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Retention: Permanent

Bioavailability of Radioactive Cesium in Old R Discharge Canal, R-Canal, Pond A, and the Adjacent Flood Plain

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1.0 EXECUTIVE SUMMARY

Risk calculations are often based on the total concentration of a contaminant within the sediment, however, such calculations may over-estimate risks because not all of the contaminant is available for uptake by the biota. Determining the fraction of the total contaminant within the sediment that is not bioavailable could make risk calculations more realistic and potentially reduce the extent (source term) of required cleanup.

The objectives of this study were to provide site-specific data for human and ecological risk analysis associated with ^{137}Cs contamination along the P and R reactor cooling canal system. This area, which contains wetlands, was contaminated with ^{137}Cs during the operation of R Reactor prior to 1964. For this study, bioavailability was measured directly from ^{137}Cs concentrations in paired sediment and plant samples collected at the study site. These data were used to calculate concentration ratios (CR = activity concentration in the dried plant / activity concentration in dried sediment). Additionally, the nature of the bond between ^{137}Cs and the sediments at this site were characterized through sequential extraction techniques. These measurements provided insight into how strongly ^{137}Cs is bound to the sediment and provide indirect information about the tendency for ^{137}Cs to be taken up by plants. Among the key findings from this research are the following:

- Sequential extraction results indicated that between 50 and 85% of the ^{137}Cs in these sediments was strongly bound and likely in a chemical form that was of limited bioavailability.
- Estimates of concentration ratios were often in excess of 1.0, indicating that the plants concentrated ^{137}Cs in their tissue to levels greater than existed in the sediment.
- Concentration ratios of plants growing on terrestrial sites were significantly greater than those of the same species growing in aquatic locations.
- Mean (\pm SD) ^{137}Cs concentrations in the Old R Discharge Canal (189 ± 167 pCi g^{-1}) were no different from the average for R Canal (224 ± 213 pCi g^{-1}) or Pond A (213 ± 205 pCi g^{-1}). Part of the motivation for sampling the Old R Discharge Canal was that gamma over-flight data indicated that ^{137}Cs contamination levels were higher there than in Pond A. This apparent discrepancy is likely the result of increased shielding of the gamma emissions by the greater depth of water in Pond A as compared to the Old R Discharge Canal. These findings indicate that remediation decisions about ^{137}Cs -contaminated wetlands could be misdirected if based solely on gamma over-flight data.

Since predictions of ^{137}Cs concentrations in plants are tenuous when based solely on sediment ^{137}Cs concentrations, and the predictions are not captured well by a single parameter, such as a CR, it is recommended that risk assessments involving the ingestion pathway be based directly on measured concentrations in plants. Relying solely on sediment ^{137}Cs concentrations, either total or that which is biologically available, could cause large errors in risk estimations.

2.0 INTRODUCTION

2.1 BACKGROUND

Over 1,200 hectares (3,000 acres) of wetlands on the Savannah River Site (SRS) are contaminated with 21 TBq (564 Ci) of ^{137}Cs and pose a significant remediation challenge to the Department of Energy (DOE). The problems of contaminated aquatic systems and their associated risks are heightened on the SRS because ^{137}Cs is more mobile here than at any other DOE facility. Reasons for the enhanced bioavailability of ^{137}Cs are largely due to the low clay content of SRS sediments, and that the clays that are present are dominated by kaolinite and iron oxides. The abundance of clay is important in determining bioavailability because ^{137}Cs can be bound within the physical structure of clay minerals (Francis and Brinkley 1976), in essence, trapping ^{137}Cs and reducing its biological uptake until physical decay removes it from the system. Thus, as the percentage of clay increases, availability of ^{137}Cs generally decreases. Kaolinite's physical structure, however, does not bind ^{137}Cs as do clays that are in 2:1 layers, such as micas.

Cesium undergoes a seasonal cycle of enhanced bioavailability on the SRS. In summer, lakes on the SRS thermally stratify and the deeper hypolimnion layer becomes anoxic. During this period, ^{137}Cs is released. The cause for the release under anoxic conditions may be attributed to either the dissolution of Fe(III)-oxides, which sorb ^{137}Cs , or to the microbial generation of NH_4^+ from NO_3^- . Ammonium has a higher affinity for the clay binding sites to which ^{137}Cs is adsorbed, and thus displaces ^{137}Cs into the interstitial waters, thereby increasing the mobility and biological availability of the contaminant. Although only a small percentage of the total inventory, the released ^{137}Cs is sufficient to cause measurable concentrations in all components of the wetlands (e.g. water, plants, aquatic insects, fish, etc).

Increased ^{137}Cs availability was documented in these systems during the 1960s era of weapons fallout (Whicker and Schultz 1982), and the high mobility of ^{137}Cs still exists today, as is evident by the propensity of ^{137}Cs to be taken up from the sediment by plants. This propensity can be measured using a concentration ratio (CR), where $\text{CR} = \text{Bq kg}^{-1} \text{ dried plant} / \text{Bq kg}^{-1} \text{ dried sediment}$. The National Council on Radiological Protection and Measurements recommends a default ^{137}Cs CR of 0.1 for use in risk calculations. In comparison, ^{137}Cs CRs for the SRS are generally well above 1.0. Indeed, they are among the highest recorded ^{137}Cs CRs in the world. Research conducted in the 1970s found plants from Steel Creek and Par Pond to have CRs that ranged from 3 to 7 (Garten and Paine 1977; Pinder et al. 1980). More recent research found that garden vegetables grown on the exposed lake bed of Par Pond had CRs of 6.8 ± 2.3 (Seels et al. 1995), and early successional species invading Par Pond's exposed lake bed had very high CRs of 14.9 ± 2.3 (Hinton et al. 1999). Such large CRs indicates how readily ^{137}Cs is taken up by plants and how weakly the ^{137}Cs is held by SRS sediments. Because of these large CRs, ^{137}Cs can be the critical contaminant when human and ecological risk analyses are conducted for the SRS. Part of the work reported herein uses CRs to examine the bioavailability of ^{137}Cs in wetlands of the SRS.

Human and ecological risk calculations will likely be used to determine the extent of remediation required within these wetlands. Often, risk calculations are based on the contaminant concentrations within the sediments. Such calculations, however, may over-estimate risks because not all of the contaminant is available for uptake by the biota. Some portion of the ^{137}Cs is sorbed onto the sediment and is not readily taken up by plants or dissolved into the overlying waters. Determining the available fraction of the total contaminant within the sediment (i.e. that which is available for plant uptake) could make risk calculations more realistic and potentially reduce the extent of required cleanup. More directly, the bioavailable fraction could be based directly on the amount of contaminant measured in field samples. Remediation of sensitive wetlands is expensive and is also an extremely difficult procedure to accomplish without destroying the fragile ecosystems.

Use of total concentration as a criterion to assess the potential effects of Cs in the sediment environment is generally not satisfactory. To provide a more precise understanding of the potential impact of elevated levels of Cs in sediments, it is necessary to identify and quantify the forms in which Cs is present in the sediment. Sequential extractions are often used to estimate the available fraction of a contaminant. Sequential extractions are used to examine the solubility of a contaminant in progressively stronger reagents. The reagents are chosen to target specific components within the sediment matrix that the contaminant may be residing in. Part of the work reported herein uses sequential extractions to examine the distribution of ^{137}Cs in SRS wetland sediments.

This study was conducted at R-Canal, Pond A, and the Old R Discharge Canal because they have been shown to contain contaminant ^{137}Cs . This area consists of wetlands, ponds, and canals with standing water that varies from a few centimeters to 150 cm in depth. The wetlands' associated flood plains were also studied and are referred to in this report as terrestrial sites.

2.2 OBJECTIVES AND SCOPE

This research was designed to improve site-specific risk calculations from ^{137}Cs -contaminated wetlands on the SRS. Risk calculations based on total ^{137}Cs concentrations in sediments may lead to an over-estimation of hazards, unnecessary destruction of sensitive wetlands, and needless expenditure of funds.

The objective of this proposal was to provide more realistic data for human and ecological risk analysis. Specifically, the objectives and scope of this study were to:

- collect plant and associated sediment samples from terrestrial flood plain sites on the Old R Discharge Canal and determine bioavailability of ^{137}Cs to specific plant species based on concentration ratios,
- collect plant and associated sediment samples from aquatic sites of the Old R Discharge Canal and to compare ^{137}Cs bioavailability in the terrestrial flood plain and aquatic systems,
- determine, through the use of sequential extraction techniques, which specific mineral phases bind ^{137}Cs , and

- estimate availability of ^{137}Cs to aquatic and terrestrial plants using concentration ratios.

3.0 MATERIALS AND METHODS

3.1 SEDIMENT CHARACTERIZATION

A total of 12 sampling locations were randomly selected along the Old R Discharge Canal located ~ 0.8 km along the cooling water outflow canal from R- reactor (Figure 1). At each location, both plant and associated sediment samples from aquatic and terrestrial flood plain sites were collected. In June, sixteen plant and sediment samples were taken from the terrestrial sites and 14 from the aquatic (Figure 1 and Table 1). Sediment samples were collected from the rooting zone of the plants. Most sediment samples were collected as a grab sample (0-7.5 cm in depth), but sediment cores were taken from 5 aquatic sites. The sediment cores were sliced into 2.5 cm sections and each slice was processed as an individual sample. Sediment samples were oven dried (60-65°C), homogenized, and sieved through a 2-mm screen. Sediment pH was determined from a 1:1 sediment/water equilibration solution (Thomas, 1996). Percent soil organic matter was estimated by loss-on-ignition at a temperature of 450 °C. Particle size distributions (percent sand, silt, and clay) were determined by the micro-pipette method (Miller and Miller, 1987). Sediment samples with organic matter content higher than 10% had their particle size distribution analysis performed after organic matter was removed with H_2O_2 .

As part of another study on R-Canal and Pond A, 16 limnocorrals have been established to study the effects of illite clay amendments on Cs mobility and bioavailability (Figure 2). Limnocorrals are flexible, plastic cylinders (3 m in diameter) that are anchored to the pond sediment and extend upwards through the water column to the atmosphere. We used these limnocorrals as additional sediment/plant sampling locations for the study reported herein. From each limnocorral three replicate sediment cores were taken.

3.2 PLANT SAMPLES

Plant species taken from the Old R Discharge Canal were royal fern (*Osmunda regalis*), netted chain fern (*Woodwardia aerloata*), and rice cutgrass (*Leersia oryzoides*) (Table 1). These species were selected because they are abundant in the area and because they were found in both aquatic and terrestrial environments. From the limnocorrals in Pond A, we collected three replicate samples of water lily (*Nymphaea odorata*) leaves and stems. All plant samples were cleaned with tap water and oven-dried at 60-65°C. Plant samples were then ground, weighed, and placed into 24-ml scintillation vials for ^{137}Cs analyses.

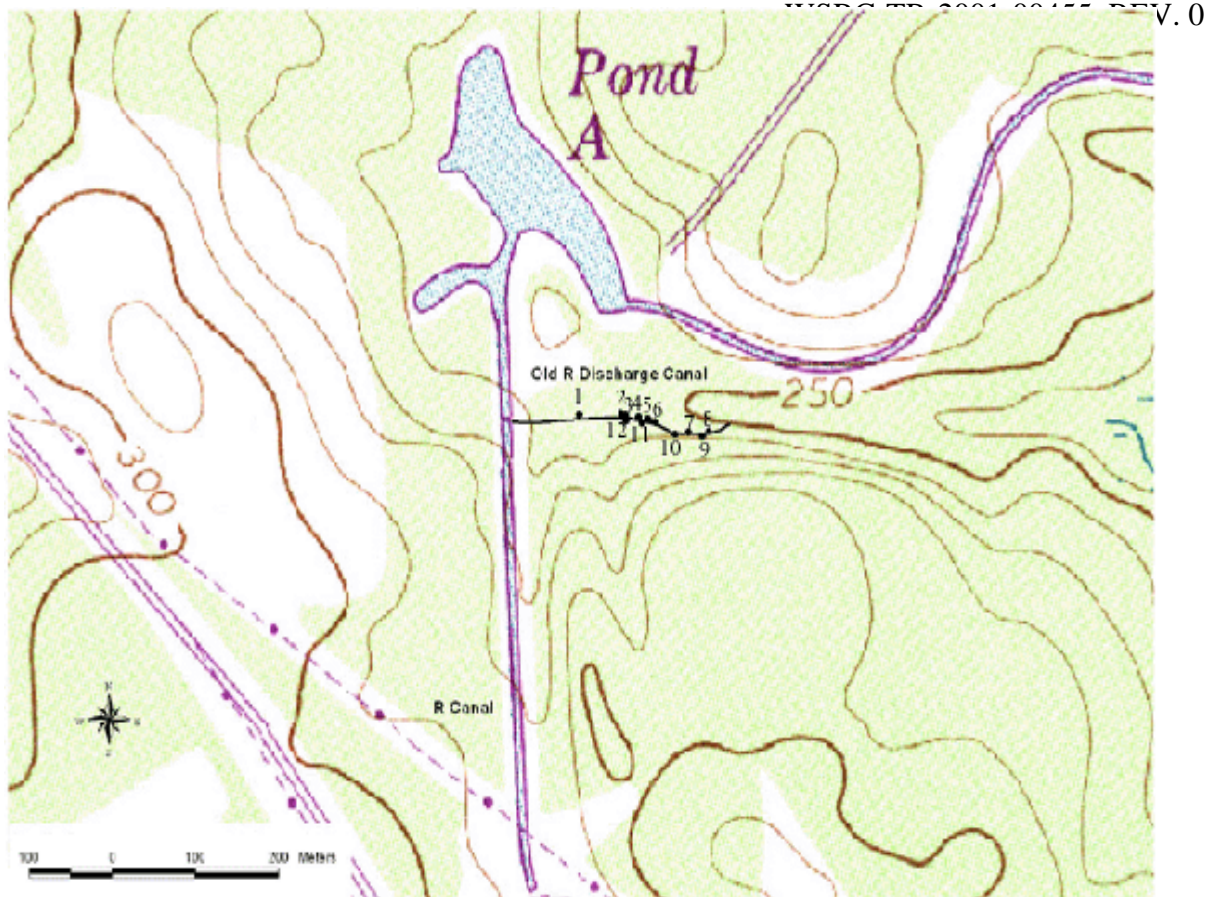


Figure 1. Old R Discharge Canal's position relative to Pond A and the main R-Canal. Dots indicate 12 sampling locations. Contour lines depict topographical elevation.

3.3 ^{137}Cs ANALYSES

Analyses of ^{137}Cs in plants, sediments, and sequential extracts were performed on a Packard Autogamma instrument with a well-type NaI crystal (7.6 x 8.3 cm with a 1.5 % relative efficiency). Additional analyses of portions of the sequential extracts were performed on a 70x90 mm² high-purity germanium detector. Cs-137 standards, traceable to the National Institute of Standards and Technology, were assayed in the same physical geometry as the samples.

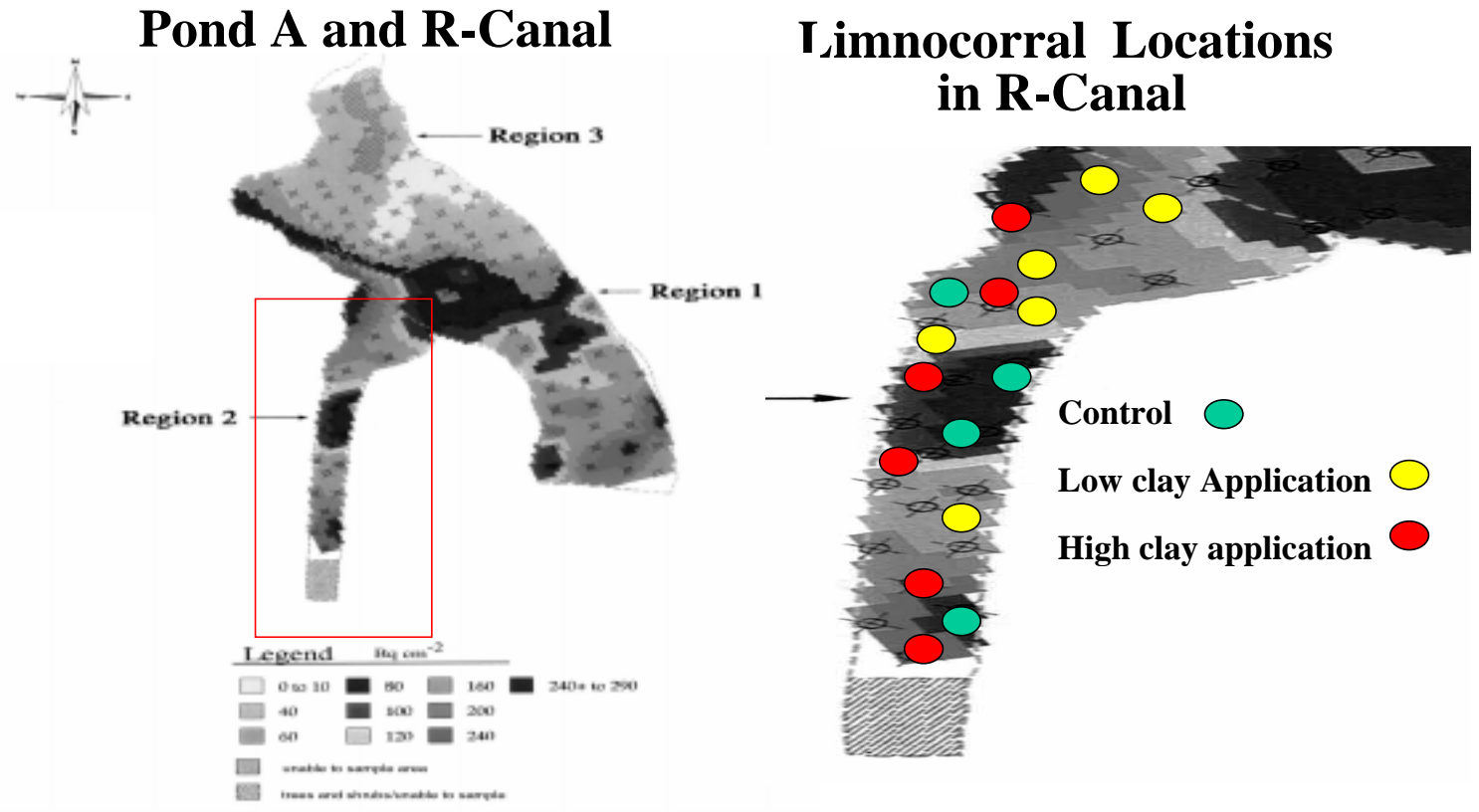


Figure 2. Location of 16 limnocorrals in R-Canal from which sediment and water lily samples were taken.

Table 1. Plant samples collected at Old R Discharge Canal; T- terrestrial, A- aquatic.

Location	Royal Fern		Netted Chain Fern		Rice Cutgrass	
	Aquatic	Terrestrial	Aquatic	Terrestrial	Aquatic	Terrestrial
1	-	X	-	-	-	-
2	X	-	-	-	-	-
	-	X	-	-	-	-
3	-	-	-	-	X	-
	-	-	-	-	-	X
4	X	-	-	-	-	-
	-	-	-	-	X	-
	-	-	-	-	-	X
5	X	-	-	-	-	-
	-	-	-	-	X	-
	-	X	-	-	-	-
	-	-	-	-	-	X
6	-	-	-	X	-	-
	-	-	-	-	-	X
7	-	-	-	-	X	-
	-	-	-	X	-	-
8	-	-	X	-	-	-
	-	-	-	-	X	-
	-	-	-	X	-	-
	-	-	-	-	-	X
9	X	-	-	-	-	-
	-	-	-	-	X	-
	-	-	-	X	-	-
	-	-	-	X	-	-
10	X	-	-	-	-	-
	-	-	-	-	-	X
11	-	-	-	-	X	-
	-	-	-	X	-	-
12	-	-	X	-	-	-
	-	-	-	X	-	-
Number of Samples	5	3	2	7	7	6
Total Number of Samples/Species	8		9		13	

3.4 SEQUENTIAL EXTRACTIONS

Chemical extraction of sediment contaminants with sequentially more aggressive reagents is commonly used to characterize various forms of metal or radionuclides in sediments. Numerous sequential extraction procedures have been developed (Tessier et al., 1979, Miller et al., 1986; Hall et al., 1995). The major problem with sequential extraction studies is that the detailed nature of the material being extracted is not known. Idealized pure phases are often named in various schemes on the basis of their response to the extraction reagents, but this approach can be misleading in that the behavior of natural samples may be variable depending on the inorganic and organic constituents with which metals are associated. Spectroscopic methods may yield more precise information regarding the identification of the metal forms, the specific phases the metal is bound to, and the nature of the sediment-metal bond (e.g., bond strength, bond distance, and bond type). But these methods are usually expensive, non-routine, and not sensitive enough to detect contaminants in low concentrations. Therefore, sequential extractions, even though they have several limitations, are still widely used.

A sequential extraction procedure, modified from Hall et al. (1995) and Tessier et al. (1979), was conducted on eight sediments collected from R-Canal and Pond A. An overview of the sequential extraction procedure used in this study is presented in Table 2. The technique differentiates five sediment fractions in which ^{137}Cs was found:

- Adsorbed/Exchangeable/Carbonate (AEC),
- Amorphous Fe-oxyhydroxide (AmFeOx),
- Crystalline Fe-oxide (CryFeOx),
- Organic Matter (OM), and
- Residual silicates (R).

Exchangeable Cs is held by electrostatic attraction on the surface of negatively charged sediment complexes. Neutral salts such as MgCl_2 or ammonium acetate can easily displace cesium held to exchange sites. This fraction is often considered the most biologically available. In the sequential extraction methods we used, exchangeable and carbonate bound (not abundant in SRS sediments) Cs was grouped together. This fraction was extracted with sodium acetate, buffered at pH 5, and designated the Adsorbed/Exchangeable/Carbonate complex (AEC). Also extracted together, in the next step, were the hydrous oxides of Mn and Fe, well known “sinks” in the surficial environment for heavy metals and radionuclides. Scavenging by these secondary oxides, present as coatings on mineral surface or as fine discrete particles, may occur by any combination of the following mechanisms: coprecipitation, adsorption, surface complex formation, ion exchange, and/or penetration of the mineral lattice. The third fraction consisted of crystalline Fe oxides. The relative importance of Fe and Mn oxides as scavengers depends upon such conditions as pH-Eh values, degree of oxide crystallization, their abundances, and the presence of organic matter. Cesium bound to organic matter was next in the sequence and represented the fourth extract. The final part of the sequential extraction procedure represented the residual fraction, and is

thought to mainly consist of primary and secondary silicate minerals that may hold Cs tenaciously within their crystal structure. This fraction was determined by gamma spectroscopy after the four prior extraction steps.

Sediment cores from R-Canal and Pond A were sliced into 2.5 cm sections and each slice was characterized for total ^{137}Cs , pH, and particle size distribution as described in Section 3.1. Eight representative sediment samples from R-Canal were selected for sequential extraction (two samples with high organic matter content, two samples with high clay content, two samples with high content of organic matter and clay, and three samples with high content of sand fraction). All extracts were performed by shaking the samples in 50 ml centrifuge tubes with the appropriate reagents at a 40:2 reagent to sample ratio, and spun for 30 min at 2800 rpm. The extracts were transferred to two 20 ml vials in preparation for determining ^{137}Cs concentration. The residue was prepared for the next extraction step by washing with 10 ml of 0.01 M CaCl_2 (three times), centrifuging at 2800 rpm for 30 min and collecting the decanted supernatant liquid into a vial for ^{137}Cs analysis. ^{137}Cs activity was measured directly in the remaining solid phase (as opposed to after total digestion by strong acids, such as aqua regia).

Table 2. Sequential extraction regime used in this study.

Sequence	Extractant: Chemical Composition	Targeted Contaminant Fraction	Ref. ^a
Initial sample	2 g of sample	Total ^{137}Cs activity of the sample was performed on high-purity germanium detector	
1	1.0 M NaOAc; pH 5; repeated twice	Adsorbed/exchangeable/carbonate (AEC)	1
2	0.25 M $\text{NH}_2\text{OH}\cdot\text{HCl}$ in 0.25 M HCl; repeated twice	Amorphous Fe oxyhydroxide (Am Fe ox)	1
3	1.0 M $\text{NH}_2\text{OH}\cdot\text{HCl}$ in 25% HOAc; repeated twice	Crystalline Fe oxide (Cry Fe ox)	1
4	0.02 N HNO_3 & 30% H_2O_2 ; repeated twice	Organic Fraction (OM)	2
5		Residual (silicates) (R)	

^aReferences: 1 = Hall et al. 1995, 2 = Tessier et al. 1979.

4.0 RESULTS AND DISCUSSION

4.1 SEDIMENT CHARACTERIZATION

Sediments collected at aquatic sites had a pH range of 3.9 to 5.1 with a mean of 4.3 (Table 3). Organic matter content was high in five samples (more than 5% but less than 10%, wt) and very high in seven samples (more than 20%, wt; Table 3). A high content of organic matter is consistent with the fact that the sediments were collected from a forested wetland. Particle size analysis showed that the textures of the sediments from aquatic sites varied substantially (Table 3). The clay-size fraction was the smallest and ranged from 0.5 to 8.9%, with a mean of 3.8%. The silt-size fraction varied substantially, ranging from 0.7 to 85%, with a mean of 44.1%. The highest fraction for aquatic sediments was sand with a mean of 52% (Table 3).

Sediment samples collected from the terrestrial flood plain sites were remarkably similar to those collected from the aquatic sites in sand and silt content ($p > 0.05$); but the clay content was significantly lower in the terrestrial site ($p < 0.01$), with a mean percentage of 1.5 (Table 4). The two types of sites had similar pH values and organic matter content (Table 4).

Table 3. Sediment characterization of samples collected from aquatic locations in the Old R Discharge Canal.

Aquatic sediment samples						
Location	Associated plant sample	OM (%, wt)	pH (unitless)	Sand (%, wt)	Silt (%, wt)	Clay (%, wt)
2	Royal Fern	6.65	3.95	68	29	3
3	Rice Cutgrass	6.41	4.25	52	44	5
4	Royal Fern	7.86	4.35	76	22	2
4	Rice Cutgrass	7.17	4.22	50	45	5
5	Rice Cutgrass	29.11	5.1	29	67	5
5	Royal Fern	12.33	5	6	85	9
7	Rice Cutgrass	22.40	4.22	7	85	8
8	Netted Chain Fern	61.25	4.17	98	1	1
8	Rice Cutgrass	33.60	3.85	63	34	2
9	Royal Fern	7.00	4.23	64	32	3
9	Rice Cutgrass	6.91	4.18	84	15	1
10	Royal Fern	43.42	4.17			
11	Rice Cutgrass	47.14	3.88	53	45	2
12	Netted Chain Fern	29.03	4.54	28	70	3
	Mean	22.88	4.42	52	44	4
	Standard Deviation	± 18.19	± 0.45	± 28	± 26	± 3

Table 4. Sediment characterization of samples collected from terrestrial flood plain sites along the Old R Discharge Canal.

		Terrestrial sediment samples				
Location	Associated plant sample	OM (%, wt)	pH (unitless)	Sand (%, wt)	Silt (%, wt)	Clay (%, wt)
1	Royal Fern	14.04	3.61	45	50	5
2	Royal Fern	53.00	3.89	63	36	1
3	Rice Cutgrass	7.07	5.43	80	20	0
4	Rice Cutgrass	20.84	3.92	78	22	1
5	Royal fern	33.07	4.72	85	22	0
5	Rice Cutgrass	29.03	5.35	55	45	1
6	Rice Cutgrass	16.66	4.3	90	10	0
6	Netted Chain Fern	69.49	3.61	36	63	1
7	Netted Chain Fern	50.33	4.09	30	68	3
8	Netted Chain Fern	19.53	4.06	72	26	2
8	Rice Cutgrass	10.12	4.31	75	23	2
9	Netted Chain Fern	14.86	3.76	49	50	4
9	Netted Chain Fern	19.50	3.98	82	17	2
10	Rice Cutgrass	3.44	5.65	83	16	0
11	Netted Chain Fern	14.07	5.06	87	13	0
12	Netted Chain Fern	7.96	4.64			
	Average	23.94	4.40	67	32	2
	Stdev	18.72	0.67	20	19	2

4.2 SEDIMENT ¹³⁷Cs CONCENTRATIONS

¹³⁷Cs concentrations in the sediments from the terrestrial and the aquatic sites of Old R Discharge Canal are presented in Table 5. The highest reported ¹³⁷Cs concentrations were in the sediment samples collected at the aquatic sites, e.g., 13.8 and 24.3 Bq g⁻¹ (373 and 656 pCi g⁻¹). The mean (± SD) concentration of ¹³⁷Cs in the sediments from the aquatic sites was 7.0 ± 6.2 Bq g⁻¹ (189 ± 167 pCi g⁻¹). In contrast, ¹³⁷Cs concentrations from the terrestrial flood plain sites were considerably lower, with a mean (± SD) of 1.3 ± 1.8 Bq g⁻¹ (35 ± 49 pCi g⁻¹) and a range from 0.08 to 6.2 Bq g⁻¹ (2 to 167 pCi g⁻¹).

¹³⁷Cs concentrations in the sediment cores taken from R-Canal and Pond A are reported in Table 6. The concentrations of ¹³⁷Cs in the sediment samples change with depth. On average, the greatest ¹³⁷Cs concentrations (9.0 ± 6.9 Bq g⁻¹; 243 ± 186 pCi g⁻¹) were measured 2.5 to 5.0 cm deep within the sediments, and concentrations then decreased with depth. This pattern is consistent with earlier published data from Pond A (Abraham et al., 2000).

The mean (± SD) ¹³⁷Cs concentration in the Old R Discharge Canal (7.0 ± 6.2 Bq g⁻¹; 189 ± 167 pCi g⁻¹) was no different from the mean for R Canal (8.3 ± 7.9 Bq g⁻¹; 224 ± 213 pCi g⁻¹) or from the mean reported for Pond A (7.9 ± 7.6 Bq g⁻¹; 213 ± 205 pCi g⁻¹) by Abraham et al. (2000) (Figure 3). Part of the motivation for sampling the Old R Discharge Canal was that gamma over-flight data indicated that ¹³⁷Cs contamination levels were higher there than in Pond A. This apparent discrepancy is likely the result of increased shielding of the gamma emissions by the greater depth of water in Pond A as compared to the Old R Discharge Canal. These findings indicate that remediation decisions about ¹³⁷Cs-contaminated wetlands could be misdirected if based solely on gamma over-flight data.

4.3 SEQUENTIAL EXTRACTIONS

The sediment samples studied by sequential extractions represent four “typical sediment types” found in Pond A and the R-Canal system; sediments with:

- 1) high organic matter content,
- 2) high clay content,
- 3) a combination of high clay and organic matter content, and
- 4) high sand content.

All 48 cores (3 replicates x 16 locations) collected from the limnocorral study conducted in R-Canal contained various proportions of these “typical sediment types.” These sediment classifications were easy to identify by unaided visual inspection of the cores. The sequential extraction technique allowed us to determine ¹³⁷Cs concentrations in five components of each sediment. The components (AEC, AmFeOx, CryFeOx, OM and R) were presented and discussed in section 3.4. ¹³⁷Cs was found in each component of all sediment types analyzed

(Figure 4). In all four “typical sediment types,” the greatest concentration of ^{137}Cs was found in the AmFeOx component (Figure 4). When compared across sediment types, the AEC component held a larger percentage (24%) of the ^{137}Cs in the sand than in the other three sediment types (Figure 4). Recall that the AEC component is the mineral matrix where Cs is most easily exchanged with other elements, and thus considered by some to be an estimate of the biologically available fraction. This fraction was greater than 15% in all sediment types. The residual fraction is expected to bind and hold Cs tenaciously, thereby reducing its bioavailability. As expected, we found the clay sediment type to contain the most ^{137}Cs in the residual fraction and sand to have the least (Figure 4).

Overall, the sequential extractions results suggest that anywhere from 10 to 50% of the total ^{137}Cs within the sediments of Pond A might be considered biologically available—depending on the seasonal site specific conditions within the sediments.

4.4 PLANT ^{137}Cs CONCENTRATIONS

Within the Old R Discharge Canal three plant species were collected: royal fern (*Osmunda regalis*), netted chain fern (*Woodwardia aerloata*), and rice cutgrass (*Leersia oryzoides*). The same species were collected at the aquatic and terrestrial flood plain sites (Table 5). The highest ^{137}Cs concentrations were observed in netted chain fern at an aquatic site; 8.1 Bq g^{-1} (219 pCi g^{-1}). Because ^{137}Cs concentrations in sediments from aquatic sites were significantly higher, $7.0 \pm 6.1 \text{ Bq g}^{-1}$ ($189 \pm 165 \text{ pCi g}^{-1}$), than those from the terrestrial flood plain, $1.3 \pm 1.8 \text{ Bq g}^{-1}$ ($35 \pm 49 \text{ pCi g}^{-1}$) one might expect ^{137}Cs concentrations in plants to be higher in aquatic sites as well. However, the mean ^{137}Cs concentrations in all plants collected from aquatic and terrestrial flood plain sites were similar, 1.9 ± 2.1 and $1.7 \pm 2.1 \text{ Bq g}^{-1}$, (51 ± 57 and $46 \pm 57 \text{ pCi g}^{-1}$). Some researchers have shown that sediment ^{137}Cs activity concentrations were positively correlated to plant uptake (Pinder et al., 1980), while others were unable to identify such a relationship (Horrill et al., 1990), indicating that plant concentrations do not necessarily depend upon sediment concentrations. Hughes et al., (1980) indicated that the main factors influencing plant uptake are sediment pH, and level and chemical species of contaminants. The influence of plant species on metal uptake may also be considerable. Different species regulate metal uptake at both the sediment-root and root-shoot interfaces to varying degrees (Archer, 1971). This was also observed in our data, in that ^{137}Cs concentrations in rice cutgrass were consistently lower than the two fern species (Table 5). Ferns are known for their tendency to take up radionuclides and heavy metals (Kaplan et al. 2001).

At R-Canal and Pond A, water lilies were collected at 16 locations. Generally, ^{137}Cs concentrations were higher in stems than in leaves. In leaves ^{137}Cs concentrations ranged from 0.9 to 1.5 Bq g^{-1} (24 to 40 pCi g^{-1}), while in stems ^{137}Cs concentrations were almost two times higher and ranged from 1.2 to 2.8 Bq g^{-1} (32 to 76 pCi g^{-1} , Table 7).

Table 5. ¹³⁷Cs concentrations in sediments, concentrations in associated plants, and concentration ratios (CR) from aquatic and terrestrial sites of Old R Discharge Canal.

Aquatic Site					Terrestrial Site				
Location	Sampled plant	¹³⁷ Cs in sediments	¹³⁷ Cs in plant	CR	Location	Sampled plant	¹³⁷ Cs in sediments	¹³⁷ Cs in plant	CR
		(Bq g ⁻¹)	(Bq g ⁻¹)				(Bq g ⁻¹)	(Bq g ⁻¹)	
2	Royal Fern	7.40	4.55	0.61	1	Royal Fern	3.52	5.90	1.68
3	Rice Cutgrass	9.23	0.87	0.09	2	Royal Fern	1.88	2.72	1.45
4	Royal Fern	4.64	0.86	0.18	3	Rice Cutgrass	0.16	0.03	0.17
4	Rice Cutgrass	10.09	1.00	0.09	4	Rice Cutgrass	0.13	0.02	0.11
5	Rice Cutgrass	3.28	0.78	0.23	5	Royal fern	3.62	2.16	0.60
5	Royal Fern	13.82	0.61	0.04	5	Rice Cutgrass	6.22	0.60	0.10
7	Rice Cutgrass	24.33	1.29	0.05	6	Rice Cutgrass	0.09	0.06	0.61
8	Netted Chain Fern	7.44	8.05	1.08	6	Netted Chain Fern	1.47	4.77	3.24
8	Rice Cutgrass	2.74	1.34	0.49	7	Netted Chain Fern	0.82	0.93	1.13
9	Royal Fern	2.17	1.57	0.72	8	Netted Chain Fern	0.18	1.27	6.88
9	Rice Cutgrass	0.13	0.20	1.59	8	Rice Cutgrass	0.14	1.35	9.45
10	Royal Fern	5.14	2.87	0.56	9	Netted Chain Fern	0.17	0.52	3.07
11	Rice Cutgrass	3.98	1.45	0.36	9	Netted Chain Fern	0.05	0.33	6.57
12	Netted Chain Fern	3.66	0.78	0.21	10	Rice Cutgrass	0.08	0.03	0.44
					11	Netted Chain Fern	2.28	5.78	2.54
					12	Netted Chain Fern	0.08	0.24	2.93
Overall Mean		7.00	1.88	0.45	Overall Mean		1.31	1.67	2.56
Overall Standard Deviation		6.16	2.09	0.45	Overall Standard Deviation		1.80	2.06	2.79
Mean (±SD) Royal Fern		6.6 ± 4.4	2.1 ± 1.6	0.4 ± 0.3	Mean (±SD) Royal Fern		3.0 ± 0.9	3.6 ± 2.0	1.2 ± 0.6
Mean (±SD) Netted Chain Fern		5.5 ± 2.6	4.4 ± 5.1	0.6 ± 0.6	Mean (±SD) Netted Chain Fern		1.0 ± 0.9	2.0 ± 2.3	3.8 ± 2.2
Mean (±SD) Rice Cutgrass		8.5 ± 8.6	1.0 ± 4.3	0.4 ± 0.5	Mean (±SD) Rice Cutgrass		1.1 ± 2.5	0.4 ± 0.6	1.9 ± 2.6

Table 6. Concentration of ^{137}Cs (Bq g^{-1}) as a function of depth in sediment cores collected at R-Canal and Pond A. Blanks within the table mean that the core was shallow and not taken to the depth indicated. Values can be multiplied by 27 to obtain concentrations in units of pCi g^{-1} .

Site	Depth of Core Slice (cm)				
	0-2.5	2.5-5.0	5.0-7.5	7.5-10	10-12.5
1	7.3	8.06	8.3	12.3	10.2
2	16.9	6.4	6.1	-	-
3	2.2	5.9	4.2	3.4	2.4
4	24.1	19.2	10.9	-	-
5	2.0	6.3	-	-	-
6	7.6	19.0	45.7	42.5	29.1
7	2.0	1.7	2.3	1.8	-
8	2.0	19.2	1.9	1.4	1.2
9	1.6	2.8	8.1	36.5	31.4
10	2.4	14.8	8.0	2.8	1.0
11	2.3	16.0	3.3	-	-
12	0.7	1.1	1.9	0.9	-
13	3.0	3.6	9.5	2.5	0.6
14	1.3	1.9	1.5	1.7	1.8
15	1.1	1.4	1.9	12.4	12.9
16	1.9	21.0	9.7	3.8	-
17	1.9	4.3	8.0	5.0	2.8
18	2.8	13.3	4.6	1.4	1.2
19	5.1	9.6	16.6	10.0	6.6
20	1.7	2.2	1.9	-	-
21	1.9	5.5	1.7	1.5	1.1
22	59.6	6.1	4.6	2.0	-
23	3.9	1.7	0.3	0.1	-
24	1.8	4.7	-	-	-
25	3.6	19.7	7.9	2.8	3.5
26	9.2	19.2	4.6	1.7	0.6
27	1.6	13.3	2.5	0.7	0.5
28	3.5	11.8	4.3	2.6	1.9
29	1.8	1.7	1.8	3.7	6.3
Mean	6.1	9.0	6.7	6.7	6.4
SD	11.5	6.9	8.6	10.9	9.4

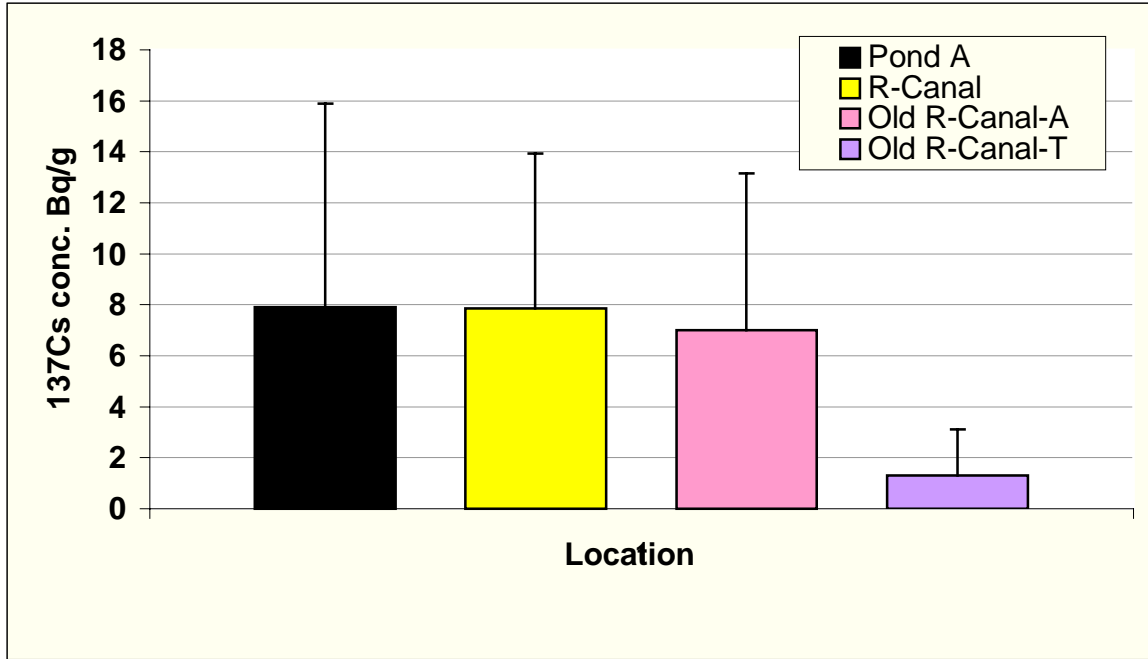


Figure 3. Average concentrations of ¹³⁷Cs (Bq g⁻¹) in sediment at Pond A, R-Canal and Old R-Canal. Data for Pond A are from Abraham et al. (2000). Data for both aquatic (A) and terrestrial sites (T) are reported for Old R-Canal. Values can be multiplied by 27 to obtain results in units of pCi g⁻¹.

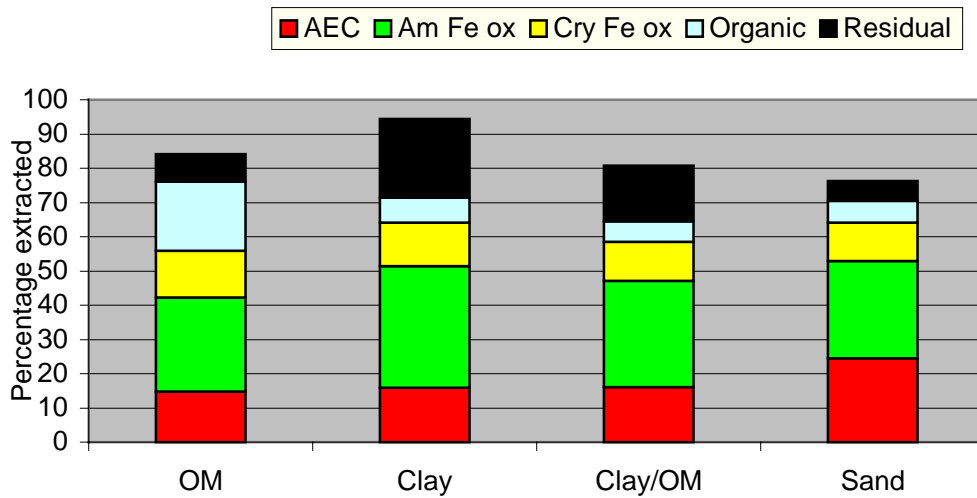


Figure 4. Partition of ¹³⁷Cs among five fractions in four types of sediment from R-Canal and Pond A; AEC – adsorbed/exchangeable/carbonate phase, Am Fe ox – amorphous Fe oxyhydroxide, Cry Fe ox – crystalline Fe oxide, Organic – organic phase, Residual – silicates.

Table 7. ^{137}Cs concentrations in leaves and stems of water lily collected at R-Canal and Pond A.

Leaf ^{137}Cs Concentrations (Bq g^{-1})						Stem ^{137}Cs Concentrations (Bq g^{-1})					
Location	Rep 1	Rep 2	Rep 3	Average	Stdev.	Location	Rep 1	Rep 2	Rep 3	Average	Stdev.
1	1.53	1.18	1.69	1.46	0.26	1	2.20	1.90	2.95	2.35	0.54
2	1.54	1.71	1.28	1.51	0.21	2	2.62	3.19	2.44	2.75	0.39
3	1.14	1.47	1.45	1.35	0.18	3	1.74	1.50	1.87	1.70	0.18
4	1.09	1.05	1.55	1.23	0.27	4	1.82	2.07	2.24	2.04	0.21
5	1.18	1.39	1.02	1.20	0.18	5	1.96	2.16	2.23	2.12	0.13
6	0.80	1.30	0.94	1.01	0.25	6	1.20	1.58	1.37	1.39	0.19
7	1.10	1.28	0.95	1.11	0.16	7	1.98	4.24	1.63	2.62	1.41
8	1.0	1.09		1.07	0.02	8	2.30	1.81		2.05	0.35
9	1.29	1.11	1.20	1.20	0.09	9	2.77	1.85	2.31	2.31	0.46
10	0.87	1.05	0.89	0.94	0.10	10	0.58	1.76	1.26	1.20	0.59
11	0.97	1.07	1.11	1.05	0.07	11	1.72	1.81	2.45	1.99	0.40
12	1.28	1.33	1.10	1.24	0.11	12	1.92	2.35	1.69	1.99	0.33
13	1.02	0.98	0.92	0.97	0.04	13	1.25	1.41	1.52	1.39	0.13
14	0.77	0.90	0.89	0.85	0.07	14	1.55	1.66	1.91	1.71	0.18
15	1.22	0.81	0.89	0.97	0.21	15	1.32	1.48	1.55	1.45	0.11
16	0.91	0.89	0.73	0.85	0.09	16	1.69	1.28	1.48	1.48	0.20
Mean				1.13	0.24					1.91	0.60

4.5 ¹³⁷Cs BIOAVAILABILITY – CONCENTRATION RATIOS

In addition to determining activity concentrations of ¹³⁷Cs in the plants (Bq g⁻¹ dry plant), ¹³⁷Cs uptake was normalized for the varying sediment levels by calculating concentration ratios (CR = activity in dried plant / activity in dried sediment). The concentration ratios were calculated separately for each paired plant/sediment sample, and then the ratios were averaged across plants within each species. It is important to normalize the plant contaminant concentrations in this manner to provide insight as to the cause for low plant contaminant concentrations. Low plant contaminant concentrations could be the result of low sediment contaminant concentrations, the contaminant existing in a non-biologically available form, or avoidance of the sediment contaminant by plant roots. By expressing the plant uptake as concentration ratios, it is possible to rule out the first cause, that is, that sediment contaminant concentrations were low.

The greatest average concentration ratio was obtained for netted fern from the terrestrial flood plain (3.8, Table 5). The average concentration ratios for water lily from R-Canal and Pond A for leaves and stems were 0.18 and 0.3, respectively (Fig. 5: Table 8). At the Old R Discharge Canal, the average ¹³⁷Cs concentration ratios for the three plant species recovered from the aquatic sites were 0.4 ± 0.4 and for the terrestrial flood plain sites were 2.6 ± 2.8 . This indicates that the CRs from plants collected in terrestrial sites were higher (Table 5). Higher concentration ratios for plants growing in terrestrial sites do not follow the findings of previous research by Hinton et al. (1999). They found CR values to be lower for terrestrial species than for wetland species (plants collected at Par Pond, SRS, Aiken SC), 8.1 and 14.8, respectively. Concentration ratios from Hinton's study are among the largest reported in the literature. Some recorded values were as high as 365, and the overall arithmetic mean for all species across 3 years was 12.5 (n = 239). Hinton et al. (1999) were studying Cs uptake in a contaminated reservoir whose water levels had recently decreased by 50%. They attributed the high CRs they observed, in part, to the conditions during the draw down of the reservoir. The species studied differed from those reported here.

Even though the CR values in this study were not as high as what Hinton et al. (1999) reported, they were 4.5 and 25.6 times greater (for aquatic and terrestrial sites) than what the National Council on Radiological Protection and Measurements recommend for a default value (0.1; NCRP, 1989). This points out, yet again, that Cs mobility is greater on the SRS than at most other sites.

Table 8. ^{137}Cs concentration ratios (Bq kg^{-1} dried plant / Bq kg^{-1} dried sediment) for water lily samples collected at R-canal and Pond A. Sediment concentrations of ^{137}Cs were averaged from the 0 - 12.5 cm depth.

Location	^{137}Cs Concentration Ratios of Leaves					^{137}Cs Concentration Ratios of Stems				
	Rep 1	Rep 2	Rep 3	Average	Stdev	Rep 1	Rep 2	Rep 3	Average	Stdev
1	0.058	0.045	0.0641	0.056	0.010	0.084	0.072	0.112	0.089	0.021
2	0.134	0.148	0.1115	0.131	0.019	0.227	0.277	0.212	0.239	0.034
3	0.123	0.158	0.1564	0.146	0.020	0.187	0.162	0.202	0.184	0.020
4	0.102	0.098	0.1439	0.114	0.026	0.169	0.192	0.208	0.190	0.019
5	0.223	0.263	0.1940	0.227	0.035	0.371	0.410	0.421	0.401	0.026
6	0.080	0.130	0.0944	0.102	0.026	0.121	0.159	0.138	0.139	0.019
7	0.371	0.432	0.3223	0.375	0.055	0.671	1.432	0.550	0.884	0.478
8	0.289	0.297	0.0000	0.196	0.169	0.628	0.493	0.000	0.374	0.331
9	0.092	0.079	0.0849	0.085	0.007	0.196	0.131	0.164	0.164	0.033
10	0.087	0.105	0.0891	0.094	0.010	0.058	0.176	0.126	0.120	0.059
11	0.171	0.189	0.1951	0.185	0.013	0.302	0.318	0.431	0.350	0.070
12	0.178	0.185	0.1535	0.172	0.016	0.266	0.327	0.235	0.276	0.047
13	0.217	0.208	0.1964	0.207	0.010	0.265	0.299	0.323	0.296	0.029
14	0.201	0.235	0.2311	0.222	0.019	0.403	0.433	0.497	0.444	0.048
15	0.370	0.248	0.2718	0.297	0.065	0.400	0.449	0.470	0.440	0.036
16	0.208	0.204	0.1680	0.193	0.022	0.386	0.292	0.337	0.338	0.047
Mean	0.181	0.189	0.155	0.175	0.033	0.296	0.351	0.277	0.308	0.082
SD	0.097	0.096	0.082	0.083	0.040	0.176	0.314	0.160	0.192	0.129

5.0 SUMMARY

The objectives of this study were to provide more site-specific data for human and ecological risk analysis on the SRS. In this study the mobile fraction of ^{137}Cs , i.e., the fraction that could be taken up by plants, in sediments collected from R-Canal and Pond A was determined by sequential extractions of the sediments, and by directly measuring plant uptake. The propensity for plants to take up ^{137}Cs was calculated using a concentration ratio (CR = activity per dried plant / activity per dried sediment).

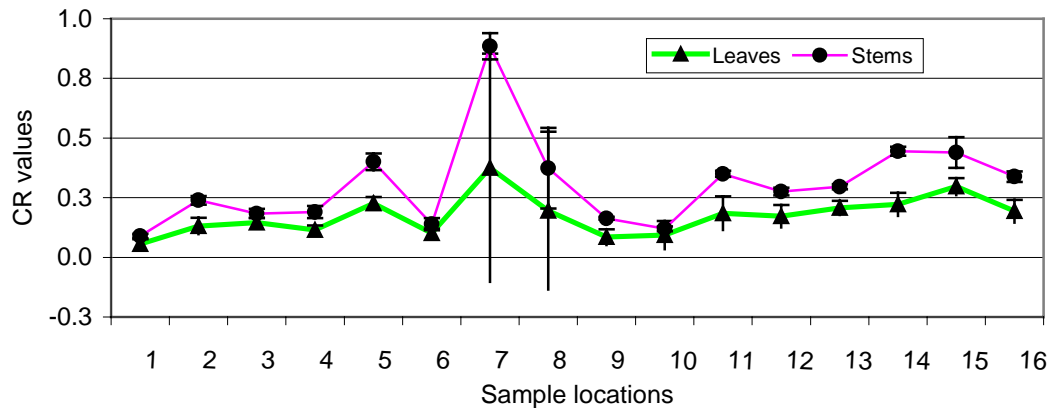


Figure 5. Mean (\pm SD) ^{137}Cs concentration ratios for water lily leaves and stems collected at R-Canal and Pond A; 3 replicates per location.

The study was conducted in R Canal, Pond A, and the Old R Discharge Canal. All these systems are considered wetlands and were contaminated with ^{137}Cs when R reactor was operating (prior to 1964).

The mean (\pm SD) ^{137}Cs concentration in the Old R Discharge Canal ($189 \pm 167 \text{ pCi g}^{-1}$) was no different from the mean for R Canal ($224 \pm 213 \text{ pCi g}^{-1}$) or Pond A ($213 \pm 205 \text{ pCi g}^{-1}$). Part of the motivation for sampling the Old R Discharge Canal was that gamma over-flight data indicated that ^{137}Cs contamination levels were higher there than in Pond A. This apparent discrepancy is likely the result of increased shielding of the gamma emissions by the greater depth of water in Pond A as compared to the Old R Discharge Canal. These findings indicate that remediation decisions about ^{137}Cs -contaminated wetlands could be misdirected if based solely on gamma over flight data.

Sequential extraction data indicated that between 50 and 85% of the ^{137}Cs in these sediments was strongly bonded and likely in a chemical form that was of limited bioavailability.

Estimates of concentration ratios were often in excess of 1.0, indicating that the plants concentrated ^{137}Cs in their tissue to levels greater than existed in the sediment. A review of

the literature indicated that ^{137}Cs concentration ratios >1.0 are common at the SRS, and have been attributed, in part, to the low abundance of Cs-binding clays.

Since predictions of plant ^{137}Cs concentrations based on sediment ^{137}Cs concentrations is tenuous and not captured well by a single parameter, such as a concentration ratio, it is recommended that risk assessment related to plants be based directly on plant ^{137}Cs concentrations. Relying solely on sediment ^{137}Cs concentration data, either total or that which is biologically available, could result in large errors in risk estimations.

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