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# URANIUM ATTENUATED BY A WETLAND 50 YEARS AFTER RELEASE INTO A STREAM

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## ABSTRACT

Wetlands have several important roles in the hydrological cycle, including maintaining water quality by removing surface and groundwater contaminants. Over time, the wetlands themselves can become contaminated, posing a secondary environmental threat. The objective of this study was to calculate the inventory of uranium (U) remaining in a wetland 50 years after the Fuel Fabrication Facility on the Savannah River Site located in South Carolina released 43.5 Mg of U into the nearby environment. Over 232,700 gamma spectra and their associated GPS coordinates were collected and collated into a map of the contaminated land area. Five core samples were also collected that contained U concentrations as high as 14,099 mg/kg (background levels are about 2.7 mg/kg U). The contaminated area was 278,000 m<sup>2</sup> and it contained 36.2 Mg U, about 83% of the U released. About 80% of the U in the wetland was concentrated in a former beaver pond, a 73,000 m<sup>2</sup> area (26% of the contaminated area). This contaminated wetland area was almost 2 km from the source, indicating that it comprised unique hydro-biogeochemical properties for immobilizing the released U. To our knowledge, this is the first data-rich study to quantify the long-term effectiveness of a wetland to immobilize inorganic contaminants. Significant environmental changes to the system, such as those associated with hydrology, forest fires, or anthropogenic land use, may alter the complex hydro-biogeochemical interactions necessary for the long-term immobilization of the U.

**Key Words:** Natural Attenuation, Wetlands, Gamma Spectroscopy, Contaminant Inventory, Source Terms, Depth Profiles, Global Positioning System

## INTRODUCTION

Wetlands possess a diverse set of biogeochemical properties that originate from their unique hydrological regime. These wet environments promote conditions that accumulate organic matter (OM), and create steep biogeochemical redox, organic matter, and microbial gradients that together can enhance binding of groundwater and surface water contaminants.<sup>1-3</sup> Because wetlands have the tendency to accumulate contaminants, they can become a naturally occurring secondary source term.<sup>4-7</sup>

Among the most important parameters needed to assess the potential threat posed by a secondary wetland source is the mass of contaminant in the system. Quantifying the non-point source term is commonly among the largest sources of uncertainty in risk models.<sup>8</sup> The longevity of contaminant attenuation by wetlands is susceptible to well documented anthropogenic activities, as well as natural perturbations that can be chronic or episodic, such as droughts, floods, and fires.<sup>9-11</sup> The drying of wetlands may promote oxidization of the soil, which in turn may promote the breakdown of OM and the release of contaminants bound to small OM fragments.<sup>12</sup> Conversely, episodic flooding may promote resuspension of contaminated particles, while extended flooding may kill plants, the primary source for soil OM and an anchor for stabilizing soil particles. Finally, fires may destroy plants and soil OM, increasing run-off, and altering hydrological regimes by reducing evapotranspiration.<sup>11</sup>

Most long-term studies evaluating wetland attenuation of contaminants focus on a wetland's capacity to remove mobile aqueous contaminants by monitoring stream-contaminant concentrations before and after entering a wetland.<sup>13, 14</sup> Another approach is to compare the soil contaminant concentrations inside and outside the contaminated wetland.<sup>15</sup> Neither approach is intended to provide an areal estimate of the contamination zone nor to provide a quantitative estimate of the long-term contaminant retention by the wetland.

The Fuel Fabrication Facility on the Savannah River Site in South Carolina made uranium (U) fuel and target assemblies between 1965 and 1988 for use in the production of nuclear materials used for defense purposes, including plutonium-238 (<sup>238</sup>Pu) and -239 (<sup>239</sup>Pu).<sup>16</sup> Uranium waste was generated at several steps during this process, including acid etching (nitric, phosphoric, and sulfuric), cutting, milling, and chlorinated solvent washing. The waste, reported at concentrations >300,000 mg/L U,<sup>17</sup> was treated with hydroxide to promote precipitation and then passed through a settling basin and a filtration system. A total of 43.5 Mg of U was accidentally and purposely released to a nearby stream, the A-14 Tributary, or in later years to an unlined seepage basin, of which, approximately 61% of the U was released during a 3-year period, from 1968 to 1971 (Supporting Information, Figure S1).<sup>17</sup> Based on historic records, released U was present in dissolved, colloidal, and particulate phases and consisted predominantly of depleted U. Releases likely included U in zero, tetravalent, and hexavalent oxidation states and potentially the pentavalent state. The U colloids and particulates likely included U bonded to clay particles, metallic fines, and waste processing precipitates, the latter of which may have included sodium diuranate, hydrogen uranyl phosphate, uranium(IV) oxide and uranium mixed oxides (U<sub>4</sub>O<sub>9</sub>, and U<sub>3</sub>O<sub>8</sub>). A more detailed description of the waste generated from the Fuel Fabrication Facility is presented in the Supporting Information.

Uranium from the Fuel Fabrication Facility flowed from the A-14 Tributary and concentrated in the Tims Branch riparian wetland (Supporting Information Figure S2).<sup>17, 18</sup> Uranium concentrations as high as 6000 mg/kg were reported in a portion of the stream referred to as Steed Pond, an abandoned farm pond predating the Savannah River Site.<sup>17</sup> Steed Pond exists

today as a wetland following the failure of its dam structure in 1984. Transport of U out of the Tims Branch system during stream baseflow is not a concern, as stream water monitoring data indicates that dissolved U concentrations are at or below regulatory limits (EPA Maximum Contamination Level is 30 µg/L U).<sup>19</sup> Elevated U concentrations associated with suspended solids in the stream water have been reported during high rainfall events.<sup>20, 21</sup> Dissolved U generally binds strongly to Savannah River Site sediments ( $K_d$  values >300 mL/g), especially in wetland sediments.<sup>22-25</sup>

Tims Branch offers a unique opportunity to evaluate the long-term capacity for a wetland to attenuate U because reasonable records exist for the mass released, and there have been limited impacts from anthropogenic activities since becoming contaminated, other than stream levels were lowered as a consequence of curtailed facility operations in 1988. The objective of this study was to calculate the inventory of U remaining in the Tims Branch wetland 50 years after release from the Fuel Fabrication Facility. The approach was to estimate the mass of U remaining in the wetland and then compare it to the initially deposited 43.5 Mg U.

## MATERIALS AND METHODS

A walk-over survey of Tims Branch was conducted to determine the areal distribution of U in the wetland using portable gamma and X-ray spectroscopy equipment. Integration of the gamma spectral data with U depth profile data from soil cores provided an estimate of the U inventory in the contaminated area.

**Portable Gamma and X-Ray Spectroscopy Survey.** The land area within Tims Branch that was surveyed was about 500,000 m<sup>2</sup> (0.5 km<sup>2</sup>). This area was selected in order to directly measure the spatial extent of the near surface U deposition to define contamination boundaries along Tims Branch. The following is a brief discussion of the hardware and software of the portable gamma and X-ray spectroscopy equipment, the approach and organization of the survey, and data management.

To facilitate multiple surveyors working concurrently, Tims Branch was segmented into survey areas, each with parallel lines spaced 3.3 m apart, using ArcMap software (ArcGIS for Desktops, ESRI Software). These parallel lines were perpendicular to Tims Branch and visible on a computer tablet provided to each surveyor. Each surveyor was assigned a gamma spectral backpack and a segment of Tims Branch each day. As many as eight surveyors worked at a time until all segments of Tims Branch were surveyed. To complete the project approximately 560 transects were surveyed in six days, with approximately 740,000 gamma and X-ray spectra collected in total. Gamma spectral analyses and radiation sediment-shielding calculations are presented below.

Backpacks were equipped with a 3.8cm x 6.4cm x 20.3cm NaI detector, integrated GPS unit, and computer tablets. Gamma spectral measurements and positions were saved every second on the computer and backpack control module (multi-channel analyzer). The computer and software provided both navigation and visualization of the gamma spectral data with one second updates (images of equipment in Figure S3 of the Supporting Information). Detection spot size of each measurement was approximately 2-m diameter. At the end of each survey day the data stored in each backpack was downloaded and archived.

**Soil Cores and ICP-MS Analyses.** Soil cores were collected to provide insight into how U was distributed at depths below those probed by the gamma measurements (~5cm for U, depending on detector height). Five soil cores (2.5cm D × 100cm L or 2.5cm D × 25-cm L)

stored in butyrate plastic sleeves were collected from three high-U concentration locations identified by the gamma survey: A-14 Delta, Steed Pond, and Beaver Pond (Figure 1). Upon arrival to the laboratory, cores were sectioned into 3 to 5 cm segments (Steed Pond and Beaver Pond cores) or ~1 cm segments (A-14 Delta core) based on visible sediment boundaries and soil characteristics. This variable versus fixed length sectioning of the cores was selected to maximize the chances of identifying U-enriched layers.

Total U concentrations in sediment samples were determined by digesting 1-g of homogenized sediment samples using 90 to 95 °C heated concentrated HNO<sub>3</sub> and 30% H<sub>2</sub>O<sub>2</sub> per the EPA 3050B method.<sup>26</sup> All measurements were conducted in duplicate. An aliquot of each digestate was analyzed in a 2% HNO<sub>3</sub> matrix via Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Thermo X-Series II). In addition to a water blank, a background soil sample collected from Tims Branch upstream of the A-14 confluence was included in the analysis. The U concentration in all samples were well above the instrument's detection limit.

## RESULTS AND DISCUSSION

**Gamma Spectra Analysis.** Prior to the spectral analysis, a review was conducted of the Fuel Fabrication Facility's operational and material release history into the A14 outfall.<sup>16, 17</sup> Records show that the U utilized in Fuel Fabrication Facility was processed U, indicating all decay products were removed. Understanding the source and approximate age of the U after processing provides insight into the current gamma and X-ray emissions.

Analysis of more than 740,000 spectra acquired from the contaminated and surrounding area of Tims Branch was accomplished with the software AVID (Advanced Visualization and Integration of Data, Department of Energy NA-84, Washington, DC). Uranium analysis of the field spectra identified two isotopes, <sup>238</sup>U and <sup>235</sup>U. Both isotopes' activities were well above the activities of the primordial U and its decay products. AVID data processing and Lidar ground elevation contour data (1-m resolution) of Tims Branch were used to determine the spatial area of the near-surface deposited U. Three regions of Tims Branch were defined, and the area of each region was determined with GIS software ESRI ArcMap 10.8 (Figure 1). These regions were named Beaver Pond (54,129 spectra with an area of 33,551 m<sup>2</sup>), north of the Beaver Pond (70,730 spectra with an area of 73,173 m<sup>2</sup>), and south of the Beaver Pond (107,850 spectra with an area of 171,148 m<sup>2</sup>) (Table 1). A ranking of the normalized composite spectra by peak height is as follows: Beaver Pond Region >> South Region > North Region (Figure 1).

**Gamma maps.** While the total area of the survey was approximately 500,000 m<sup>2</sup>, the U deposition area as defined by the gamma spectral survey was narrowed to 277,872 m<sup>2</sup> (Figure 1). Consistent with the composite spectra, a ranking of the regions by their U activity is: Beaver Pond >> South of the Beaver Pond (Steed Pond) > North of the Beaver Pond (A-14 Delta). It is noteworthy that elevated U activity was not detected in the A-14 Tributary, the initial 2 km before reaching Tims Branch (Figure 1; Figure S2). As previously suggested by Evans et al.<sup>17</sup>, this can likely be attributed to the much steeper topography in this portion of the study site. The A-14 Tributary has a 4% grade, whereas Tims Branch has a 0.3% grade (Figure 2). Finally, the most southern boundary of the U deposition zone is the sharp U demarcation of Steed Pond's most southern bank, where the breached dam was located. Based on aerial photography taken in 1970, about the time of the greatest U releases to the streams, Steed Pond was about 4.5 ha.<sup>17</sup>

**Depth Profiles.** The locations of the five soil cores are presented in Figure 1 and their profiles of U concentration and cumulative mass fractions are presented in Figure 3. Soil

concentrations were as high as 14,099 mg/kg U (Sample #807a collected from the Beaver Pond region), whereas background U concentrations were ~2.7 mg/kg (sample collected in Tims Branch wetland upstream of the A-14 Tributary confluence). There were two general U concentration trends of the depth profiles. Cores from the former Steed Pond, #1003 and #1004, had a steady decreasing concentration profile. About 90% of all the U was in the upper 19 to 25 cm depth. The three upper-stream cores from the former Beaver Pond (#807a and #807b) and the A-14 Delta (#HW; with 1-cm sampling depth resolution) had maximum U concentrations in the subsurface, at a depth between 5 to 10 cm.

**Uranium Inventory.** To estimate the contaminant U inventory, it was necessary to account for not only the near surface U measured by the gamma spectral measurements, but also the deeper U observed in the core samples (Figure 3). To accomplish this a Gamma Correction Factor was developed that was used with the gamma spectral activity data corrected for background levels to account for the underlying U, whose emissions were shielded by the soil. RadDetect software (Department of Energy NA-84, Washington, DC) was used to calculate a detector response factor for each spectral backpack.<sup>27</sup> Calculation of the detector response factor is based on photon emissions associated with depleted uranium, including emissions from <sup>234</sup>Th (63.3keV, 92.4keV, 92.8keV), <sup>234m</sup>Pa (766.4keV, 1001.0keV) and <sup>235</sup>U (89.9keV, 93.4keV, 105keV, 109.2keV, 142.6keV, 163.4keV, 185.7keV, 205.3keV). Source geometry was assumed to be a distributed source with no shielding resulting in a factor of  $3.62 \times 10^{-3}$  cps/Bq/m<sup>2</sup> <sup>238</sup>U (cps, counts per second; 133 cps/μCi/m<sup>2</sup> <sup>238</sup>U). The estimated minimum detectable activity of <sup>238</sup>U was calculated to be  $3.93 \times 10^4$  Bq/m<sup>2</sup> (1.06 μCi/m<sup>2</sup>). Spectral responses associated with each region of Tims Branch were calculated with AVID. Spectra associated with the Beaver Pond region were assigned Subsurface Enriched Profiles (based on core #807a), while spectra from the north and south of the Beaver Pond were assigned a Steady Decline Profile (based on core #1004). Soil shielding of underlying gamma and X-ray emissions was quantified by basic radiation shielding calculations and parameterized as the half-value layer coefficient,<sup>28</sup> defined as the thickness that attenuates half of the photon emissions. The half-value layer coefficient is dependent on the energy of the photon emission and the composition of the attenuating material, in this case the soil. NIST XCOM<sup>29</sup> calculations were carried out to determine the photon transmission mass attenuation factors based on photon cross sections and the elemental composition of a soil. The soil composition was based on a NIST reference soil (#2710a) that was enriched in Fe to better represent the study site. Once the half-value layer coefficients were determined, it was possible to estimate the Gamma Correction Factors, which were 18.61 for the Subsurface Enriched Profile, and 1.88 for the Steady Decline Profile. These Gamma Correction Factors indicate that the gamma spectral estimates for U activity in the Subsurface Enriched Profile regions of the study site needed to be multiplied by a factor of 18.61 and the Steady Decline Profiles by a factor of 1.88 to account for the U present throughout the sediment profile. The larger Gamma Correction Factor for the Subsurface Enriched Profiles is attributed to greater concentrations of U in the lower portions of soil profile.

Additionally, a first-approximation estimate of the U inventory based entirely on soil core U depth profiles was calculated and compared to the gamma spectral estimate. The area defined by spectral near surface U measurements for each region, and profile type were used to estimate the total U concentration.

Based on the spectral calculation, there was a total mass of 36.2 Mg U (also referred to as a metric tonne) in the wetland, which is 83% of the estimated 43.5 Mg released from the Fuel Fabrication Facility (Table 1). The remaining 17%, 7.3 Mg U, was presumably released

downstream. Approximately 80% of all the U in the wetland was in the Beaver Pond region. This region comprised only 26% of the contaminated area, yielding an area-based concentration of 0.4 kg U/m<sup>2</sup>. The first-approximation estimate of the U inventory yielded a near identical result, 37.4 Mg U (Table 1). This similarity is not entirely unexpected because two important parameters in the calculations were identical for both calculations, the area and the depth profile of the U. The similarity of these calculations indicates that both methods estimated about the same mass of U near surface and in the subsurface.

Based on a 1984 soil survey, Evans et al.<sup>17</sup> estimated that 30.7 Mg U, or 70% of the U released from the Fuel Fabrication Facility, was in Steed Pond. While they obtained somewhat similar values as presented in Table 1, it is a coincidence. They selected the 15 cores with the greatest U concentrations out of the 30 cores collected from Tims Branch to base their calculation. In their single mass calculation, they assumed that all the U was accumulated in the 53,000 m<sup>2</sup> of Steed Pond, the average U concentration was 581 mg U/kg dry-weight soil, and the contaminant depth was 60 cm, resulting in a total of 30.7 Mg U (not stated, but also assumed was a soil bulk density of 1.65 Mg/m<sup>3</sup>). The present study had the advantage of building on the data presented by Evans et al.<sup>17</sup> Among the improvements in understanding of the U inventory at the study site is that: 1) U is not only in Steed Pond but also in the Beaver Pond and the A-14 Delta (Figure 1), 2) the contamination area is much greater than previously believed, closer to 278,000 m<sup>2</sup> rather than 53,000 m<sup>2</sup> (Table 1), and 3) most U is presently located in the upper 20 cm, not 60 cm soil depth (Figure 3).

**Implications.** This wetland has been extremely effective for the last 50 years at immobilizing the released U. This conclusion is supported by stream water measurements collected over the last several decades indicating that U concentrations have been consistently below drinking water regulatory limits (i.e., the EPA Maximum Contaminant Level, 30 µg/L<sup>19</sup>). To our knowledge, this is the first estimate quantifying the effectiveness of a wetland to retain inorganic contaminants spanning decades. This estimate was made possible by several characteristics of the site. First there were few land use changes to the system during the last 50 years and no attempts were made to remediate the contaminant U. Secondly, there was a reasonable estimate of total U released by the Fuel Fabrication Facility.<sup>17</sup> Finally, recent advances in GIS and the handling of large amounts of data from the gamma and X-ray detection equipment made it possible to determine accurately the areal distribution of the contamination.

The fact that the Beaver Pond region is almost 1 km downstream from where the stream water U first entered the Tims Branch wetland indicates that this portion of the wetland comprised unique hydro-biogeochemical properties for immobilizing the released U. Significant changes to the wetland system in the future, such as in hydrology, forest fires, or anthropogenic changes in land use, may alter the complex hydro-biogeochemical processes responsible for the U immobilization.

## ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge.

Figures include: 1) Historical U releases to streams from the Fuel Fabrication Facility, 2) expanded map of U activity in Tims Branch, and 3) photographs of the equipment used

during survey. Additional background information is provided about the waste generated from the Savannah River Site's Fuel Fabrication Facility.

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### Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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## REFERENCES

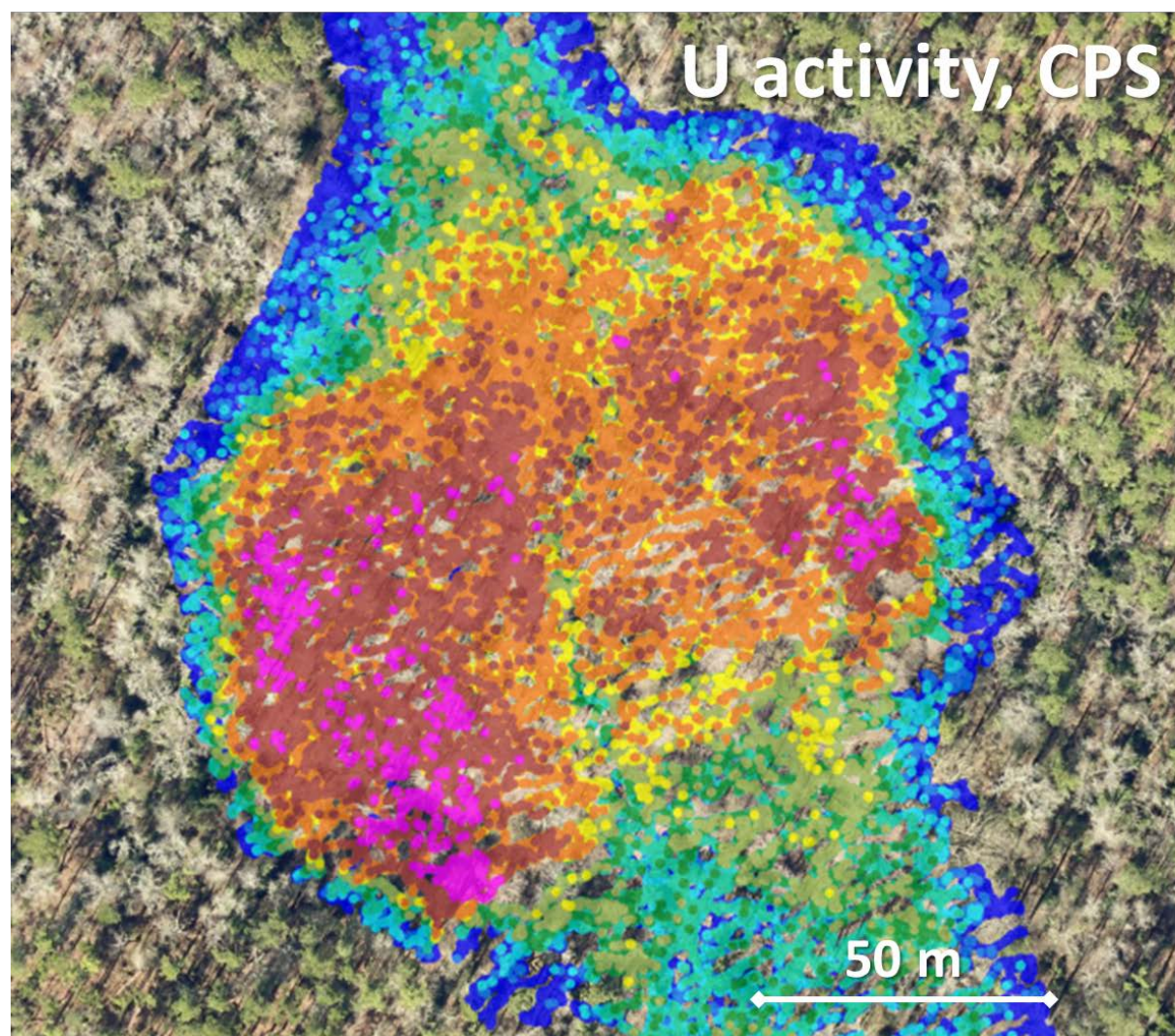
1. O'Geen, A. T.; Budd, R.; Gan, J.; Maynard, J. J.; Parikh, S. J.; Dahlgren, R. A., Mitigating nonpoint source pollution in agriculture with constructed and restored wetlands. In *Advances in Agronomy, Vol 108*, Sparks, D. L., Ed. 2010; Vol. 108, pp 1-76.
2. Gambrell, R., Trace and toxic metals in wetlands - a review. *Journal of Environmental Quality* **1994**, 23, (5), 883-891.
3. Frohne, T.; Rinklebe, J.; Diaz-Bone, R. A., Contamination of floodplain soils along the Wupper River, Germany, with As, Co, Cu, Ni, Sb, and Zn and the impact of pre-definite redox variations on the mobility of these elements. *Soil & Sediment Contamination* **2014**, 23, (7), 779-799.
4. Sheoran, A.; Sheoran, V., Heavy metal removal mechanisms of acid mine drainage in wetlands: A critical review. *Minerals Engineering* **2006**, 19, (2), 105-116.
5. Wang, Y.; Frutschi, M.; Suvorova, E.; Phommavanh, V.; Descostes, M.; Osman, A. A. A.; Geipel, G.; Bernier-Latmani, R., Mobile uranium(IV)-bearing colloids in a mining-impacted wetland. *Nature Communications* **2013**, 4, 1-9.
6. Wang, Y.; Bagnoud, A.; Suvorova, E.; McGivney, E.; Chesaux, L.; Phommavanh, V.; Descostes, M.; Bernier-Latmani, R., Geochemical control on uranium(IV) mobility in a mining-impacted wetland. *Environmental Science & Technology* **2014**, 48, (17), 10062-10070.



7. Kaplan, D. I.; Zhang, S.; Roberts, K. A.; Schwehr, K.; Xu, C.; Creeley, D.; Ho, Y.-F.; Li, H.-P.; Yeager, C. M.; Santschi, P. H., Radioiodine concentrated in a wetland. *Journal of Environmental Radioactivity* **2014**, *131*, 57-61.
8. Zheng, C.; Bennett, G. D., *Applied contaminant transport modeling*. Wiley-Interscience New York: 2002; Vol. 2.
9. Davis, J. A.; Froend, R., Loss and degradation of wetlands in southwestern Australia: Underlying causes, consequences and solutions. *Wetlands Ecology and Management* **1999**, *7*, (1-2), 12-23.
10. Meng, W.; He, M.; Hu, B.; Mo, X.; Li, H.; Liu, B.; Wang, Z., Status of wetlands in China: A review of extent, degradation, issues and recommendations for improvement. *Ocean & Coastal Management* **2017**, *146*, 50-59.
11. Brinson, M. M.; Malvárez, A. I., Temperate freshwater wetlands: types, status, and threats. *Environmental conservation* **2002**, *29*, (2), 115-133.
12. Sobolewski, A., A review of processes responsible for metal removal in wetlands treating contaminated mine drainage. *International Journal of Phytoremediation* **1999**, *1*, (1), 19-51.
13. Dean, A. P.; Lynch, S.; Rowland, P.; Toft, B. D.; Pittman, J. K.; White, K. N., Natural wetlands are efficient at providing long-term metal remediation of freshwater systems polluted by acid mine drainage. *Environmental science & technology* **2013**, *47*, (21), 12029-12036.
14. Callaway, J. C.; Delaune, R. D.; Patrick Jr, W. H., Heavy metal chronologies in selected coastal wetlands from Northern Europe. *Marine Pollution Bulletin* **1998**, *36*, (1), 82-96.
15. Jacob, D. L.; Yellick, A. H.; La Toya, T. K.; Asgary, A.; Wijeyaratne, D. N.; Saini-Eidukat, B.; Otte, M. L., Cadmium and associated metals in soils and sediments of wetlands across the Northern Plains, USA. *Environmental pollution* **2013**, *178*, 211-219.
16. Reed, M. B.; Swanson, M. T. *300/M-Area-Fuel and Target Fabrication*; 1189; New South Associates: Stone Mountain, GA, 2006.
17. Evans, A. G.; Bauer, L. R.; Haselow, J. S.; Hayes, D. W.; Martin, H. L.; McDowell, W. L.; Pickett, J. B. *Uranium in the Savannah River Site Environment*; WSRC-RP-92-315; Westinghouse Savannah River Company, Aiken, SC, 1992.
18. Pickett, J. B. *Heavy Metal Contamination in Tims Branch Sediments*; OPS-RMT--900200; Westinghouse Savannah River Company: Aiken, SC, 1990.
19. *SRNS Savannah River Site Environmental Report 2018*; SRNS-RP-2019-00022; Savannah River Nuclear Solutions: Aiken, South Carolina, 2019.
20. Batson, V. L.; Bertsch, P.; Herbert, B., Transport of anthropogenic uranium from sediments to surface waters during episodic storm events. *Journal of Environmental Quality* **1996**, *25*, (5), 1129-1137.
21. Hayes, D. W.; Ouzts, C. D. *Sediment transport studies in Tims Branch*; DPST-86-468; E.I. du Pont de Nemours & Co.: Aiken, South Carolina, 1986.
22. Li, D.; Kaplan, D. I.; Chang, H.-S.; Seaman, J. C.; Jaffé, P. R.; Koster van Groos, P.; Scheckel, K. G.; Segre, C. U.; Chen, N.; Jiang, D.-T., Spectroscopic evidence of uranium immobilization in acidic wetlands by natural organic matter and plant roots. *Environmental Science & Technology* **2015**, *49*, (5), 2823-2832.
23. Li, D.; Seaman, J. C.; Chang, H. S.; Jaffé, P.; Groos, P. K. v.; Jiang, D. T.; Chen, N.; Lin, J.; Authur, Z.; Pan, Y.; Scheckel, K.; Newville, M.; Lanzirotti, A.; Kukkadapu, R., Retention and chemical speciation of uranium in an oxidized wetland sediment from the Savannah River Site. *Journal of Environmental Radioactivity* **2014**, *131*, 40-46.

24. Kaplan, D. I.; Serkiz, S. M., Quantification of thorium and uranium sorption to contaminated sediments *J. Radioanalytical and Nuclear Chemistry* **2001**, *248*, 529-535.
25. Kaplan, D. I. *Geochemical Data Package for Performance Assessment Calculations Related to the Savannah River Site*; SRNL-STI-2009-00473. Rev. 1.; Savannah River National Laboratory: Aiken, South Carolina, 2016.
26. EPA, Method 3050B. Acid digestion of sediments, sludges, and soils. Revision 2. In *Test Methods for Evaluating Solid Wastes: Physical/Chemical Methods*, EPA, Ed. Environmental Protection Agency: Washington DC, 1996.
27. Knoll, G. F., *Radiation detection and measurement*. John Wiley & Sons: 2010.
28. Johnson, T., *Introduction to health physics*. McGraw Hill: 2017.
29. Gerward, L.; Guilbert, N.; Jensen, K. B.; Levring, H., X-ray absorption in matter. Reengineering XCOM. *Radiation Physics and Chemistry* **2001**, *60*, (1-2), 23-24.

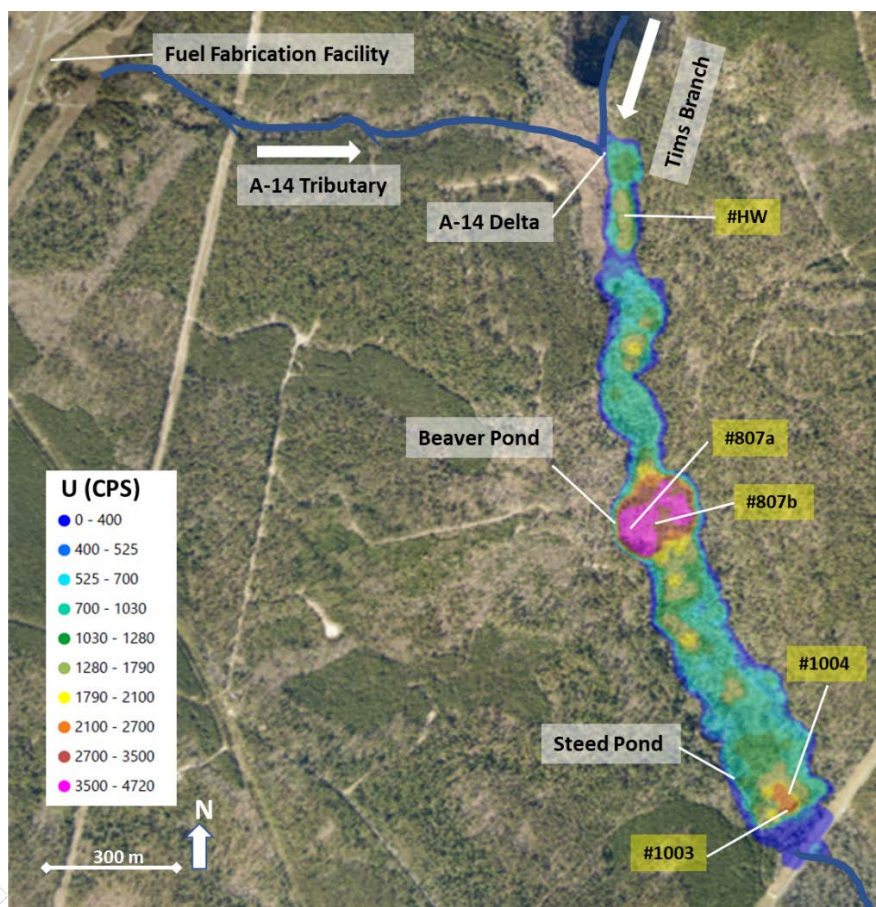
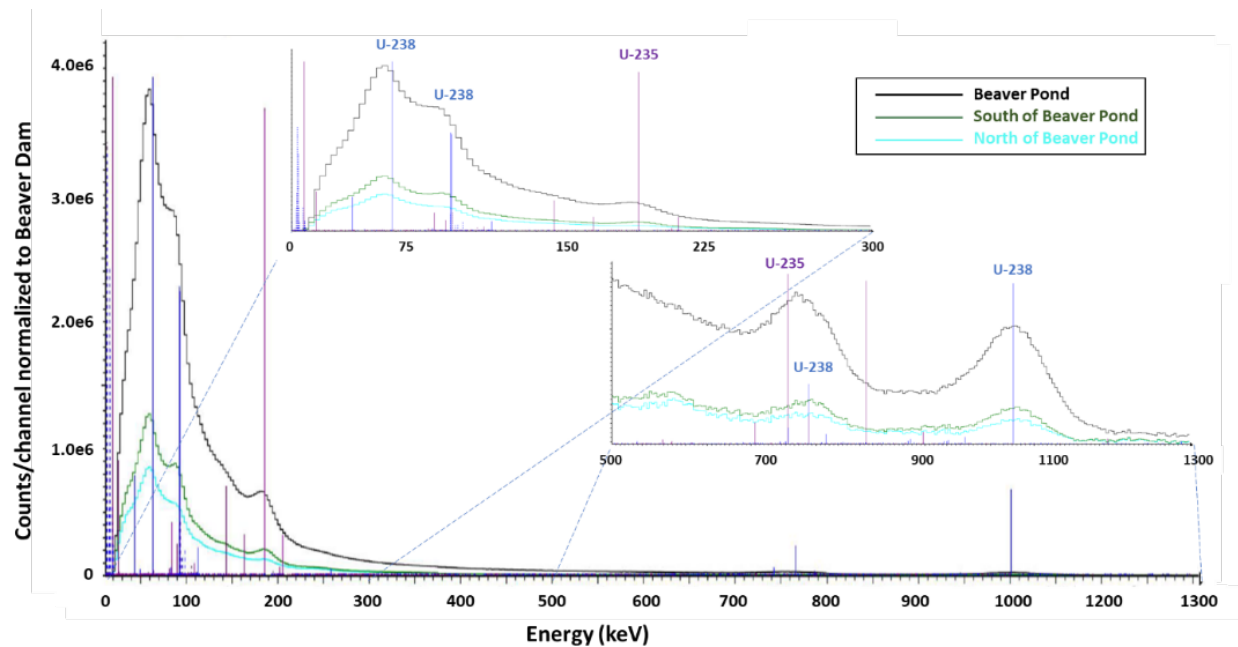
**Graphical Abstract**



**Table 1.** Estimates of uranium inventory in Tims Branch corrected for background U concentrations.

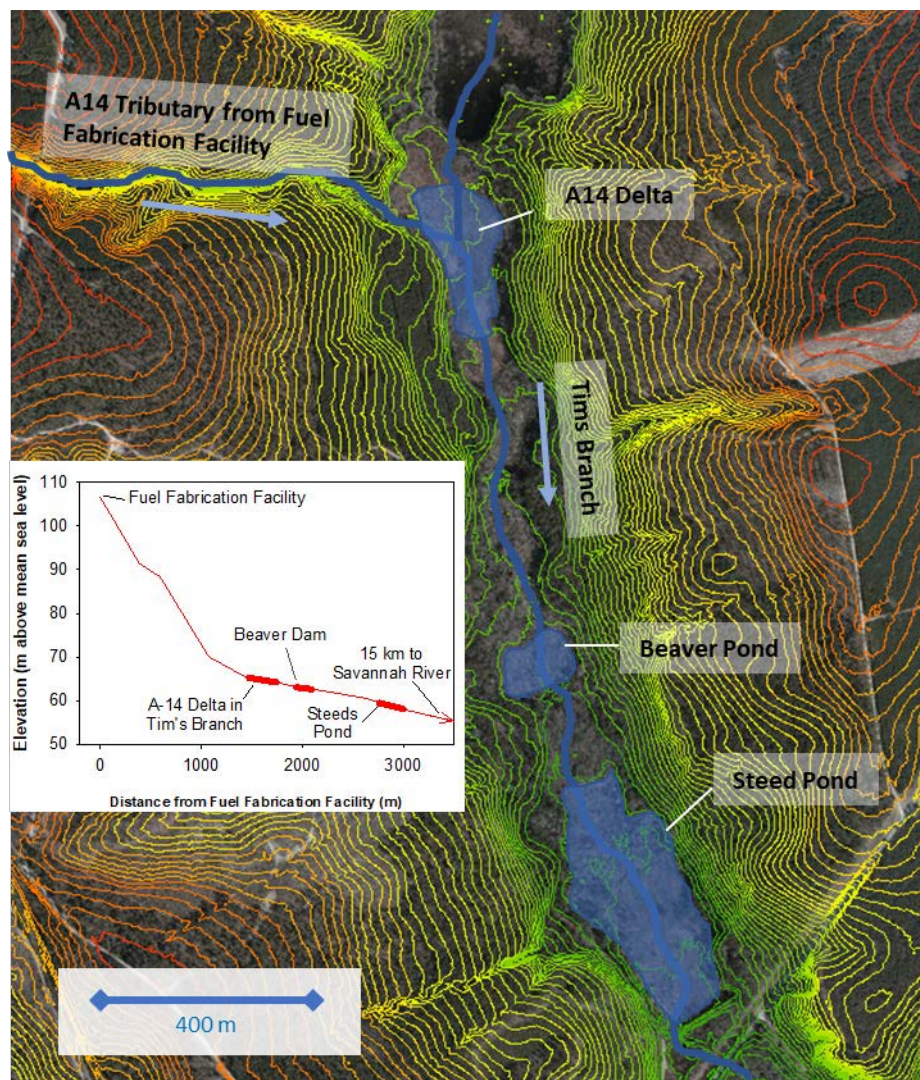
<b>Tims Branch Region</b>	<b>Area</b>	<b>Number of Spectral Acquisitions</b>	<b>Total U Based on Gamma-shielding</b>	<b>First Approximation of Total U</b>	<b>U mass per Area</b>
	<b>(m<sup>2</sup>)</b>		<b>Mg</b>	<b>Mg</b>	<b>kg U/m<sup>2</sup></b>
North of Beaver Pond	33,551	54,129	1.4 (4%)	2.1 (6%)	0.042
Beaver Pond	73,173	70,730	29.9 (83%)	29.8 (81%)	0.41
South of Beaver Pond	171,148	107,850	4.9 (14%)	4.9 (13%)	0.029
Total	277,872	232,709	36.2	36.9	0.13





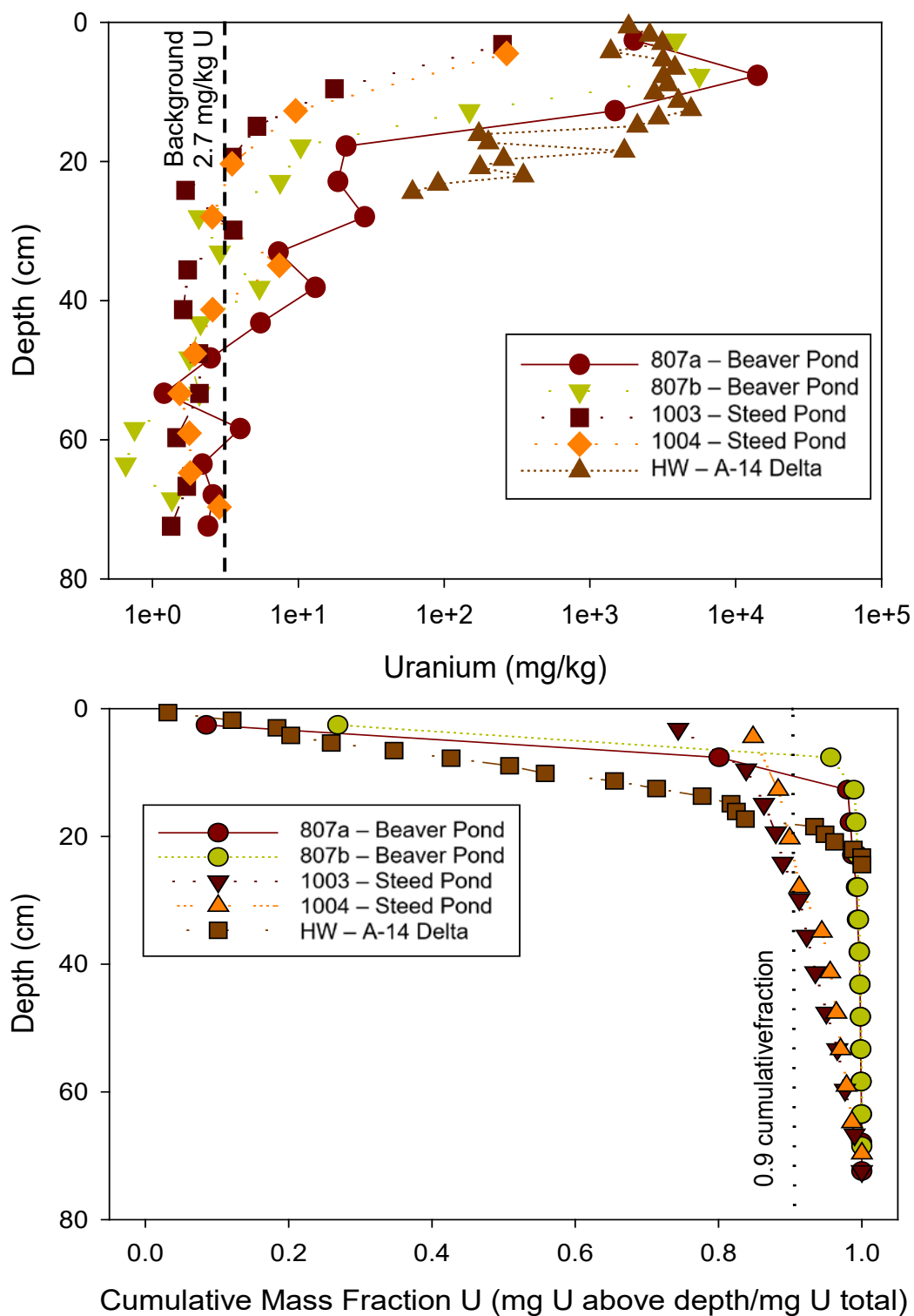
**Figure 1. (Top)** Tims Branch *in-situ* composite spectra for the Beaver Pond region (54,129 spectra) north of the Beaver Dam region (70,730 spectra) and south of Beaver Dam region (107,850 spectral acquisition. Composite spectra normalized to the former Beaver Pond region. **(Bottom)** Gamma map generated from 232,709 spectra, each with 1 second acquisition times collected over 277,872 m<sup>2</sup> in Tims Branch. Soil core locations identified in yellow boxes.

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**Figure 2.** 1-m Lidar elevation topography map of Tims Branch wetland area. Note steep gradient of A14 Tributary between Fuel Fabrication Facility and Tims Branch (3% grade) compared to shallower grade of Tims Branch (0.4%). A14 Delta, Former Beaver Pond, and Former Steeds Pond are depositional areas.

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**Figure 3. (Top)** Uranium concentration and **(Bottom)** cumulative mass fraction of U as a function of depth from five cores collected from Tims Branch wetland.