uranium concentrations in granite as 4.1 ppm and 4.8 ppm, respectively. Converting those values as described in the materials and methods section yields eU values of 50 and 59 Bq kg⁻¹, respectively. Additionally, Taylor (1964) reported an average uranium concentration in continental crust of 2.7 ppm (corresponding to an eU concentration of 33 Bq kg⁻¹). The mean and median uranium concentrations from the *in-situ* data fall between the average values for continental crust and granite, as reported above. Comparing the *in-situ* measurements with the average data reported by Galbraith and Saunders (1983) and Taylor (1964), it appears the uranium concentration in the surface soils surrounding many of the creeks closely resemble the expected (average) values for similar geologic materials. The range of values that we report for the *in-situ* eU in soil are similar to the *ex-situ* ²³⁸U concentrations measured in sediment.

4. Conclusion

Concentrations of uranium and other radionuclides were measured in river sediments from the Reedy River and surrounding creeks near Simpsonville, SC. Additionally, *in-situ* gamma spectroscopy was used to measure the uranium concentration in soils in the surrounding area. The uranium concentration in the soils and river sediments were consistent with the average reported values for similar geologic materials. The Simpsonville area is predominantly underlain by granite, with some amphibolites present (Sodja, 2005). Additionally, the area overlays a principal monazite belt which runs through the Piedmont of the Carolinas. The monazite placers

are presumed to be the source of elevated concentrations of uranium, thorium, and other naturally occurring radionuclides in the area. Mertie (1953) reported percent U_3O_8 fractions in monazite placers ranging from 0.18 to 0.98. These would yield ^{238}U activity concentrations ranging from 20 to 100 $kBq\ kg^{-1}$, which are significantly higher than the
5 sediments measured in this work. This monazite is presumed to be the source of elevated uranium concentrations in private drinking water wells. However, based upon the low uranium concentrations measured in these stream sediments relative to groundwater concentrations, the elevated uranium concentrations do not appear to be affecting the stream sediments in the local area.

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Table 1: Concentration of ^{238}U , ^{232}Th , ^{226}Ra , and ^{40}K in sediments. Error reported as 2σ standard deviation based on counting statistics

5

Sediment ID	Concentration in Sediment (Bq/kg)			
	$^{238}\text{U}^{\text{a}}$	$^{232}\text{Th}^{\text{b}}$	$^{226}\text{Ra}^{\text{c}}$	$^{40}\text{K}^{\text{d}}$
1	13.2 ± 16.9	17.0 ± 2.4	14.2 ± 0.9	385.9 ± 17.0
2	34.6 ± 21.9	64.2 ± 2.3	31.1 ± 1.1	641.3 ± 19.0
3	12.5 ± 15.9	13.9 ± 4.4	11.4 ± 1.7	386.5 ± 33.8
4	30.1 ± 18.2	32.6 ± 0.0	21.4 ± 0.9	672.8 ± 0.7
5	74.2 ± 24.6	185.8 ± 8.5	41.2 ± 2.4	846.6 ± 41.1
6	30.6 ± 17.3	13.0 ± 4.5	12.5 ± 1.8	568.4 ± 36.5
7	62.7 ± 20.2	42.3 ± 5.5	27.6 ± 2.1	800.5 ± 40.2
8	11.1 ± 17.3	19.2 ± 4.5	14.6 ± 1.8	434.3 ± 34.6
9	43.1 ± 20.0	18.1 ± 5.1	19.5 ± 2.0	417.8 ± 37.8
10	23.2 ± 38.5	16.4 ± 5.0	12.5 ± 1.9	665.7 ± 41.8
11	44.7 ± 49.4	154.0 ± 8.6	28.7 ± 2.4	525.4 ± 40.1
12	38.5 ± 40.5	18.0 ± 5.0	17.9 ± 2.0	650.0 ± 41.4
13	38.8 ± 41.0	33.5 ± 5.4	16.3 ± 2.0	671.6 ± 41.6
14	39.9 ± 44.4	38.9 ± 5.8	25.6 ± 2.2	1046.9 ± 46.9
15	34.6 ± 42.1	34.3 ± 5.6	17.8 ± 2.1	457.8 ± 38.8
16	72.9 ± 43.6	23.3 ± 5.2	30.0 ± 2.2	577.2 ± 40.6
AVERAGE	37.8 ± 29.5	45.3 ± 4.9	21.4 ± 1.8	609.3 ± 34.5
UNSCEAR (mean)	35	35	40	370
UNSCEAR (range)	4-140	4-130	8-160	100-700
NURE (mean)	212.4 ^e	-	-	-
NURE (range)	6.2-4143 ^e	-	-	-

^aAs determined from ^{235}U concentration assuming a $^{238}\text{U}/^{235}\text{U}$ activity ratio of 21.6

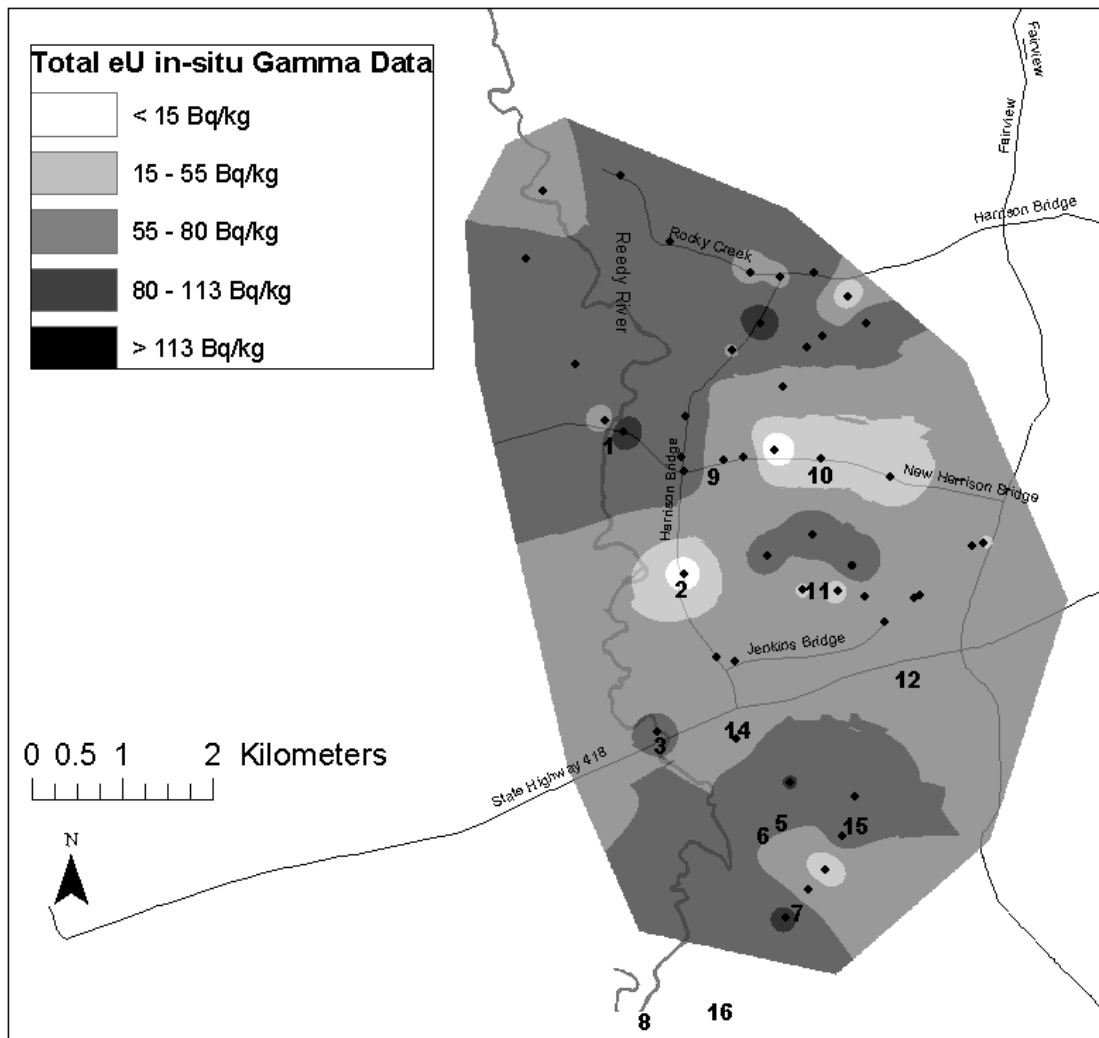
^bAs determined from ^{228}Ac concentration

^cAs determined from the average of ^{214}Bi and ^{214}Pb concentration

^dAs determined from 1461 keV gamma

^eDetermined using the value of 12347.5 Bq of ^{238}U per g of natural uranium reported by Nir-El et al., 1999

10



5 Figure 1: Contour maps developed using eU concentration obtained from *in-situ* gamma-ray spectroscopy. The numbers represent the location of the river and stream sampling while the diamonds represent the location of the *in-situ* measurement. Sampling area is in the vicinity of N 24° 39.0' and W 82° 16.5'.