Bio-monitoring for uranium using stream-side terrestrial plants and macrophytes

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Abstract

This study evaluated the abilities of various plant species to act as bio-monitors for environmental uranium (U) contamination. Vegetation and soil samples were collected from a U processing facility*. The water-way fed from facility storm and processing effluents was the focal sample site as it represented a primary U transport mechanism. Soils and sediments from areas exposed to contamination possessed U concentrations that averaged 630 mg U kg^{-1}. Aquatic mosses proved to be exceptional accumulators of U with dry weight (dw) concentrations measuring as high as 12,500 mg U kg^{-1} (approximately 1% of the dw mass was attributable to U). The macrophytes (*Phragmites communis, Scripus fontinalis* and *Sagittaria latifolia*) were also effective accumulators of U. In general, plant roots possessed higher concentrations of U than associated upper portions of plants. For terrestrial plants, the roots of *Impatiens capensis* had the highest observed levels of U accumulation (1,030 mg kg^{-1}), followed by the roots of *Cyperus esculentus* and *Solidago speciosa.* The concentration ratio (CR) characterized dry weight (dw) vegetative U levels relative to that in associated dw soil. The plant species that accumulated U at levels in excess of that found in the soil were: *P. communis* root (CR, 17.4), *I. capensis* root (CR, 3.1) and *S. fontinalis* whole plant (CR, 1.4). Seven of the highest ten CR values were found in the roots. Correlations with concentrations of other metals with U were performed, which revealed that U concentrations in the plant were strongly correlated with nickel (Ni) concentrations (correlation: 0.992; r-squared: 0.984). Uranium in plant tissue was also strongly correlated with strontium (Sr) (correlation: 0.948; r-squared: 0.899). Strontium is chemically and physically similar to calcium (Ca) and magnesium (Mg), which were also positively-correlated with U. The correlation with U and these plant nutrient minerals, including iron (Fe), suggests that active uptake mechanisms may influence plant U accumulation.

*It is often difficult to gain access to contaminated sites for experimental monitoring on a voluntary basis. However, having access is essential to the advancement of environmental monitoring science. Privileged access was granted to us for the purposes of gaining a better understanding the dynamics of contamination at this site and for the advancement of monitoring science. In return, we were asked to not divulge the location of the site in this presentation.

1. Introduction

Recent environmental impacts from nuclear accidents have had a strong impression on the world's acceptance of nuclear power. There are multiple public safety concerns that have followed the traumatic incidences at the recent Fukashima Daiichi nuclear disaster in Japan and the Chernobyl reactor explosion in Pripyat, Ukraine. Responsible growth in this research field requires monitoring strategies that are capable of characterizing ecological and human exposure risks associated with all the anthropogenic processes involved in nuclear energy, including the mining, reprocessing and disposal activities related to nuclear fuel. Furthermore, effective methodologies for environmental remediation are required, when contamination is a result. Radionuclides present high profile hazards to the biosphere, but this has not yet provoked the extensive development of environmentally benign methods to extract them from soils.[1]

A developing monitoring strategy uses vegetation as radionuclide bio-monitors for potential contamination. Plants represent the exposure pathway of contamination moving from the soil through plants to humans, which can contribute significantly to the overall dose from radionuclides.[2] Radioactive contamination of vegetation may be highly dependent upon edaphic (soil) and climatic conditions. Therefore, measuring plant concentrations relative to soil concentrations provides insight into the bio-availability of radioactive contaminants.

Research on the levels of anthropogenic radionuclides in plants, such as cesium-137 (^{137}Cs), has been relatively extensive in contrast to that of uranium (U). This is somewhat due to the extensive global fallout of radiocesium from Chernobyl. However, at US Department of Energy (DOE) facilities, U is the most frequent radionuclide contaminant, with more than 50% of these facilities reporting U to be the primary contaminant in the soil and groundwater.[3] Uranium, at even natural levels, can provoke a radiation dose and chemical toxicity concern.[4] The biochemical toxicity of U is estimated to be six orders of magnitude higher than that of its radioactivity. When compared to other heavy metals, its chemical toxicity lies between mercury and nickel (Ni).[5] This is important as there have been reports of the high bioavailability of U to agricultural plants.[6,7] With the rising demand for U as a fuel source and for use in depleted U munitions, there is an increased need to monitor and characterize the occurrence of natural and technologically-modified forms of U in the environment.[8] Whereas assessments of soil or water for contamination provide information on limited spatial and temporal concentrations, terrestrial and aquatic plant sampling provides a mechanism for continuous, ubiquitous, *in-situ* sampling for purposes of environmental monitoring.

Plants have been used successfully in characterizing U contamination in the environment. In a previous study, the use of *Samucus nigra* as a bio-monitor provided more detailed information regarding U distribution than soil analyses alone. The plants not only indicated the location of U mineralization but

also the migration pathway of U-containing soil-water.[9] A previous study used bog-growing *Quercus velutina* as a bio-monitor to successfully determine by isotopic fractionation that a U contamination source contained depleted U. Results were considered to be a viable and inexpensive alternative to drilling wells to monitor for shallow groundwater U contamination.[10] Plants have also been explored as an economical approach to remediation of soils contaminated with heavy metals and radionuclides.[11,12] In a rhizofiltration greenhouse study, *Helianthus annuus* reduced the concentration of U in water taken from a DOE site (0.1-0.4 mg U kg^{-1}) by 95% within 24 hrs to help achieve the EPA groundwater standards of 0.03 mg U kg^{-1}.[13]

The bio-availability of U contamination is largely determined by mineralogical and physiochemical conditions.[14] In most soils, U exists primarily in the +6 oxidation state [U(VI)] as the uranyl [$U(VI)O_2^{2+}$] ion.[15] Oxidized U(VI) is the more mobile form of U as compared to the reduced U(IV) form. Soil pH will impact the solubility and thereby mobility of U by influencing its speciation: uranyl cation (acidic), uranyl hydroxides (neutral) and uranyl carbonates (basic). As U(VI) is transported via groundwater, uranyl will readily sorb to mineral sites, restricting transport. Increasing pH exposes more negatively-charged binding sites which would be expected to increasingly restrict U transport. However, increases in soil water pH are usually associated with increases in dissolved carbonate ion (CO_3^{2-}) which is the predominant complexant for environmental U.[16,17] Uranium can bond strongly to common groundwater species of CO_3^{2-} and Ca^{2+} to form stable dissolved complexes, which can compete with mineral surfaces, thus maintaining relatively high mobility of U.[16~~16~~,17~~17~~,18] Desired immobilization of U within such a contaminated zone can be further confounded by the inhibition of microbial reduction of U(VI) to U(IV) in the presence of Ca.[18~~18~~] Iron oxides and hydroxides with a range of structural crystallinity (amorphous, polycrystalline, and single phase) and many soil organic materials can be very important sorbents of U(VI) as well, thus reducing the bioavailability of U(VI).[19,20] Conversely, soil organic complexes and chelates can also increase soil U mobility.[2] These factors make it difficult to predict the bioavailability of U based on soil properties due to the complexity of possible interactions.[21]

The bioavailability of U is effectively quantified through examination of U concentrations in a biological component, such as vegetation. Consideration of plant behavior regarding U accumulation is required to characterize U behavior in the soil. Plants growing in neutral to basic soils have been shown to accumulate relatively higher U concentrations than plants grown in acidic soils.[15~~15~~,16~~16~~,22] Despite the uranyl ion being preferential to uptake by plants [as opposed to U(IV)], increased mobility of U ligands (carbonate and phosphate) enhances opportunities for root interception of U. Complexed forms of U in the soil have not been shown to hinder plant U uptake rates.[23] Because desorption of rhizospheric U complexes is not promoted in alkaline soils, a portion of the measured concentrations of U attributed to

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roots may include U adsorbed to the roots.[11] Other factors associating higher plant U concentrations with neutral to basic soils could also include: the increased variety of U speciation at near neutral pH thereby widening the range of plant U uptake opportunities; or the decreased competition with hydrogen for plant uptake sites in soils with higher pH.

Translocation of U within a plant is not fully understood and it varies by plant species. Regardless of pH, U will accumulate primarily in the roots.**Error! Bookmark not defined.**[23] Of the small portion translocated to the shoots, a higher proportion of shoot U is reported for plants growing in alkaline soils.[24] These results are only partially explained by higher concentrations of U in soil solutions of alkaline soil.[24][25] This indicates physiological barriers of U transfer to aerial portions of the plant that may be influenced by U speciation.[25] The discussed variables make CR values unique to each soil and plant and plant tissue combination making consistency of linearity with soil concentrations difficult to establish.[26] While not pursued in this study, it has been proposed that the likeliest example of a linear relationship would be established between U concentrations in roots and to a lesser degree, shoots of plants with soil pore-water.[27] While perhaps not representative of total soil contamination concentrations, dissolved radionuclides in the soil solution are mobile and their levels offer the ability to assess exposure risk to bio-available U. The use of available pore-water U instead of dw soil concentration in the calculation of CR values should also considerably reduce the variability within results from sites with differing soil properties.[28]

The objective of this study was to collect and identify plant species that would serve as bio-monitors for U contamination in the environment. We chose to examine the levels of U in various portions of the common biota that were collected at different locations that were potentially accessible to aerial as well as aquatic U contamination. We often sampled plants that had some documented history of U uptake. In some cases, we performed analyses of plant species that had little documented history of U accumulation to potentially identify new species for use as bio-monitors. Our study also examined the correlation between plant concentrations of U and other heavy metals. As the accumulation of certain metals may infer an ability to accumulate other metals, correlation results could serve as an alternative predictor for U accumulator species. The identified plants could potentially be utilized for research regarding the phytoremediation of U contamination in soils, sediments, and waters that are contaminated by low to moderate levels of radionuclides.

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2. Materials/Methods

2.1 Background Information on Sampling Areas

The facility has U contamination (most likely in the form of the highly soluble U(VI) species] that was hypothesized to have originated from the facility processing-water and the facility borne storm-water. Plant, soil, water and insect samples were taken from five sites exposed to facility borne U. The sample areas proximal to the facility are in the outfall area: the facility *processing-water outfall* (1), adjoining to and sharing a basin with the *storm-water outfall* (2) combining to drain the facility. Sources of plant U uptake have been known to include atmospheric deposition on aerial tissue.[26~~27~~] Because of this, terrestrial plants were sampled from immediately *above outfalls* (3). Approximately 300 m downstream of the outfall area were sample sites referred to as: the *stream outfall* (4), as well as samples from the *swamp* (5) that exists adjacent to and is hydraulically connected to the stream outfall. All sample sites were compared to a nearby *control site*, up-gradient of the facility, and thus not comparatively exposed to high levels of processing water or storm-water runoff from U activities at the facility.

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2.2 Sampling of Materials for Radiochemical Analysis

This study was intended to identify plant species that displayed the potential to accumulate U. As such, the number of species sampled was high, but the replication was low. A total of 17 native and non-native plants, considered abundant to the area were sampled in 2009. Samples included: **1) macrophtes:** aquatic mosses (unidentified, but assumed to all be the same), *Typha latifolia* (common cattail), *Phragmites communis* (giant reed), *Sagittaria latifolia* (broadleaf arrowhead), *Scirpus fontinalis* (bulrush) and **2) stream-side terrestrials:** *Bromus tectorum* (downy brome), *Sorghum halepense* (johnsongrass), *Setaria pumila* (yellow foxtail), *Cyperus esculentus* (yellow nutgrass), *Eupatorium rugosum* (white snakeroot), *Impatiens capensis* (spotted touch-me-not), *Urtica dioica* (stinging nettle), *Bidens aristosa* (tickseed sunflower), *Solidago speciosa* (showy goldenrod), *Diervilla sessilifolia* (bush honeysuckle), *Cardaria chalepensis* (lenspod white-top) and *Salix nigra* (black willow). When possible, species that were sampled from the U exposure sites were also sampled from the control site.

The insect *Chauliognathus pennsylvanicus* (Pennsylvania leatherwing) was sampled by hand. This species feeds primarily on the nectar and pollen of flowers, particularly *Solidago*. Incorporation of insects in this study was considered to potentially examine the bioaccumulation of U at higher trophic levels.

Plant root samples were repeatedly dipped in ambient water to detach soil and sediment aggregates from the roots. The washing was not thorough as it was our intent to maintain as many root hairs as possible. Conservatively, up to 10% of the total root mass analyzed could be attributed to soil or

sediment mass. All vegetation samples were dried to a constant weight at a temperature of 60°C and then ground to a particle size of 1 mm before analysis. The drying process caused more of the root adhering soil and sediment particles to detach. Soil and sediment samples were taken by a standard soil corer at each location from the root zone of an associated plant sample at a depth of 0 to 5 cm. Soil and sediment samples were dried to a constant weight at a temperature of 60°C and milled to a uniform particle size of <1 mm before analyses.

Radiochemical and stable element analyses were performed by GEL Labs, LLC (Charleston, SC). Soil and sediment pH was determined by electrometric measurement of a slurry, created by mixing a sample with reagent water, using the Environmental Protection Agency analytical protocol established in SW846 9045C/9045D. Vegetation and soil/sediment samples were analyzed after acid dissolution and chemical separation for U using liquid scintillation alpha (α) spectrometry. Activities of U isotopes were determined by alpha spectrometry applicable to method DOE RP 800 1997. Soil and vegetation samples were aliquoted and digested. The elements were then separated through ion exchange resins. The elements were then prepared for the measurement of U by coprecipitation with neodyminum fluoride. The neodyminum fluoride was trapped on a filter, mounted on a stainless steel disk and placed in a partially evacuated chamber for measurement of isotopic alpha emission during a four hour count time. To account for losses during separation, ^{232}U was used as a tracer. All activity data for these isotopes are presented in Bq kg^{-1} dry weight. Alpha spectrometry may be one of the most widely used methods for the determination of U. However, this method cannot resolve the energies of the α-particle emissions of ^{233}U, ^{234}U and ^{235}U, ^{236}U peaks due to their similar energies and this requires these two sets of isotopes to be reported as single values. Neither ^{234}U nor ^{236}U isotopes were expected to be present at this site as only natural U is handled at the facility. All isotopic activity data are presented as dry weight (dw) values in Bq U kg^{-1} dw (for soil data only). The typical method detection limit is 37 Bq kg^{-1}. Unlike the plant and insect samples, the soil U levels always exceeded the minimum detection limit (MDL) so they are reported here.

The analysis of total U and other metals was performed on a Perkin Elmer ELAN 6100E inductively coupled plasma mass spectrometer (ICP-MS). The instrument is equipped with a cross-flow nebulizer, quadrupole MS, and dual mode electron multiplier detector. Internal standards of scandium, germanium, indium, tantalum, and/or lutetium were utilized to calibrate the mass spectrum. Values for metals are expressed in mg kg^{-1} dw. Our comparisons were largely drawn from total U as measured by ICP-MS with instrument error calibrated to be less than 10%. Conversely, many of the uncertainties using α-spectrometry were much higher as the activity levels were often near the MDL.

This study anticipated the identification of plant species that were capable of bio-accumulating U at levels in excess of their soil concentration. This relationship was determined through calculation of a CR: a value representative of the dw concentration of U in the plant relative to the dw concentration of U in the associated soil. Unless linearity can be established, a CR value greater than one is desirable for a potential bio-monitor species. A regression analysis was performed on U with each element of: antimony (Sb), barium (Ba), cerium (Ce), cesium (Cs), chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), lead (Pb), lithium (Li), manganese (Mg), Ni, phosphorous (P), potassium (K), ruthenium (Ru), strontium (Sr), calcium (Ca), zirconium (Zr) and zinc (Zn). Results from this analysis provided correlation and r-squared values that were used to determine the relationships between plant uptake of these elements and the uptake of U.

3. Results

3.1 Soil-Uranium Characterization

Uranium concentrations for comparable surface soils in this area have a mean concentration of 3.4 mg U kg^{-1}.[29] As shown in **Table 1**, soil levels from contaminated sites had a mean of 630 mg U kg^{-1}, with a range between 291 to 1,460 mg U kg^{-1}. The highest U concentration from this study was measured at the outfall site of storm and processing-waters effluents. Concentrations of U from the control site were 26 mg U kg^{-1}. It is presumed that atmospheric deposition has contaminated this site beyond natural background levels.

The soil pH was 6.17 in the control site and a range between 7.72 and 8.64 in the sites exposed to contamination. It is expected that soil pH values above neutral will promote speciation of uranyl (UVI) carbonate species as well as U(VI) organic species (such as with humic and fulvic organic acids). These types of U(VI) species can interact strongly with algae and result in elevated plant uptake of soil U via the roots.[14,22,23,30] The soil solution in this pH range is typically dominated by bicarbonate ion and although we did not measure the soil organic matter or the levels of organic acids in the soil solutions, we observed high levels of surface organic material in the wetland areas that were part of this study. We anticipate the organic matter in the surface sediment materials in the swamp areas also had an effect on the speciation and retention behavior of the elevated levels of U that were previously observed. Although these soils and sediments were from swamp areas, we also noted a lack of highly reducing conditions due to the absence of volatile sulfides. Therefore, we do not suspect the elevated levels of soil U to be due to the presence of reduced forms of U [as the sparingly-soluble U(IV) species].

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3.2 Vegetation-Uranium Characterization

Although U is not essential or beneficial to plants, many species will absorb U and incorporate it into their biomass along with other metals. The concentration of U in a plant has been shown to be dependent upon the degree of U contamination in the soil.[31] To a lesser degree, it has also been observed that plant U concentrations vary between plant species independently of soil concentrations.[32,33] Plants exhibit a complex degree of control over the rhizosphere in the regulation of material acquisition from the soil.

Values for plant concentrations of mg U kg^{-1} are shown in **Table 2**. By statistically clustering total U levels for vegetation into groups of similar values we are able to relatively identify high, medium and low accumulators. The highest group contains the four aquatic moss samples and the roots of plants *P. communis* (outfall swamp) and *I. capensis*. Plant concentrations of U from this group were within the range of 1,250 – 1,030 mg U kg^{-1} dw. The second group contains the cluster: whole plants of *S. fontinalis*, and S. *latifolia* and the roots of *C. esculentus, P. communis* (stream outfall) and *S. speciosa*. Plant concentrations of U from this group were within the range of 286 – 148 mg U kg^{-1} dw. All remaining plant species and plant tissue parts are considered to be the low accumulators of this group. Plant concentrations of U from this group were within the range of 26.2 – 1.6 mg U kg^{-1} dw.

Each plant species from the high accumulator group was only sampled in areas of U facility exposure, providing no comparison with a species sampled from the control group. A previous study that found concentrations in moss from a background sample to be 33 mg U kg^{-1} dw.[34] The aquatic mosses had U values that ranged between 12,500 mg U kg^{-1} at the storm outfall, as compared to the more distal stream outfall site with concentrations of 2,480 mg U kg^{-1}. These values are comparable to U concentrations of 1,800 mg U kg^{-1} measured in the ash of aquatic mosses taken from a spring percolating through natural U deposits.[35] The study concluded that the sampling of mosses was more effective than the sampling of spring water for U as the ability of mosses to concentrate U and integrated U fluctuations that occurred in the spring water.

The *P. communis* root sampled from the outfall swamp also possessed a high U level of 5,050 mg U kg^{-1}. The concentration found in the upper portions of *P. communis* from the outfall site was in the low accumulator grouping, measuring only 12.9 mg U kg^{-1}, or an accumulation rate of 390 times less than concentrations from the roots. There has been some investigation into the removal of heavy metals, including U from contaminated waters via constructed wetlands using *P. communis*. With a relatively low water concentration of 0.37 to 2.44 mg U kg^{-1}, concentrations between 14 and 99 mg U kg^{-1} from whole plant samples have been previously measured.[36]

The roots of the *I. capensis* species were also comparable with the high accumulator group at 1,030 mg U kg^{-1}. The disparity between U concentrations in the roots relative to concentrations in the

upper portions was also large in *I. capensis* as the upper portions of the plant possessed a concentration of only 6 mg U kg^{-1} (171 times less).

From the second cluster of medium accumulators, there were two plant species that were whole plant samples: *S. fontinalis* and *S. latifolia* (415 and 161 mg U kg^{-1} respectively). Both macrophytes were sampled from the shallow wetlands of the outfall swamp. The roots of *C. esculentus*, *P. communis* and *S. speciosa* accumulate U in the roots at a respective rates of 3.7, 55.8 and 33 (respectively) times greater than that of the upper portions of the plant.

Some species or families of plants from the low accumulator grouping have been previously studied for U accumulations. Certain sunflower and mustard plants are known to accumulate high concentrations of U.[12~~12~~] Results are compared with the *B. aristosa* (of the Aster family) and *C. chalepensis* (of the *Brassicaceae* family), species used in this study. A previous study of the Aster *Helianthus annus* (sunflower) provided a range of soil U contamination levels applicable to both stream outfall and control soil contamination levels.[37] Unlike *H. annus*, concentrations of *B. aristosa* were considered low and did not reflect a linear relationship between soil and plant U concentrations. Uranium measured in *B. aristosa* at the control site had concentrations that were roughly half of those measured in *H. annus* for both root and above ground plant tissues. At the higher soil U concentrations of the stream outflow site, the levels measured in tissues of *B. aristosa* were less than a tenth of those measured in *H. annus* from a comparable soil concentration level. High U accumulations have also been reported in *Brassica juncea* with shoot concentrations of 22 mg U kg^{-1} dw from soil contamination levels of only 100 mg U kg^{-1} dw.[15~~15~~] Despite being located in the outfall site with a contamination level of 1,460 mg U kg^{-1}, *C. chalepensis* shoots only accumulated 4.7 mg U kg^{-1}.

The beetle *Chauliognathus pennsylvanicus* was compared with the upper portions of the *S. speciosa* sampled from the outfall site as this insect primarily feeds on the pollen and nectar of this plant species. The α-activity values for ^{238}U (actual isotopic data for all plants not shown) were about 1.5-fold greater in the insects (86.21±32.49 Bq ^{238}U kg^{-1}, which equates to 2.33±0.878 pCi ^{238}U g^{-1} or ~0.79 mg ^{238}U kg^{-1}) than were measured from the upper portion of the plant (47.36±25.86 Bq ^{238}U kg^{-1}, which equates to 1.28±0.698 pCi ^{238}U g^{-1} or ~0.435 mg ^{238}U kg^{-1}). However, few conclusions could be drawn from this observation due to the large error range of the α-spectrometric analysis (total U levels by ICP-MS were far below detection). Analysis from *S. speciosa* was below the MDL for $^{235,236}U$ while the insect sample was above MDL. This is interesting as the *S. speciosa* sample weighed about 180 g, while the *C. pennsylvanicus* sample weighed less than 10 g.

3.3 Influence of site on U contamination

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We could infer a spatial relationship to contamination from the U concentrations found in the aquatic moss samples that were taken from the outfall region as previously discussed. The highest concentration of U in moss was measured in the storm-water outfall area. The third highest moss concentration was measured in the processing-water outfall. Moss collected from where these two effluents mixed had the second highest U concentration. The moss sampled at the greatest distance from the contamination source was the stream outfall sample, which had the lowest U concentration of the mosses. These results suggest the primary source of U contamination could be the storm-water (i.e., run-off) and not the processing-water effluent.

Site selection for plants used to bio-monitor and phytoremediate U contaminated sites need consider the influence of lentic vs. lotic environments on the accumulation of U in vegetation. Results from a no longer operating mining facility have shown that plants grown in still water have increased U concentrations when compared to the same plants grown in moving waters.[38] We found examples that support the hypothesis of positively correlating water flow characteristics with plant U concentrations. Despite the U soil concentration in the stream outfall being similar or slightly greater than measured in the swamp outfall, the *P. communis* swamp outfall roots accumulated U at a levels that were 24 times greater than that in the sample from the stream outfall. Furthermore, a higher percentage of total plant U was accumulated in the upper portions of *P. communis* in the outfall swamp than in the stream outfall sample. Similarly, *T. latifolia* has been previously evaluated for U accumulation as it is a common species used for phytoremediation in wetlands. Results from a wetland study reported concentrations between 21 and 77 mg U kg^{-1}.[36~~37~~] In contrast, our study found a much lower concentration of 1.6 mg U kg^{-1} from the stream outfall.

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Bromus tectorum has been considered a desirable hazardous waste vegetative cover as it has a shallow root system with a low affinity for radionuclide uptake from the soil.[39] The plant species *B. tectorum*, *S. pumila* and *S. halepense* were sampled from terrestrial soils above the storm and processing water outfall pipes. Plants from these locations would have expectedly higher concentrations than those measured in control areas if sufficient atmospheric deposition of U from the outfalls effluent and/or from the facility occurred. Plants that were sampled above the outfall pipes had measured values of: 4.4 mg U kg^{-1} (*B. tectorum*) 3.4 mg U kg^{-1} (*S. pumila*) and 4.9 mg U kg^{-1} (*S. halepense*). These species sampled from the control area had respective values of 1.0, 0.2 and 0.3 mg U kg^{-1}. These findings support atmospheric deposition of U from outfall aerosol as a contributing factor in terms of a U contamination source.

3.4 Concentration ratios

Calculated CR values normalize the contaminant concentration in plants by using the contaminant concentration in the soil. These ratios are typically calculated for plants in biological monitoring studies. The results indicated that progressively increasing soil U concentrations corresponded with relatively decreasing concentrations of plant U (results of correlation analysis not shown). This is in agreement with the observed behavior of U in previously studied plant species and it suggests a saturation-type mechanism that is indicative of non-linear accumulators.2

Sediment U concentrations from just below the outfall were 56-fold greater than soil levels from the control site and roughly 3-fold greater than levels found elsewhere amongst the outfall sites. High U concentrations in outfall sediments produced substantially lower CR values for the plants sampled from this area. This did not affect the CR values of the samples with the highest U concentrations, as they were sampled elsewhere.

Using CR values, some plant species could be considered high U accumulators as concentrations from plant tissue were higher than those found in soil/sediment. These were: *P. communis* root (CR, 17.4), and *S. fontinalis* whole plant (CR, 1.4) from the swamp outfall and *I. capensis* root (CR, 3.1) from the stream outfall. The plant species *C. esculentus* (root) and *S. speciosa* (root) were both identified as medium accumulators from the section considering plant concentration yet CR values were amongst the highest in this study with values of 0.88 and 0.34 respectively. Concentration ratios support the consideration of these species as U bio-monitors. As expected, seven of the top ten CR values were found in the roots. The remaining three from the top ten plant CR values were the whole plant samples including: *S. fontinalis*, *S. latifolia* (CR, 0.55) from the swamp outfall and also *S. speciosa* (CR, 0.12) from the control site. The root of *B. aristosa* from the control site had a relatively high CR value of 0.33 but as soil U concentration increased, as in the stream outfall, the CR value decreased to 0.05.

Aquatic mosses lack true roots, but a projected CR that could be calculated based on the sediments associated with the habitats for the four moss samples averaged to be slightly higher than 6. A previous study found a CR value of 9 for aquatic mosses measured in an area with large concentrations of granitic U.[34~~34~~]

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3.5 Correlations of plant uranium with other metals

Beneficial and toxic metals often display similar chemical characteristics. Metal uptake by plants may be accelerated by roots during nutrient deficiency through the release of carriers or solubilizing agents such as organic acids and gelatinous mucilage to the rhizosphere.[40,41] These exudates and chelates can potentially complex with plant micronutrient metals like Fe, Mn and in doing so U may also be acquired.[42] As plant metal uptake mechanisms are similar, the ability to hyperaccumulate any one metal

may infer some ability to accumulate other metals.[43] Regression analysis of U and other metals in plant tissues provided correlations and r-squared values as shown in **Table 3**. The plant accumulated concentrations of Ni were very similar to U (correlation: 0.992; r-squared: 0.984). There was also a strong correlation between plant concentrations of Cu and U (correlation: 0.966; r-squared: 0.933). As neither U, Ni nor Cu are required mineral nutrients for plants, it can be assumed that the tight correlations are due to non-specific ion uptake mechanisms. Plant accumulated concentrations of Sr were also similar to U (correlation: 0.948; r-squared: 0.899). Strontium and Ca are chemically and physically similar alkaline earth elements, as such, plant uptake mechanisms cannot readily differentiate between Sr and plant required Ca. The correlation and r-squared values for Ca and U were 0.930 and 0.865 respectively. The six plant species with the highest accumulation of Ca were in the same ranked order as U. Regression analysis found a similar correlation among U and the other two alkaline earth metals analyzed: Ba and Mg (average correlation: 0.66; r-squared: 0.44). There was also a notable relationship between Fe and U plant concentrations (correlation: 0.783; r-squared: 0.613). As Ca, Mg (macronutrients) and Fe (micronutrient) are required by plants, the active attainment of these mineral nutrients by root systems appears to impact the uptake of U.

It has been proposed that a plant containing >0.1% of Ni, Co, Cu, Cr and Pb or 1% of Zn on a dw basis is a "metal accumulator", irrespective of the soil metal concentration.[44,45] The 0.1% criteria for Ni, Co, Cu, Cr and Pb and 1.0% Zn did not sufficiently discriminate such species as only a third of the sampled species had concentrations below these levels. By examining each metal independently, the top-third metal accumulators were identified. One set of species was identified as being among the highest accumulator of each metal of interest. These were also those species that had the highest concentrations of U: *I. capensis*, *P. communis* and the four aquatic moss samples. Results are displayed in **Table 4**. The exclusion of samples that were not in the top-third group improved the r-squared value in each instance aside from Co and Cr. This is to be expected as the typically low biological availability of these metals offers evidence of a plants status as metal accumulator.

The presence of phosphorous (P) in the soil has been shown to inhibit the bio-availability of U to vegetation.[46] The P levels from the swamp outfall soil were approximately 2.5 times greater than P levels from the stream soils. Yet, three of the top six CR values were from plants sampled at the swamp. If P does in fact exert influence on the availability of U, this finding supports the importance of water flow characteristics on plant U concentrations. When a plant has sufficient P, it is translocated from the roots to new leaves, possibly as a uranyl phosphate complex.[47] A significant regression was not established in this study between P and U from samples of the upper portions of plants.

4. Conclusion

Previous studies of U uptake by plants have focused primarily on field and garden crops important in the human food chain, or the U content of native plants growing in environments that have high U contamination.[16,33] This application uses plants as bio-indicators and bio-remediators of U in water and soil. Plants as bio-monitors are used to characterize the concentration and behavior of contaminants in the soil, including mobility and bio-availability. This information is useful in containment and remediation if an accident in an aboveground radioactive waste repository or in a nuclear fuel facility were to occur. Mechanical remedial processes such as ion exchange, reverse osmosis, microfiltration, precipitation, or flocculation are widely used to remove heavy metals from aqueous streams.[48] These methods, however, may be ineffective for large water volumes and low metal ion concentrations.[12] In stream waters, U is usually complexed with a ligand, such as carbonate, hydroxide, sulfate, phosphate, fluoride and possibly silicate.[49] These complexes increase the solubility of U and make U precipitation less likely. For these reasons, U is one of the best candidates for a biological monitoring and removal process.[12]

While roots are thought to accumulate more U than upper portions of plants, whole plants and upper portions of plants accounted for five of the top ten non-moss samples (that were analyzed for U): *S. fontinalis*, *S. latifolia*, *D. sessilifolia*, *U. dioica* and *C. esculentus*. This could be useful as it would allow for harvesting foliar portions of plants used in phytoremediation. Also, terrestrial dicots have been thought to bio-accumulate higher concentrations of U than monocots,2 yet the hydrophytic monocots (*P. communis*, *S. fontinalis* and *S. latifolia*) of this study were amongst the highest accumulators of all the species. Plant exposure to U was expanded in these hydrophytes to include foliar uptake of U derived from stream-water. Furthermore, when water flow rates were decreased in the outfall swamp, plant U concentrations were further increased. This would support the use of constructed wetlands to enhance phytoremediation.

The plant species tissues that accumulated U concentrations that exceeded those found in the soil were: *P. communis* (root, CR of 17.4), *I. capensis* (root, CR of 3.1) and *S. fontinalis* (whole plant, CR of 1.4). Nearly all of the U accumulated in *Phragmites* was found in the roots. This would require whole plant harvesting if *Phragmites* were to be used for U phytoremediation. The invasive nature of this hardy plant species may support such a remedial program.

There was little evidence that plants were linear accumulators of U. This is in agreement with the compilation review conducted by Sheppard and Evenden.2 Plants were shown to generally concentrate

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less U in their tissue as soil concentrations increased. The lowest CR values were found in the plants sampled above the outfall area where the highest soil U concentrations were found. Two plant tissue samples with U concentrations that approached unity with soil U concentrations were *C. esculentus* and *S. fontinalis*. There was only the one sample for the *S. fontinalis* species, but a second sample from the control site of *C. esculentus* showed extremely low plant U concentrations with a dissimilarly low CR value.

Mosses do not have roots, nor do they possess a vascular system, making them independent of their substrate. However, they are effective indicators of water quality and contaminant transport. Aquatic mosses possess a high capacity to rapidly accumulate and retain metals from solutions due to a large surface area, the absence of a cuticle in their tissues and an abundance of cation exchange sites on their cell walls. This, combined with a tolerance to elevated levels of organic and inorganic pollutants, makes these species good candidates for bio-monitors of U contamination.[50,51] Mosses also possess the ability to detect and differentiate between chronic and acute U exposure. Extracellular metal is easily absorbed and readily exchangeable with its environment reflecting current or sporadic spikes in environmental contamination.[52] Intracellular metal is not as easily exchangeable and therefore is more representative of average conditions.[53] While it has been hypothesized that the measured levels of U in terrestrial mosses may underestimate contamination concentrations due to adaptations that allow them to exist in highly contaminated areas, this relationship has not been observed in aquatic mosses.[54] If a CR value were calculated for mosses associated with the outfall and stream outfall areas, the average speculated value would be ~6.

Plants that displayed abilities to hyperaccumulate metals also had the highest concentrations of U. This provides another tool in the identification of potential plant species as bio-monitors. Plant concentrations of Ni and Cu with U were very strongly, positively correlated. As neither of these metals are plant mineral nutrients, this is evidence of non-specific metal uptake by plant roots. Also correlated with U were Fe and the alkaline metals of: Sr, Ca, Ba and Mg. While Sr is the only element of this group that is not required by a plant, it is chemically and physically similar enough to Ca that plants are unable to differentiate between the two. The ability to actively acquire essential plant elements appears to impact the uptake of metals such as U. This information may be useful in the further identification of plant species as U accumulators. These findings also project plant species that are effective in the phytoremediation of metals such as Cu and Ni to be equally effective in U phytoremediation.

5. **Acknowledgements**

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Table 1. Soil U concentrations by species and/or location. Alpha activity is in Bq U kg^{-1} dw. Those values in parenthesis were below the MDL are expressed as the determined MDL. Lab error values (as opposed to standard deviations) are also reported after the measured alpha. Total U values (by ICP-MS) had an error of 20%.

Soil Sample Description	**pH**	**^{232}U**	**^{233,234}U**	**^{235,236}U**	**^{238}U**	**Total U in mg kg^{-1}**
B. aristosa soil at control site	6.17	(22.46)	275.28 ± 55.13	(17.46 ± 99.90)	261.59 ± 53.65	26
S. latifolia soil at swamp outfall	7.72	(24.12)	3100.60 ± 208.31	142.45 ± 44.77	3037.70 ± 206.09	291
Outfall soil	8.64	(284.16)	17205.00 ± 1650.20	939.80 ± 388.50	19388.0 ± 1750.1	1460
S. speciosa soil at outfall	8.14	(23.16)	3418.80 ± 207.20	193.14 ± 49.21	3415.10 ± 206.83	441
Stream outfall soil	7.85	(33.86)	3700.00 ± 216.08	253.08 ± 56.61	3885.00 ± 220.89	327

Table 2. Plant U concentrations by species.

Plant Sample Description and Sampling Locations				**Contaminated Site Total U**		**Control Site Total U**	
Common Name	**Scientific Name**	**Plant Tissue¥**	**Collection Site**	**Total U mg kg^{-1}**	**Total U CR**	**Total U mg kg^{-1}**	**Total U CR**
Aquatic moss	Unknown	AP	Storm/process mix	9240	31.75	*No data*	
Aquatic moss	Unknown	AP	Process water	2900	9.97	*No data*	
Aquatic moss	Unknown	AP	Storm water	12500	42.96	*No data*	
Aquatic moss	Unknown	AP	Outfall	2480	7.58	*No data*	
Willow (black)	*Salix nigra*	AGP	Below outfall	2.27	0.00	0.34 (AGP)	0.01
Scirpus	*Scirpus fontinalis*	AP	Outfall swamp	415	1.43	*No data*	
Cattail (common)	*Typha latifolia*	AGP	Stream outfall	1.63	0.00	*No data*	
Sunflower (tickseed)	*Bidens aristosa*	AGP	Stream outfall	1.68	0.01	0.50 (AGP)	0.02
Sunflower (tickseed)	*B. aristosa*	RT	Stream outfall	16.00	0.05	8.55 (RT)	0.33
Arrowhead (broadleaf)	*Sagittaria latifolia*	AP	Outfall swamp	161	0.55	*No data*	
Reed (giant)	*Phragmites communis*	AGP	Outfall swamp	12.90	0.04	*No data*	
Reed (giant)	*P. communis*	AGP	Stream outfall	3.75	0.01	*No data*	
Reed (giant)	*P. communis*	RT	Outfall swamp	5050	17.35	*No data*	
Reed (giant)	*P. communis*	RT	Stream outfall	209	0.64	*No data*	
Goldenrod (showy)	*Solidago speciosa*	AGP	Below outfall	4.48	0.00	3.06 (AP)	0.12
Goldenrod (showy)	*S. speciosa*	AGP	Stream outfall	5.12	0.02	3.06 (AP)	0.12
Goldenrod (showy)	*S. speciosa*	RT	Stream outfall	148	0.34	*No data*	
Spotted touch-me-not	*Impatiens capensis*	AGP	Stream outfall	6.02	0.02	*No data*	
Spotted touch-me-not	*I. capensis*	RT	Stream outfall	1030	3.15	*No data*	
Nutgrass (yellow)	*Cyperus esculentus*	AGP	Below outfall	5.37	0.00	1.11 (AP)	0.04
Nutgrass (yellow)	*C. esculentus*	RT	Below outfall	286	0.88	1.11 (AP)	0.04
Honeysuckle (bush)	*Dieruilla sessillifolia*	AGP	Outfall swamp	26.20	0.09	0.37 (AGP)	0.01
Honeysuckle (bush)	*D. sessillifolia*	AGP	Stream outfall	2.50	0.01	0.37 (AGP)	0.01
Brome (downy)	*Bromus tectorum*	AGP	Above outfall	4.42	0.02	1.03 (AP)	0.04
Johnsongrass	*Sorghum halepense*	AGP	Stream outfall	10.30	0.03	*No data*	
Johnsongrass	*S. halepense*	AP	Outfall near	3.94	0.01	*No data*	
Johnsongrass	*S. halepense*	AGP	Above outfall	4.91	0.02	0.30 (AP)	0.01
Nettle (stinging)	*Urtica dioica*	AP	Stream outfall	25.70	0.08	0.77 (AP)	0.03
Nettle (stinging)	*U. dioica*	AGP	Control	*No data*		0.25 (AGP)	0.01
Nettle (stinging)	*U. dioica*	RT	Control	*No data*		2.8 (RT)	0.11
Lenspod white-top	*Cardaria chalepensis*	AGP	Below outfall	6.24	0.00	0.24 (AGP)	0.01
Lenspod white-top	*C. chalepensis*	AGP	Stream outfall	4.70	0.01	0.24 (AGP)	0.01
Lenspod white-top	*C. chalepensis*	AGP	Control	*No data*		5.48 (RT)	0.21
Snakeroot (white)	*Eupatorium rugosum*	AGP	Below outfall	4.13	0.00	*No data*	
Foxtail (yellow)	*Setaria pumila*	AGP	Above outfall	3.40	0.01	0.22 (AGP)	0.01

¥ (AGP): Above ground portions: (AP): All portions; (RT): Root.

Table 3. Regression of U and other metals, expressed with correlation and r-squared values. Correlation values for Li were insignificant and Ru levels were below detection and not regressed. Those plants with r-squared values >0.6 are shown in **bold**.

Significant at p<0.01	**Sb**	**Ba**	**Ce**	**Cs**	**Cr**	**Cu**	**Co**	**Fe**
Correlation	0.687	0.676	0.563	0.749	0.597	**0.960**	0.655	**0.783**
r-squared	0.472	0.457	0.317	0.561	0.356	**0.923**	0.429	**0.613**

Significant at p<0.01	**Pb**	**Mg**	**Ni**	**K**	**Sr**	**Ca**	**Zr**	**Zn**
Correlation	**0.879**	0.649	**0.992**	0.354	**0.874**	**0.930**	0.706	0.460
r-squared	**0.773**	0.421	**0.984**	0.125	**0.763**	**0.865**	0.499	0.212

Table 4. Regression of U with various metals for the plants that were the highest accumulators of Ni, Co, Cu, Cr, Pb and Zn.

Metal	**Nickel**		**Cobalt**		**Copper**	
	mg Ni kg^{-1}	*r-squared*	*mg Co kg^{-1}*	*r-squared*	*mg Cu kg^{-1}*	*r-squared*
Concentration Range	1,000 to 10,000	0.799	1,000 to 4,500	0.006	1,000 to 60,000	0.120
	>10,000	0.964	>4,500	0.044	>60,000	0.856
Metal	**Chromium**		**Lead**		**Zinc**	
	mg Cr kg^{-1}	*r-squared*	*mg Pb kg^{-1}*	*r-squared*	*mg Zn kg^{-1}*	*r-squared*
Concentration Range	1,000 to 30,000	0.045	1,000 to 30,000	0.006	1,000 to 80,000	0.183
	>30,000	0.013	>30,000	0.942	>80,000	0.891