1. Introduction

The nuclear fuel cycle generates a considerable amount of radioactive waste, which often includes nuclear fission products, such as strontium-90 (^{90}Sr) and cesium-137 (^{137}Cs), and actinides such as uranium (U) and plutonium (Pu). When released into the environment, large quantities of these radionuclides can present considerable problems to man and biota due to their radioactive nature and, in some cases as with the actinides, their chemical toxicity. Radionuclides are expected to decay at a known rate. Yet, research has shown the rate of elimination from an ecosystem to differ from the decay rate due to physical, chemical and biological processes that remove the contaminant or reduce its biological availability.[1] Knowledge regarding the rate by which a contaminant is eliminated from an ecosystem (ecological half-life) is important for evaluating the duration and potential severity of risk. To better understand a contaminants impact on an environment, consideration should be given to plants. As primary producers, they represent an important mode of contamination transfer from sediments and soils into the food chain. Contaminants that are chemically and/or physically sequestered in a media are less likely to be bio-available to plants and therefore an ecosystem.

It is widely accepted that the sorption capacity of a soil, particularly particle size distribution (sand, silt and clay percentages) and mineralogy, play a large role in the availability of radionuclides to plants.[2,3,4,5] However, these parameters are unable to account for the variations of concentrations found in different plant species.[6,7,8,9] Plants will often accumulate contamination as a consequence of mineral and nutrient attainment. Plants that have shown an ability to accumulate high concentrations of radionuclides have garnered recent interest for their potential to act as bio-monitors in helping identify areas of ecological and transport risk.[10] The purpose of this study was to reexamine established and identify potential new plant species that are active accumulators of radionuclides in terrestrial, lotic and lentic environments.[11,12,13] As such, this study took place in an area known to be impacted by nuclear processing: the Savannah River Site (SRS) in Aiken, South Carolina (SC). The SRS is a Dept. of Energy (DOE) site that produced primarily tritium and Pu used in the fabrication of nuclear weapons from 1952 to 1988. The SRS utilized natural streams, series of canals and reservoirs to disperse heat from nuclear reactors. As a result of routine discharges, routine leaks in the heat exchangers, and occasional leaks in fuel and core elements, over 50 radionuclides were released to the atmosphere, to onsite streams, and to seepage basins.[14] Since production was halted in the late 1980s, the primary mission of the DOE at the SRS has been environmental cleanup.

Macrophytes and stream-side terrestrial plants from areas impacted by operations were

selected for this study. These plants grow in edge habitats, potentially exposing a large diversity of fauna to contaminants. Also, streams present a primary mode of radionuclide transport at SRS. Temporal and spatial parameters make stream characterization particularly problematic for contemporary methodologies; whereas plant bio-monitors offer a mechanism for ubiquitous, random, *in situ* sampling of the water column and sediments for contamination.

This study identifies ten SRS plant species, considered to have well established populations at the SRS, for their ability to accumulate the following isotopes of interest: ^{137}Cs, ^{40}K, ^{238}Pu and ^{239,240}Pu. Radiocesium is of interest to this study as it is associated with all nuclear testing and some nuclear accidents. It is considered the radionuclide with the greatest health impact from the Chernobyl nuclear disaster of 1986. It is also one of the principal radionuclides released from SRS operations. Approximately 148 GBq of radiocesium have been released into the atmosphere and approximately 70,300 GBq of radiocesium have been released to seepage basins and streams, largely resulting from spills.[15] Most of the radiocesium that was released to on-site streams primarily adsorbed to stream or impoundment sediments near the point of release. Concentrations of Cs suspended in the water column are very low, with sediments retaining over 90% of the total inventory of aquatic systems. The soil of SRS tends to be sandy, low in clay, acidic and K depleted. The clay at SRS is usually kaolinite, which has a low level of ion exchange capacity but trace levels of interlayer chlorite-vermiculite are well known to fix Cs and K ions. Due to the K depletion in the soil and water at the SRS, high and persisting soil and sediment uptake of ^{137}Cs is facilitated.[16] These characteristics are expected to affect the bioavailability of ^{137}Cs.

Interest in ^{40}K stems from its chemical and physical similarity (hydrated radius) to Cs isotopes. As such, plants have difficulty distinguishing between K isotopes and the radioactive isotope ^{137}Cs. While K is required in large quantities by plants as a macronutrient the vast majority of K exists in relatively unavailable forms in the soil. Acquisition of K from a growing media against a concentration gradient necessitates active uptake processes. Plant metal accumulation of these monovalent ions is usually attributable to protein complexes located in the plasma membrane of root cells that function by specifically recognizing, binding, and transporting the metal of interest from the growing media into the cell. These active transporter or ion channels may range in specificity to the substrate, sometimes allowing similar metals such as Cs to bind to K transporters allowing for plant uptake.[17] Cesium and K uptake may occur through the soil or foliar absorption. Foliar absorption is a method of elemental (nutrient or non-nutrient) uptake by plants and lichens whereby gaseous uptake occurs via the stomata and the negative charge of the pectic material in the leaf cuticle

functions as a cation exchanger. While it might be expected that root uptake would be the principal mechanism for Cs absorption by macrophytes, some species absorb at least a portion of their K content from the water column.[18,19] Plants avoid K deficiencies through the excess consumption of K as soluble quantities become available. The implication is that a plant need not be deficient in K to remove ^{137}Cs from the air, water or soil when exposure occurs.[20] The highly plant mobile elements of K and Cs are easily translocated in the plant to areas of new plant growth. There is even evidence of root K being released into the sediments to displace ammonia to augment a plant's N supply.[21] Rooted macrophytes function as a link between bed sediments and the water column in element cycling. Despite the potential for K over consumption, it has been suggested that the uptake of ^{137}Cs is suppressed by the presence of available stable K and Cs in the soil.[7,22,23] This study will further investigate the ^{137}Cs and ^{40}K congener relationship for the SRS plants.

The interest in ^{238}Pu and ^{239}Pu stems from their extreme toxicity to mammals as well as their long half lives ($t_{½}$) of 87 and 24,000 yrs, respectively. Nearly 481 GBq of Pu have been released to onsite streams and seepage basins at SRS, while about 148 GBq have been released to the atmosphere.[14] Plutonium is known to form compounds, complexes, or alloy with virtually every other element. It is fixed by clay minerals and complexed by organic matter, decreasing its availability for uptake.[24] A study previously conducted, when SRS was a functioning facility, determined Pu uptake in a wheat crop occurs almost entirely (>95%) by physical foliar deposition and not root uptake.[25] In general, much of the early Pu plant uptake and translocation research had been conducted from a dosimetry standpoint, regarding transfer factors to humans, specifically from consumable portions of agricultural plants.[26] Yet, more recent research, focused on phytoremediation of Pu, was able to document uptake and translocation in two plant species.[27] While metal uptake by plants is often a combination of passive and metabolic mechanisms, plants exhibits a great deal of control over the rhizopshere with specific regards to the acquisition of highly insoluble nutrients. The release of root exudates such as organic acids and gelatinous material called mucilage by plant roots during growth can increase the solubility of essential nutrients (like iron (Fe), nitrogen, phosphorous, and magnesium (Mn)). Subsuquent chelation of iron oxide, Fe(III) impacts (to a lesser degree) the chemically and structurally similar Pu oxide Pu(IV), increasing the mobility of each in the soil[28] The movement of metals from the growing media into the cells walls is a passive process driven by diffusion or mass flow of cations that are attracted to the negatively charged root cell walls. The concentration gradient across the cell wall membrane facilitates transport of the metal into the cell. As an active process, the transport of Fe across the root plasma membrane occurs following

plant enhanced reduction, via a Fe(II) transporter (IRT1). This transporter might also facilitate the transport of heavy-metal cations.[29] An ability of the plant to mobilize Pu in soils and sediments, infers information regarding the solubility, mobility and bioavailability of Pu in the system. It may also have implications regarding bio-remediation in areas of low level contamination.

2. Materials/Methods

2.1 Background Information on Sampling Areas

The SRS consists of an 800 km^2 reservation, located at north latitude 33^o 14', west longitude 81^o 39' in Aiken and Barnwell counties of SC (United States of America). The climate of the area is moderate, having warm, humid summers and mild winters. Average daily temperatures range from 7.5°C in January to 27.1°C in July. Precipitation averages 120 cm annually and is generally highest in March and least in November. Evapotranspiration returns roughly 70% of incoming precipitation to the atmosphere.[30] Neighboring locations served as reference sites for this study. Monitoring for contamination in soils and vegetation on site and in neighboring areas has occurred for over 40 years. The SC Dept. of Health and Environmental Control has independently verified DOE findings since 1995. The reference site areas of Ellenton Bay and Tinker Creek were found to have alpha-emitting radionuclides (including ^{238}Pu and $^{239,240}Pu$) and gamma-emitting ^{137}Cs to be at levels deemed to be "low" and "consistent with background".[31] These reference sites are (as shown on the map of **Figure 1**) in relative proximity to the SRS, possessing similar soils and climates, but they were not impacted by site operations.

Sampling in this study occurred primarily around creeks and floodplains of third order tributaries of the Savannah River. Most soils in the basin are classified as Ultisols and are mapped in the Blanton-Lakeland soil association.[32] Because of their high sand content, the soils are well-drained, acidic and have low base saturation capacities.[33] Soils found in the streams are mapped in the Pickney Series (siliceous, thermic cumulic humaquepts) and are characterized as being poorly drained and acidic with a high organic content.[32] The dominant minerals in both soil types are quartz, feldspar, kaolinite, with some interlayered chlorite-vermiculite.[33]

The SRS sampling of contaminated sites occurred in three general locations: Fourmile Creek, Upper Three Runs and F-Area (**Figure 1**). F-Area no longer contains operational facilities, but sampling of plants known for atmospheric accumulation from this area provided insight regarding atmospheric contamination levels to be expected at nearby Fourmile Creek and Upper Three Runs. The majority of sampling occurred at two locations: Fourmile Creek and the water bodies created by

the damming of Upper Three Runs Creek. The Upper Three Runs Creek area includes: R-Canal, Pond A, Pond B and Par Pond. These water bodies formed the cooling basin for R-Reactor.

In 1967, there was a large spill of 55,500 to 74,000 GBq of ^{137}Cs into Fourmile Creek which is a likely source of the ^{137}Cs contamination.[15] But, there are also several SRS facilities draining into this watershed basin of approximately 57 km^2: C-Reactor, N-Area Central Shops, F, H, and E General Separation Areas and the Solid Waste Disposal Facility. Contamination in this basin has multiple sources and modes of pollution.

2.2 Sampling of Materials for Radiochemical Analysis

Sampling occurred during February, April, May, July and November of 2008. Growth rate and growth strategies are expected to influence a plants accumulation of nutrients and indirectly contaminants. Such a discussion is beyond the scope of this study. As these growth characteristics are influenced by climatic conditions, we attempted to account for these conditions by sampling (often the same plant) in multiple seasons.

Several plant species with populations common to SRS were sampled—including macrophytes, stream-side terrestrials, and epiphytes (shown in **Table 1**). The floating macrophytes that were sampled include: *Nymphaea odorata* (water lily) and the partially submerged *Utricularia inflata* (bladderwort). The emergent macrophytes that were sampled included: *Sparganium americanum* (bur-reed), *Carex* spp., *Arundinaria gigantean* (giant cane), *Juncus effusus* (*Juncus*) and *Typha latifolia* (cattail). The following terrestrial plants were sampled: *Pinus palustris* (longleaf pine), and *Myrica cerifera* (wax myrtle). The lichen *Cladina rangiferina* (reindeer lichen) and the epiphytic bromeliad, *Tillandsia usneoides* (Spanish moss) were also sampled. Some species were sampled in replications large enough for statistical analysis while other species samplings were considered to be survey samples used to identify potential species for future studies.

This study anticipated the identification of plants that were capable of bio-accumulation of radionuclides at levels in excess of their soil concentration. This relationship is determined by calculating a concentration ratio (CR), which represents the concentration of the radionuclide in the plant relative to the concentration of the radionuclide in the associated soil. The CR value reveals the ability of the part of a plant to bio-accumulate a contaminant from soil (or from sediment, in the case of rooted aquatic species) that can be compared with that of other plants. A CR value greater than 1 (equilibrium), is desirable for a potential bio-monitor species.

The roots of plants were rinsed with ambient water upon sampling. This rinsing was not thorough as the intent was to maintain an intact root system as well as detach large, readily removable soil and sediment aggregates. Aerial portions of plants were not washed. This protocol expectantly confounded the assessment of contaminant transfer within a plant. An objective of this study was to increase the understanding of contaminant uptake potentials of plants as it related to plant species. Multiple plant species were usually sampled from the same sites allowing us to differentiate contamination in plants as it related to species under similar atmospheric exposure conditions. Our study also assessed the bioavailability of contaminants with regard to their potential for food chain transfer in a contaminated area. This information could not be obtained from washed leaves. It was presumed that a study to evaluate the transfer of contaminants from the roots to the leaves would be best evaluated in a lab setting where aerial deposition and absorption could be controlled. Atmospheric surveillance of SRS from the year preceding sampling found no detectable gamma or alpha radiation levels that exceeded normal background levels.[31] Environmental monitoring of SRS process area stacks in the year preceding sampling measured releases of 0.044 GBq of ^{137}Cs and 0.043 GBq of total Pu.[34] These values are consistent with measurements from the previous four years examined.

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. Composite soil and sediment samples were taken at each location from the root zone of an associated plant sample at a depth of 0 to 5 cm. This depth attempted to capture the primary zone of expected contamination for the radioisotopes of interest. Soil and sediment samples were dried to a constant weight at a temperature of 60°C and milled to a uniform particle size of less than 1 mm before analyses. Stream water was not sampled for analysis. An Environmental Report compiles annual monitoring and surveillance activities at the SRS. Transport of Pu in site streams is no longer quantified because of the historically low levels and ^{137}Cs is not detectable in stream discharges. An extremely large sample would have to have been taken to achieve analysis above the MDA (minimum detection activity) levels.

Radiochemical and stable element analyses were performed by GEL Laboratories, LLC (Charleston, SC). Vegetation and corresponding soil (or sediment) samples were analyzed after acid dissolution and chemical separation for: ^{40}K, ^{137}Cs using gamma spectrometry applicable to method EPA 600/4-80-032 method 901.1. The gamma detector consisted of a high purity germanium detector. This method measures gamma photons emitted from radionuclides without separating them from the sample matrix. A typical detection limit is 0.1 pCi/g for a 100 minute counting time, with a

method precision (calculated by the typical relative percent difference) being less than 20%. Concentrations of ^{238}Pu and ^{239,240}Pu 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 exhange resins. The elements are then prepared for the measurement of Pu by coprecipitation with neodyminum fluoride. The neodyminum fluoride is trapped on a filter, mounted on a stainless steel disk and placed in a partially evacuated chamber for measurement of isotopic alpha emission. To account for losses during separation, Pu^{242} is used as a tracer. The typical method detection limit is 1 pCi/g. All activity data for these isotopes are presented in Bq kg^{-1} dry weight. The alpha spectrometry method cannot resolve the ^{239}Pu and ^{240}Pu peaks due to their similar energies, requiring these two isotopes to be reported in this study as a single value.[35]

2.3 Data Analysis

There were some samples that were below the MDA level. These values were excluded from the analysis, represented in the tables by an asterisk. The total number of samples analyzed for each species/location in a set is represented as *n* in the tables. The number reported as a subscript represents the number of samples that were above the MDA when different from *n.*

Upon receipt of radiological data, averages for the radionuclides of interest were compiled by plant species (**Table 2**). As many species were sampled in areas of varying levels of contamination, it was expected that there would be a range in values. Standard deviations values are included in parenthesis following values to characterize this variation. The mean values of species sampled from the contaminated site were separated using a Tukey multiple range test using SYSTAT 12 Statistical Software.[36] For this test, including samples taken from areas of greater or lesser contamination concentration made separation of means more difficult but better simulated situations where contamination concentrations may be largely unknown.

A statistical comparison of ^{137}Cs and ^{40}K concentrations by species studied any anticipated relationship of these isotopes; specifically, their ability to act as analogues. If applicable, high concentrations ^{40}K (or total K) would be expected to preclude the accumulation of radiocesium (or stable Cs).

3. Results and Discussion

3.1 Concentrations of Radionuclides by Individual Plant Species with Respect to Tissue

Plants in this study were able to clearly distinguish between contaminated and reference sites with regards to ^{137}Cs and Pu isotopes (**Table 3).** A comparison of all plant species from the contaminated sites is shown in **Table 2**. The two highest concentrations of ^{137}Cs were found in *J. effusus* root (5,010 Bq kg^{-1}) followed by the leaves of *J. effusus* (2,037 Bq kg^{-1}). The samples of *S. americanum (281* Bq kg^{-1} root and 195 Bq kg^{-1}leaves), *T. latifolia (702* Bq kg^{-1} root and 485 Bq kg^{-1}leaves) and *J. effusus* were all taken from sediments at the edges of Fourmile Creek. Of these macrophytes, *J. effusus* proved itself to be an accumulator of ^{137}Cs as roots and leaves possessed concentrations statistically greater than those found in the corresponding tissues of these proximally located plants.

Other high accumulators of ^{137}Cs were the macrophytes with floating leaves fully exposed to stream water; the one survey sample of *U. inflata* (3,922 Bq kg^{-1}) and *N. odorata* (1,653 Bq kg^{-1} roots and 1,493 Bq kg^{-1} leaves). One of the highest tissue concentrations of ^{137}Cs was measured in the *N. odorata* leaf from Pond B (3,090 Bq kg^{-1}). Sediments analysis showed this area to have the highest concentrations of ^{137}Cs (13,339 Bq kg^{-1}) for this study. Similar leaf concentrations of ^{137}Cs were found in *N. odorata* from Pond B in a study from 1983.[37]

Plants of the *Pinus* genera possess the ability to grow in nutrient poor soils through a strategy of nutrient retention, as opposed to the type of nutrient cycling that occurs through leaves in broadleaf forests. The *Pinus* genus has also been determined to have high Cs and K mobility within the plant.[38] Nutrient uptake strategies, combined with the large surface area of the leaf and year round plant activity have been attributed to the success of *Pinus* needles as successful bio-monitors for low level ^{137}Cs contamination.[39] The *P. palustris* sample from our study accumulated 283 Bq kg^{-1} of ^{137}Cs while growing in the Pond A area which possessed the second to lowest soil concentrations of ^{137}Cs (251 Bq kg^{-1}). The *P. palustris* sample growing in the nearby Pond B area accumulated 1,522 Bq kg^{-1} of ^{137}Cs while growing in a soil ^{137}Cs concentration of 858 Bq kg^{-1}.

The roots of sampled plants generally had the highest concentrations of ^{238}Pu and ^{239,240}Pu (as opposed to upper portions) (**Table 2**). Roots from *S. americanum, T. latifolia* and *J. effusus* statistically possessed the highest Bq kg^{-1} values for measured Pu isotopes. All other species were statistically similar. The highest individual Pu concentrations were found in the four *S. americanum* root samples from Fourmile Creek at Road 4, with average concentrations of ^{238}Pu 678 Bq kg^{-1} and ^{239,240}Pu concentrations of 61.6 Bq kg^{-1}. These levels are approximately ten times greater than Pu levels measured in the sediment. While we don't have sediment samples associated with *T. latifolia*

or *J. effusus* from this sample location, Pu concentrations in these roots are roughly four to five times greater than the soil sample from *S. americanum*. Despite a suspected limited uptake potential, Pu appears to concentrate within and/or on the outside of these roots, thus making Pu more bio-available than sediment concentrations may suggest.

Plutonium analysis was primarily performed on samples from Fourmile Creek as this is an area of suspected exposure. Both of the species analyzed for Pu from the R-Reactor basin (*N. odorata* and *P. palustris*) were below the MDA. While ^{238}Pu and ^{239,240}Pu were below the MDA for *P. palustris* needles in this study, Pu concentrations have been reported as high as 68.1 Bq kg^{-1} of ^{238}Pu and 39.6 Bq kg^{-1} of ^{239,240}Pu for pine needle litter.[40] This older study was conducted while the SRS was still a functioning Pu production facility and they support the importance of aerial deposition of Pu for the *Pinus* genus.

A plant species that was not discussed previously due to lack of sample replication is *U. inflata*. Only one sample was taken from a contaminated site, but concentrations from this highly reticulate plant were extremely large for ^{137}Cs (3922 Bq kg^{-1}), ^{238}Pu (803 Bq kg^{-1}), and ^{239,240}Pu (862 Bq kg^{-1}). These rootless plants can become established in oligotrophic waters due in large part to their association with periphyton; including obtaining nutrients through carnivory. The autotrophic nature and large surface to volume ratio of periphyton facilitates the concentration of certain radionuclides by several orders of magnitude over water concentrations.[41] A previous study found *U. inflata* to be a high foliar accumulator of Cs (^{133}Cs was used as the contaminant spike) as compared to other macrophytes.[42] Further investigation is warranted to validate concentrations found in our study, and if accurate, assess the impact of these concentrations on the ecosystem.

3.2 Correlations between ^{40}K and ^{137}Cs in Plants

Among all plant species sampled, a regression analysis was performed that used plant concentrations of ^{40}K as a predictor for plant concentrations of ^{137}Cs. It was significant at the 0.05 confidence level, but the R^2 value is very low as some plants accumulated very little of both ^{40}K and ^{137}Cs. However, no plant sample possessed high concentrations of both ^{40}K and ^{137}Cs. The data generally showed that ^{40}K and ^{137}Cs concentrations exhibited an inverse relationship in the plant. As examples, *S. americanum* leaves possessed the highest concentrations of ^{40}K and the lowest concentration of ^{137}Cs for rooted plants. The highest concentrations of ^{137}Cs were found in the *J. effusus* root; however, ^{40}K concentration of the *J. effusus* root was statistically amongst the lowest concentration of levels observed.

The analysis run on plant and soil samples also included an assessment of total K. This was useful as most ^{40}K soil/sediment samples were below the MDA. Results were fairly uniform aside from two exceptions. Associated soils of *P. palustris* possessed total K concentrations that were two and a half times lower than the average K concentration of all the soils from the study. *Pinus palustris* proved to be strong accumulator of K in that it was able to deplete soil K. Another example is *S. americanum*; at a minimum, total K plant concentrations were two times greater than levels found in any other sample. The high CR values for both ^{137}Cs (1.59) and K/^{40}K concentrations have identified a plant with strong K congener uptake tendencies. For this study, K measurements were most useful in assisting of the identification of strong ^{137}Cs accumulators.

3.3 Concentration Ratios for Cs and K

The use of CR values is often criticized as it suggests that the variation of contaminants within plants is completely explained by the variation in the soil concentration.[43,44] While CR values may not be dependably used to infer a soil concentration from the analysis of a plant concentration, CR values are useful in determining the bio-availability of soil contamination. The plant tissue samples that possessed the highest ^{137}Cs CR value was the *P. palustris* needles (2.18), the leaves of *S. americanum* (1.59), the roots of *T. latifolia* (1.46) and the whole plant of the sedge *Carex* spp. (1.37) as shown in **Table 4**. In a previous study conducted at SRS, the sedge *Cyperus spp.* (similar to the sedge *Carex* spp.) possessed amongst the highest CR value for ^{137}Cs (24.6).[12] When only plant concentrations of ^{137}Cs were considered, *S. americanum* samples ranked as below average accumulators, yet consideration of CR values identified this species for potential use as bio-monitors of bio-available ^{137}Cs. As shown in **Table 5**, there was also a strong linear correlation between ^{137}Cs measured in the *P. palustris* needles with values measured from associated soil (R^2=0.76, p=0.006). From another study, soil and needle concentrations were comparable to those measured at SRS (soil with 176 and 298 Bq kg^{-1} of ^{137}Cs and needles with 163 and 504 Bq kg^{-1} of ^{137}Cs).[45] Correlating these two sites from the previous study, with the averages from the two SRS sites at Pond A and Pond B reveals a significant R^2 value of 0.992. The data supports the use of *P. palustris* needles as a bio-monitor for ^{137}Cs and suggests a potential to act as a predictor for soil concentrations.

The ^{137}Cs CR value for *T. latifolia* leaves was 0.87. *Typha latifolia* is discussed in the literature as a possible ^{137}Cs indicator species as it has shown to predictably reflect the concentrations in the components of the food web and/or ecosystem.[13] In a previous three-year study, the CR for *T. latifolia* leaves were very similar (0.7, 1.8) with the third year being considerably higher (75.2).[12]

While a CR value near 1 may be low, it reflects the contamination level of the soil, maintaining the importance of this species as a potential ecological bio-monitor.

There were few CR values for ^{40}K as there were only a total of eight soil or sediment samples with concentrations above the MDA. Six of the eight were from soil associated with *M. cerifera* samples. This is of note as there are only two remaining detectable ^{40}K concentrations in the study (*S. latifolia*). Concentrations of ^{40}K and ^{137}Cs in *M. cerifera* are statistically amongst the lowest for all species. While this would support the conclusion that *M. cerifera* is not an active accumulator of ^{137}Cs, the low CR values could also be indicative of a low bio-availability of terrestrial K congeners. This will be explored further with regards to the influence of site on ^{137}Cs concentration.

3.4 Concentration Ratios for Pu

Despite soil concentrations of ^{238}Pu generally being an order of magnitude higher than soil concentrations of ^{239,240}Pu, the CR values of ^{238}Pu and ^{239,240}Pu were similar within species. This is in agreement with a previous study that determined plants to be indiscriminate in the uptake and absorption of ^{238}Pu and ^{239,240}Pu.[46] A previous study determined that Pu concentrations in the above ground biomass of plants did not reflect soil contamination concentrations, meaning the CR value of Pu would be expected to decline as the soil concentration increased.[47] The leaves of *S. americanum* appeared to be an exception in this study. *T. latifoli*, *A. gigantean*, *J. effusus* and *S. americanum* were all sampled from the same area along Fourmile Creek (Road 4), but uniquely, *S. americanum* possessed leaf concentrations that were approximately twenty times greater than levels found in these other proximally located plants. The CR value for the five *S. americanum* samples from Road 4 was 3.6 while the CR value for the two *T. latifolia* leaves with associated sediments was 0.03. For all *T. latifolia* samples, the CR value was 0.02, and seemed unrelated to sediment Pu concentrations. The CR for *S. americanum* leaves from all sample sites averaged 1.75 for ^{238}Pu and 2.90 for ^{239,240}Pu. Leaf samples from this species were correlated with Pu values measured in corresponding sediments for ^{238}Pu and ^{239,240}Pu (0.73 and 0.59 respectively). The linear relationship respective to R^2 values for ^{238}Pu was 0.536 (p=0.025) and 0.346 (p=0.096) for ^{239,240}Pu. In this study, plant concentrations of Pu increased as sediment concentrations increased.

All of the macrophytes were able to accumulate detectable levels of Pu in their roots and rhizopshere. The highest CR values for Pu were found in the *S. americanum* roots with values of 5.86 for ^{238}Pu and 4.84 for ^{239,240}Pu. The CR value for *T. latifolia* roots improved dramatically from those found in the leaves (1.74 for ^{238}Pu and 1.6 for ^{239,240}Pu). Assuming uptake of Pu by the roots of

these macrophytes, only *S. americanum* was able to also translocate Pu within the plant. Therefore, the leaves of *S. americanum* were more representative of the bioavailability of Pu than were concentrations measured from the sediment. If Pu was not translocated within the plant, than *S. americanum* has proven itself to be an effective bio-monitor for assessing aquatic Pu transport while the other macrophytes were not.

The CR value of 5.13 for *Carex spp.* does not accurately characterize ^{238}Pu uptake for this species. There was only one plant sample above the MDA for ^{238}Pu. This associated sediment sample possessed a concentration that was almost a 100% less than the average of the other two sediments.

3.5 Concentrations of Radionuclides by Plant Species with Respect to Sample Site

Past studies have shown *M. cerifera* to accumulate large concentrations of ^{137}Cs (approximately 155,500 Bq kg^{-1} in 1971 and 2,200 Bq kg^{-1} in 1989).[12] While natural decay and physical transport has considerably lessened the ^{137}Cs inventory at SRS, the capacity to accumulate ^{137}Cs is apparent. Despite inhabiting soils that were amongst the highest in ^{137}Cs concentration for this study, the *M. cerifera* leaf samples from R-Canal possessed amongst the lowest leaf concentrations of ^{137}Cs (169 Bq kg^{-1}). The trend of disparity between leaf concentrations and soil concentrations for *M. cerifera* leaves continues with samples from Par Pond and Pond A (respectively for leaves: 42 and 31 Bq kg^{-1} of ^{137}Cs). The exceptions are the six leaf samples from Pond B with an average ^{137}Cs concentration of 673 Bq kg^{-1}. These are concentrations that are more than eight times greater than the combined average concentrations for leaves from R-Canal, Par Pond and Pond A. Pond B also possess the lowest ^{40}K concentration of soils associated with *M. cerifera* as only one of five samples was above the MDA. Similarly, the average CR value for the five *P. palustris* needle samples from Pond B was 3.1 whereas the CR value for the six samples from Pond A area was 1.5. Atmospheric deposition is unlikely to be a significant factor as all sites mentioned here are in the R-Reactor basin with Pond B being located somewhat near the center of R-Reactor Basin sampling. From these results, unidentified soil properties have made ^{137}Cs relatively more bio-available in the Pond B area.

The concentrations of ^{137}Cs measured in the sediments of Pond B are approximately twice those of Fourmile Creek. This could be expected as sedimentation and transport is not occurring in the pond at the same rate as in the creek. A comparison of plants between sites is difficult as the species are not duplicated between sites, but it is worth noting that some of the highest plant

concentrations of ^{137}Cs (Juncus root and leaves being the highest in the study) were from Fourmile Creek. This suggests that ^{137}Cs may be more bio-available in Fourmile Creek than sediment analysis would suggest.

With respect to site, Pu concentrations in the roots and leaves of *S. americanum* sampled from Road 4 of Fourmile Creek exceeded those found in associated sediments. Plant tissue samples, approximately 1 km downstream, decreased four times faster (on average) than sediment concentrations. The rapid decline of Pu in plant tissues could be a function of the bio-availability of Pu or it may reflect an accumulation curve that is steeper than a linear accumulation curve. Under either condition, the concentrations ^{137}Cs and Pu measured from sediments did not represent the true potential exposure risk for the ecosystem.

Samples taken from F-Area include: *C. ranigernia, T. usneiodes* and *P. palustris*. Each of these species is considered to be an atmospheric accumulator of contaminants of interest.[23,48,49] The concentrations measured were slightly higher than those measured in the reference site, yet were all very low relative to samples from Fourmile Creek and R Reactor Basin. F-Area is no longer an operating facility, but it's proximity to Fourmile Creek provides an indication of the low concentration of background atmospheric contamination in the area.

4. Conclusion

Many identified plants accumulated concentrations that were greater than those found in the soils and sediments due to an ability to forage for minerals, nutrients and consequently contaminants. Plants provided insight into the ecological exposure risk and bioavailability of contaminants that could not be assessed by only measuring soil/sediment concentrations. The following macrophytes from this study were identified as high foliar accumulators of ^{137}Cs: *J. effusus* and *N. odorata*, (respectively: 2037, 1493 Bq kg^{-1} of ^{137}Cs). It is important to note that these species were only sampled in highly ^{137}Cs contaminated areas. There were no sediment samples associated with *J. effusus*, however, from a previous study at SRS, the ^{137}Cs CR for *J. effusus* leaves was determined to be 9.3.[12] *Juncus* has also been previously considered to be an accumulator of ^{137}Cs in a study that examined phylogeny and growth stages as it relates to ^{137}Cs concentrations found in plants.[50] This species warrants further investigation as it possessed exceptionally high root concentrations of ^{137}Cs (5010 Bq kg^{-1}) as well as ^{238}Pu (237 Bq kg^{-1}) and ^{239,240}Pu (22.6 Bq kg^{-1}). While the ^{137}Cs concentration of *P. palustris* needles and *S. americanum* leaves were relatively average, the CR values were 2.18 and 1.59 respectively. Both species proved to be strong accumulators of K

416 congeners as analysis of total K determined that *P. palustris* had depleted soil K inventories, while *S.*
417 *americanum* possessed the highest total K leaf concentrations of all species in the study. In an area
418 with low sediment concentrations of ^{137}Cs, two samples of *S. americanum* accumulated ^{137}Cs
419 concentrations nearly six times greater than those of associated sediments. This could have
420 implications for fauna in the diversity abundant, edge ecosystem that macrophytes inhabit.
421 The highest concentrations of Pu in this study were found in the roots of macrophytes, in
422 particular, *S. americanum*, *T. latifolia* and *J. effusus*. The roots of these samples were not extensively
423 washed, so some of Pu contribution included associated sediments. Yet, Pu CR values for *S.*
424 *americanum* and *T. latifolia* show these species to have accumulated concentrations that were above
425 equilibrium with associated sediments (5.4 and 1.7 respectively). The standard deviation for *S.*
426 *americanum* CR values is high, but this is a result of varying soil concentrations. The correlation
427 between *S. americanum* roots and sediment for ^{238}Pu was (1.0) and the regression R^2 (0.99) was
428 significant (0.001). The regression analysis for ^{239,240}Pu also showed a strong correlation (0.98) and
429 the R^2 (0.96) was significant (0.02) (**Table 5**).
430 As was true with ^{137}Cs, sediment concentrations do not reflect the bioavailability of Pu. The
431 average CR value for foliar *S. americanum* was 2.17 for all Pu. Internal transport of Pu between
432 roots and shoots is considered to be minimal yet the CR value for the leaves of *S. americanum* were
433 approximately twenty times greater than the concentrations measured in other macrophytes from the
434 same sample area. This would identify *S. americanum* as possessing the ability for uptake and
435 translocation of Pu within the plant. Sediment concentrations associated with *S. americanum* were
436 lower than other proximally located sediment samples. This could have implications regarding
437 phytoremediation and also transport potential if leaf uptake and senescence is occurring.
438 With respect to site, as sampling of *S. americanum* proceeded downstream, plant tissue Pu
439 concentration declined more rapidly than did the rate of sediment Pu concentration. Regarding the
440 impact of site on ^{137}Cs, *M. ceriferia* leaves accumulated approximately four times more ^{137}Cs at Pond
441 B than were measured from R-Canal. Yet, some of the highest concentrations of soil/sediment ^{137}Cs
442 were sampled from R-Canal. There are many factors that may create such discrepancies:
443 contaminant bio-availability, influences of terrestrial, lentic and lotic environments, non-linear
444 contaminant accumulation are but a few. It was not the intent of this study to determine the reasons
445 for differential accumulation. Instead, results have shown that concentrations of contaminates, as
446 measured in soils and sediments, do not sufficiently characterize the exposure risk in an ecosystem.
447 To assign such meaning to contamination values, we must measure contamination in the biota.

Plants, as primary producers, offer unique insight into the important transfer pathway of contamination from soils and sediments into the food chain.

Figure 1. Map of Savannah River Site (Aiken, SC). Contaminated and reference sample sites are identified. Single, smaller sized capital letters represent various areas onsite, such as "F" for F-Area.

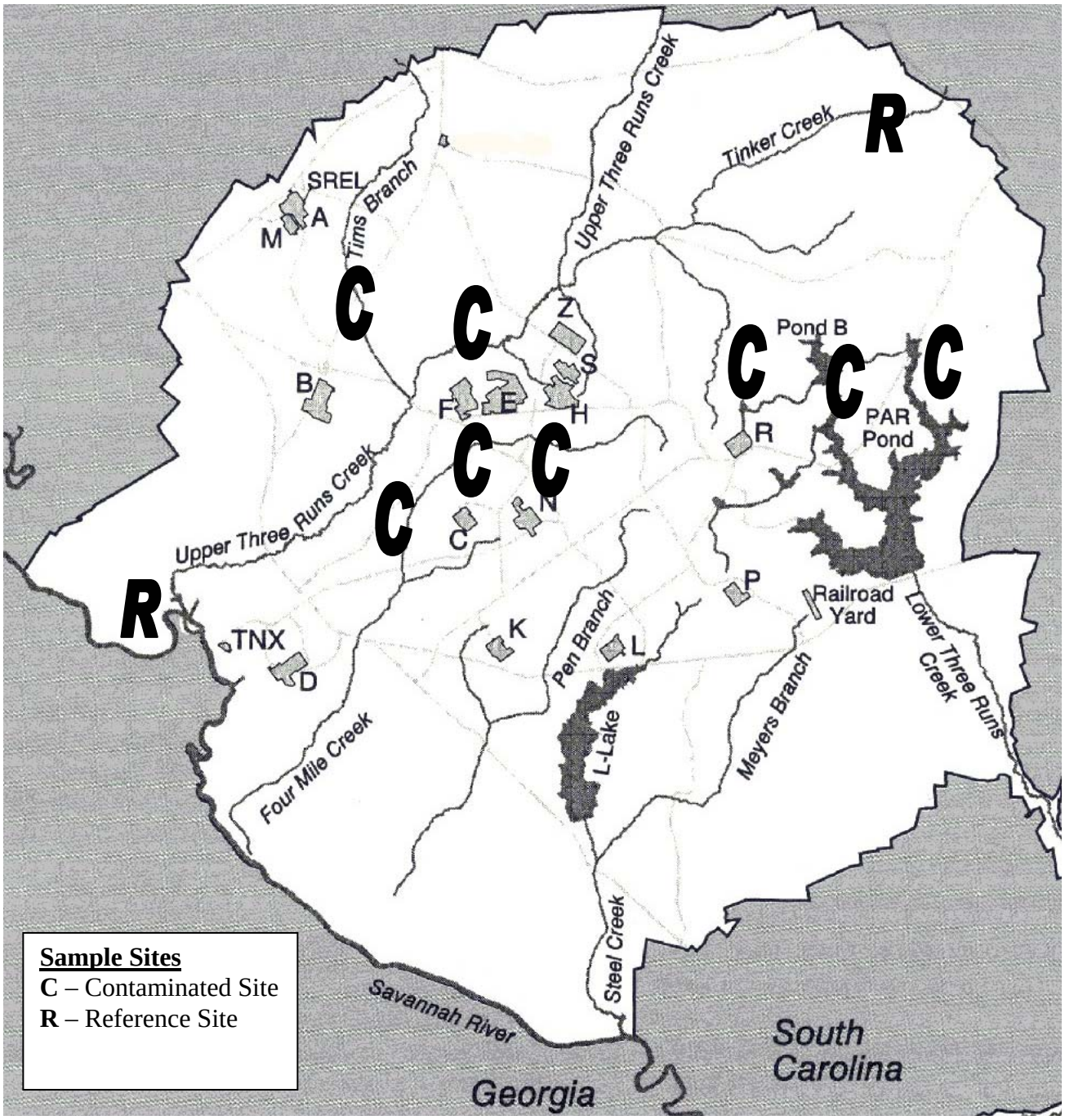

Table 1. Analysis of radioisotopes by plant species and area sampled.

Plant contamination levels by site and plant species in Bq kg^{-1} units

the subscript number in parenthesis equates the number of samples below m.d.l.

* represents species with no sample above m.d.a. NA denotes no test performed.

		Plant	Soil			F Area	^{137}Cs		^{40}K		^{238}Pu		^{239,240}Pu	
Species	***Common name***	***n***	***n***	**Tissue**	**Sampled**	**Location**	**Plant**	**Soil**	**Plant**	**Soil**	**Plant**	**Soil**	**Plant**	**Soil**
Cladina rangiferina	reindeer lichen	*1*	*0*	whole	Feb.	F-Area, North	23.68	NA	*	NA	*	NA	*	NA
Tillandsia usneoides	Spanish moss	*1*	*0*	whole	Feb.	F-Area, North	13.14	NA	346.3	NA	*	NA	*	NA
C. rangiferina	reindeer lichen	*1*	*0*	whole	Feb.	F-Area	15.54	NA	*	NA	*	NA	3.848	NA
Pinus palustris	longleaf pine	*1*	*0*	needles	Feb.	F-Area	8.84	NA	157.3	NA	*	NA	*	NA
		Plant	**Soil**			**Four Mile Creek**	^{137}Cs		^{40}K		^{238}Pu		^{239,240}Pu	
Species	***Common name***	***n***	***n***	**Tissue**	**Sampled**	**Location**	**Plant**	**Soil**	**Plant**	**Soil**	**Plant**	**Soil**	**Plant**	**Soil**
Sparganium americanum	bur-reed	*2*	*2*	leaves	March	Road 3	113.04	19.46	1513.3	*	*	0.432	*	$_{(1)}$ 0.437
Carex spp.	sedge	*3*	*3*	leaves	April	Road 3	776.38	449.67	499.5	*	$_{(1)}$ 0.641	7.799	*	4.039
Utricularia inflata	bladderwort	*1*	*0*	leaves	April	Road 4	3922.00	NA	862.1	NA	8399.000	NA	802.900	NA
S. americanium	bur-reed	*6*	*3*	leaves	March	Road 4	268.87	738.77	1111.2	*	115.989	51.997	10.152	4.655
S. americanium	bur-reed	*4*	*1*	roots	March	Road 4	379.16	740.01	471.0	*	677.655	66.971	61.577	6.956
Typha latifolia	common cattail	*11*	*2*	leaves	April	Road 4	527.25	783.85	790.1	66.1	6.348	152.440	$_{(6)}$ 0.741	13.542
T. latifolia	common cattail	*10*	*2*	roots	April	Road 4	702.15	783.85	485.2	66.1	306.393	152.440	28.817	13.542
Arundinaria gigantea	gaint cane	*1*	*0*	leaves	March	Road 4	954.60	NA	164.3	NA	1.169	NA	*	NA
Juncus effusus	*Juncus*	*8*	*0*	leaves	April	Road 4	1850.93	NA	470.4	NA	4.624	NA	$_{(3)}$ 0.479	NA
J. effusus	*Juncus*	*6*	*0*	roots	April	Road 4	5721.43	NA	$_{(1)}$ 28.6	NA	279.473	NA	25.986	NA
S. americanium	bur-reed	*5*	*2*	leaves	March	Road C	138.82	717.80	1605.8	*	9.607	16.706	$_{(2)}$ 1.011	$_{(4)}$ 2.332
S. americanium	bur-reed	*3*	*3*	roots	March	Road C	150.84	466.2	571.2	*	30.574	8.917	$_{(2)}$ 4.329	1.654
T. latifolia	common cattail	*1*	*0*	leaves	April	Road C	17.21	NA	*	NA	3.001	NA	*	NA
J. effusus	*Juncus*	*4*	*0*	leaves	April	Road C	2407.78	NA	356.6	NA	$_{(2)}$ 1.160	NA	$_{(3)}$0.839	NA
J. effusus	*Juncus*	*2*	*0*	roots	April	Road C	2876.75	NA	$_{(1)}$ 148.4	NA	110.630	NA	12.303	NA
		Plant	**Soil**			**R Reactor Cooling Basin**	^{137}Cs		^{40}K		^{238}Pu		^{239,240}Pu	
Species	***Common name***	***n***	***n***	**Tissue**	**Sampled**	**Location**	**Plant**	**Soil**	**Plant**	**Soil**	**Plant**	**Soil**	**Plant**	**Soil**
Nymphaea odorata	water lily	*3*	*3*	leaves	Feb., July	Par Pond	76.47	220.89	722.7	*	*	NA	*	NA
Myrica cerifera	wax myrtle	*1*	*1*	leaves	July	Par Pond	30.78	259.10	*	104.4	NA	NA	NA	NA
P. palustris	longleaf pine	*6*	*6*	needles	Feb., July	Pond A	283.42	251.11	$_{(3)}$ 114.7	*	*	NA	*	NA
N. odorata	water lily	*5*	*4*	leaves	Feb., July	Pond A	745.20	7311.83	967.2	*	*	NA	*	NA
N. odorata	water lily	*3*	*0*	roots	Feb., July	Pond A	555.00	NA	526.6	NA	NA	NA	NA	NA
M. cerifera	wax myrtle	*3*	*3*	leaves	Feb., July	Pond A	42.48	991.85	$_{(1)}$ 115.1	$_{(2)}$ 119.8	NA	NA	NA	NA
P. palustris	longleaf pine	*5*	*5*	needles	Feb.	Pond B	1522.18	858.33	$_{(3)}$ 125.4	*	*	NA	*	NA
N. odorata	water lily	*5*	*3*	leaves	Feb., July	Pond B	3090.24	13338.50	$_{(1)}$ 399.2	*	*	NA	*	NA
N. odorata	water lily	*3*	*3*	roots	Feb., July	Pond B	2750.33	13338.50	251.0	*	NA	NA	NA	NA
M. cerifera	wax myrtle	*5*	*5*	leaves	Feb., July	Pond B	672.66	NA	$_{(4)}$ 232.0	$_{(1)}$ 27.8	NA	NA	NA	NA
M. cerifera	wax myrtle	*6*	*6*	leaves	Feb., July	R-Canal	169.28	2621.26	$_{(4)}$ 199.4	$_{(3)}$ 39.71	NA	$_{(1)}$ 7.622	NA	$_{(1)}$ 3.448
		Plant	**Soil**			**Reference Areas**	^{137}Cs		^{40}K		^{238}Pu		^{239,240}Pu	
Species	***Common name***	***n***	***n***	**Tissue**	**Sampled**	**Location**	**Plant**	**Soil**	**Plant**	**Soil**	**Plant**	**Soil**	**Plant**	**Soil**
U. inflata	bladderwort	*1*	*0*	whole	April	Fire Pond	*	NA	506.9	NA	0.670	NA	0.907	NA
S. americanium	bur-reed	*3*	*1*	leaves	April	Tinker Creek	*	$_{(1)}$ 4.33	1064.4	*	$_{(2)}$ 0.259	*	$_{(2)}$ 0.318	*
Carex spp.	sedge	*2*	*1*	leaves	April	Tinker Creek	$_{(1)}$ 10.84	*	397.9	*	$_{(1)}$ 0.925	$_{(1)}$ 0.299	*	*
T. latifolia	common cattail	*3*	*1*	leaves	May	Ellenton Bay	*	*	762.2	165.8	*	NA	*	NA
J. effusus	*Juncus*	*3*	*3*	leaves	April	Ellenton Bay	23.69	18.16	446.0	174.4	*	$_{(1)}$ 0.221	*	0.807
C. rangiferina	reindeer lichen	*3*	*0*	whole	May	Ellenton Bay	*	NA	*	NA	*	NA	*	NA
P. palustris	longleaf pine	*3*	*1*	needles	May	Ellenton Bay	*	25.09	$_{(2)}$ 30.2	*	NA	NA	NA	NA
N. odorata	water lily	*3*	*0*	leaves	April	Ellenton Bay	*	NA	432.9	NA	NA	NA	NA	NA
M. cerifera	wax myrtle	*3*	*3*	leaves	Nov.	Ellenton Bay	*	7.75	93.1	$_{(2)}$ 37.37	NA	NA	NA	NA

Table 2. Statistical separation of radioisotope means (Bq kg^{-1}) by plant species expressed as. Each letter denotes a significantly different group. The numbers in parenthesis are the standard deviation.

Contaminated Sites	**^{137}Cs**	p <0.05	**^{40}K**	p <0.05	**^{238}Pu**	p <0.05	**^{239,240}Pu**	p <0.05
U. inflata	3922.00		862.1		8399.00		802.900	
S. americanum	194.9 (141.1)	c	1383.3 (352.3)	a	78.01 (140.69)	b	8.74 (13.2)	bc
***S. americanum* - root**	281.3 (149.8)	c	513.9 (155.0)	bc	400.34 (383.88)	a	42.495 (35.29	a
Carex spp.	776.3 (554.2)	bc	499.5 (61.8)	bc	0.640	b	*	
T. latifolia	484.7 (383.9)	c	790.1 (177.6)	b	6.07 (4.78)	b	0.799 (0.321)	c
***T. latifolia* - root**	702.3 (229.2)	bc	485.1 (184.7)	bc	306.39 (225.28)	a	28.82 (18.95)	a
A. gigantea	954.6		164.3		1.17		*	
J. effusus	2036.5 (730.4)	b	432.9 (72.7)	bcd	3.01 (3.93)	b	0.654 (0.276)	c
***J. effusus* - root**	5010.2 (2654.7	a	259.3 (109.8)	de	237.29 (169.90)	ab	22.57 (15.97)	ab
C. rangiferina	19.6 (5.8)		25.6 (0.9)		*		3.840	
P. palustris	846.5 (780.5)	bc	118.92 (58.8)	e	*		*	
T. usneoides	13.1		346.3		*		*	
N. odorata	1492.9 (137.9)	bc	716.7 (262.4)	b	*		*	
***N. odorata* - root**	1652.8 (1364.8	bc	416.4 (163.3)	cde	*n/a*		*n/a*	
M. cerifera	302.7 (389.2)	c	195.5 (117.3)	de	*n/a*		*n/a*	

All comparisons of samples by radionuclides are significant at the 0.01 level. Means are separated using Tukey HSD multiple comparison test ($p < 0.05$).

Table 3. Statistical comparison of means (Bq kg^{-1}) for vegetation with at least three replications from the control and contaminated sites.

	^{137}Cs		^{40}K		^{238}Pu		$^{239,240}Pu$	
	Contaminated	**Reference**	**Contaminated**	**Reference**	**Contaminated**	**Reference**	**Contaminated**	**Reference**
U. inflata	3922.00	*	862.10	506.90	8399.00	0.67	802.90	0.91
S. americanum	194.9	*	1383.4	1064.4	78.010	0.256	8.740	0.318
Carex spp.	776.3	10.8	499.5	397.9	5.130	0.921	*	*
T. latifolia	484.7	*	790.1	844.5	6.070	*	0.512	*
J. effusus	2036.5	23.7	432.4	446.0	3.290	0.037	0.386	0.052
P. palustris	846.5	*	123.3	301.5	*n/a*	*n/a*	*n/a*	*n/a*
N. odorata	1492.9	*	716.7	432.9	*n/a*	*n/a*	*n/a*	*n/a*
M. cerifera	302.7	*	195.5	93.1	*n/a*	*n/a*	*n/a*	*n/a*

Table 4. The concentration ratios for radionuclides in plants from contaminated sites.

Contaminated site concentration ratios

	^{137}Cs	^{40}K	^{238}Pu	^{239,240}Pu
S. americanum (n,9)	$_{(9)}$1.59 (2.4)	*	$_{(7)}$1.75(2.5)	$_{(4)}$2.90 (2.8)
***S. americanum* - root** (n,4)	$_{(4)}$0.34 (0.04)	*	$_{(4)}$5.86 (4.9)	$_{(3)}$5.66(5.2)
Carex spp. *(n,3)*	$_{(3)}$1.37 (1.5)	*	$_{(1)}$5.13	*
T. latifolia *(n,2)*	$_{(2)}$0.87 (0.9)	$_{(2)}$14.33 (4.4)	$_{(2)}$0.02 (0.0001)	$_{(1)}$0.03
***T. latifolia* - root** *(n,2)*	$_{(2)}$1.46 (1.6)	$_{(2)}$9.47 (5.5)	$_{(2)}$1.74 (1.7)	$_{(2)}$1.60 (1.6)
P. palustris *(n,11)*	$_{(11)}$2.18 (1.5)	*	*n/a*	*n/a*
N. odorata *(n,10)*	$_{(10)}$0.30 (0.2)	*	*n/a*	*n/a*
***N. odorata* - root** *(n,6)*	$_{(6)}$0.24 (0.3)	*	*n/a*	*n/a*
M. cerifera *(n,15)*	$_{(15)}$0.74 (1.1)	$_{(5)}$3.58 (1.1)	*n/a*	*n/a*

Table 5. Correlation and regression analysis of plant and soil radioisotope concentration.

Correlation and Regression Analysis of plant and soil concentrations

	^{137}Cs			^{238}Pu			^{239,240}Pu		
		Regression			*Regression*			*Regression*	
	Correl.	R^2	*p*	*Correl.*	R^2	*p*	*Correl.*	R^2	*p*
S. americanum	0.25	0.061	0.523	0.73	0.536	0.025	0.59	0.346	0.096
***S. americanum* - root**	0.97	0.940	0.030	1.00	0.999	0.001	0.98	0.956	0.021
P. palustris	0.76	0.584	0.006						
N. odorata	0.61	0.371	0.061						
M. cerifera	0.06	0.004	0.881						

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