Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy.

Changes in ^{137}Cs concentrations in soil and vegetation on the floodplain of the Savannah River over a 30 year period

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Abstract

^{137}Cs released during 1954 -1974 from nuclear production reactors on the Savannah River Site, a US Department of Energy nuclear materials production site in South Carolina, contaminated a portion of the Savannah River floodplain known as Creek Plantation. ^{137}Cs activity concentrations have been measured in Creek Plantation since 1974 making it possible to calculate effective half-lives for ^{137}Cs in soil and vegetation and assess the spatial distribution of contaminants on the floodplain. Activity concentrations in soil and vegetation were higher near the center of the floodplain than near the edges as a result of frequent inundation coupled with the presence of low areas that trapped contaminated sediments. ^{137}Cs activity was highest near the soil surface, but depth related differences diminished with time as a likely result of downward diffusion or leaching. Activity concentrations in vegetation were significantly related to concentrations in soil. The plant to soil concentration ratio (dry weight) averaged 0.49 and exhibited a slight but significant tendency to decrease with time. The effective half-lives for ^{137}Cs in shallow (0-7.6 cm) soil and in vegetation were 14.9 (95% CI = 12.5-

17.3) years and 11.6 (95% CI = 9.1-14.1) years, respectively, and rates of ^{137}Cs removal from shallow soil and vegetation did not differ significantly among sampling locations. Potential health risks on the Creek Plantation floodplain have declined more rapidly than expected on the basis of radioactive decay alone because of the relatively short effective half-life of ^{137}Cs.

Keywords: cesium, effective half-life, floodplain, contaminated soil, contaminated vegetation, Savannah River, Savannah River Site, ecological half-life, long-term change

1. INTRODUCTION

Contamination by long-lived radionuclides can affect decisions regarding land use for generations because of concerns about human and ecological health. A radionuclide of particular importance in this respect is ^{137}Cs, which has a physical half-life of 30.2 years, high fission yield, and high bioavailability due to its physiological similarity to potassium. The latter factor results in the uptake of ^{137}Cs by plants and its movement through aquatic and terrestrial food chains.

^{137}Cs contamination within an ecosystem can be expected to decline at a minimal rate based on radioactive decay. However, considerable research shows that rates of ^{137}Cs elimination from an ecosystem often differ from the radioactive decay rate due to physical, chemical, and biological processes that remove ^{137}Cs or reduce its biological availability (Whicker and Schultz 1982). The term "effective half-life", which includes both environmental processes and radioactive decay, is often used to describe the time required for a radionuclide (or other contaminant) to decrease by 50%. Effective half-lives are specific to particular media and locations because they are influenced by variable environmental factors like erosion, soil composition, chemical reactions, and leaching. In addition, effective half-lives may change over time as environmental processes change and chemical reactions progress (Smith et al. 2000). Effective half-lives are often shorter than the radioactive half-life, although not always (Stark et al. 2006).

Reductions in the activity of ^{137}Cs and other radionulides over time have been reported for a variety of environmental media contaminated by the Chernobyl accident (Prohl et al. 2006). Effective loss rates have also been described for fish, vegetation, and soil on and near Pacific islands contaminated by nuclear testing (Noshkin et al. 1997, Robison et al. 2003) and at nuclear material production sites in the United States (Bagshaw and Brisbin 1984, Paller, et al. 1999, Peles et al. 2000, Peles et al. 2002). Such work contributes information needed to intelligently plan for the long-term disposition of contaminated sites because it facilitates accurate projections regarding future risks of radiological contaminants to humans and other organisms. In the United States this information is likely to be of particular importance at Department of Energy (DOE) nuclear facilities where the focus has changed from production to remediation of contaminated land and facilities.

^{137}Cs released over 30 years ago from reactors on the Savannah River Site (SRS), a DOE nuclear site in South Carolina, contaminated a portion of the Savannah River floodplain. ^{137}Cs concentrations in floodplain soil and vegetation have been periodically measured in the contaminated area creating an opportunity to assess long-term changes in ^{137}Cs on the floodplain of a major river in a warm-temperate zone environment. Previous studies of this type have involved cool-temperate zone rivers (Kryshev et al. 1998, Aarkrog et al. 2000, Linnik et al. 2005) or smaller streams (Gladden et al. 1985). The objectives of this study were to evaluate the spatial distribution of ^{137}Cs on the floodplain and determine the effective loss-rates for ^{137}Cs in floodplain soil and vegetation.

2. METHODS AND MATERIALS

2.1 Study area

The SRS is an 800 km^2 reservation established in 1951 for the production of nuclear materials. It is located on the upper coastal plain of South Carolina near Aiken, South Carolina (USA). By 1955 there were five functioning nuclear reactors on the SRS.

Cooling water for the reactors was withdrawn from the Savannah River and released into cooling reservoirs or into streams that returned the water to the river after traveling through an extensive floodplain swamp contiguous with the river (Figure 1). Approximately 10.35 TBq of ^{137}Cs were discharged down Steel Creek from 1954–1974, mostly from P Reactor in the early 1960s. Water from Steel Creek flowed through the floodplain swamp when river levels were high, resulting in the deposition of radioactive materials between Steel Creek Landing and Little Hell Landing (Figure 1). Contamination of the floodplain occurred when the Savannah River stage exceeded 27.5 m mean sea level, which occurred approximately 21.5% of the time during 1954–1974 (Fledderman et al. 2007).

Part of the floodplain swamp affected by Steel Creek discharge lies in Creek Plantation, a privately owned area along the Savannah River on the southeastern border of the SRS. The portion of floodplain swamp within Creek Plantation is uninhabited, difficult to access, and is used as wildlife habitat and for occasional private hunts. Land elevations within the Creek Plantation floodplain are mostly about one to three meters higher than the river channel and rise near the landward border of the swamp, which is formed by a natural terrace. Sloughs and ponds occur in lower areas. The Creek Plantation floodplain swamp is forested by bald cypress *Taxodium distichum* and water tupelo *Nyssa aquatica* in wetter areas and by bottomland hardwoods at higher elevations. The soil group on the Savannah River floodplain is referred to as the Chastain-Tawcaw-Shellbluff Association (Wike et al. 2005). Chastain soils, which compose 60% of the association, are poorly drained and contain substantial clay. Tawcaw soils, about 20% of the association, are somewhat better drained and contain a combination of clay and loam.

Shellbluff soils, the smallest component at 15%, are well drained and composed mostly of loam.

2.2 Field methods

Ten trails were established through the Creek Plantation swamp in the early 1970s for the collection of soil and vegetation samples. Each trail consisted of a transect extending perpendicularly from the swamp margin to the Savannah River, except for Trail 3, which ran across the neck of a narrow peninsula formed by a loop of the Savannah River (Figure 2). With the exception of Trails 3 and 10, the trails ranged from 728 to 975 m in length and each had five to eight fixed sampling locations extending from near the river channel to near the swamp margin, although not at consistent intervals because of obstacles (such as ponds) that prevented sampling in some areas. Trail 3 had three sampling locations over 626 m and Trail 10, a very short trail where the floodplain was narrow, was 73 m long with three sampling locations.

Shallow (to a depth of about 7.6 cm) soil samples were collected from all sampling locations. In addition, core samples were collected at eight locations (one each near the middle of the floodplain on Trails 1, 3, 4, 5, 7, 8, 9, and 10) to evaluate the vertical distribution of ^{137}Cs in soil. Four samples in 7.6 cm increments were collected to a total depth of about 30.5 cm at these sites. Living herbaceous vegetation was also collected from the sampling locations. Bermuda grass (*Cynodon dactylon*) was collected when possible, but typical hydrophytic species of regional wetlands and floodplains were collected where Bermuda grass was unavailable. These species usually included members of the cut grass family (*Leersia* spp.) and sedge family (e.g., *Cyperus* spp.).

Soil and vegetation samples were dried for radiological analysis. Foreign matter (stones, twigs, etc.) was removed, and the remaining material was ground to pass through a #10 sieve and placed into a standard calibrated geometry (typically 200 or 500 g). Processed samples were weighed and counted on an HPGe detector of approximately 35% efficiency. Earlier samples were counted with NaI(Tl) solid scintillator or Ge(Li) semiconductor detectors. Samples were counted for 5000 seconds, resulting in an MDA of approximately 0.04 pCi/g (1.5 Bq/kg). All values were reported as dry weight.

Sampling was initiated in 1974, and the last year included in the analysis was 2004 for vegetation and 2005 for soil. Efforts were made to sample all 52 sampling locations during 1974 – 1977 and about every five years thereafter. A subset of 30 sites or fewer was often sampled during other years. Years in which samples were collected included 1974-1977, 1982, 1985-1991, 1993, 1994, 1996, 2000-2002, 2004, and 2005 (see Fledderman et al. 2007 for a detailed sampling history). The median number of years in which shallow soil samples were collected from each sampling location was nine with a maximum of 20 and minimum of three. Comparable values for vegetation samples were seven with a maximum of 16 and minimum of two. Soil cores for assessment of the depth distribution of ^{137}Cs were collected in 1976, 1990, 1996, 2000-2002, 2004, and 2005.

Other radionuclides besides ^{137}Cs were present in environmental media collected from the Creek Plantation floodplain. However, ^{137}Cs accounted for most of the radiation dose and is the subject of this paper.

2.3 Data analysis

The effective loss rate constant (λ_e), as used in this paper, subsumed all processes (including environmental processes and radioactive decay) that reduced ^{137}Cs concentrations over time. Reductions in ^{137}Cs activity concentrations were described by the equation $dc/dt = - \lambda_e c$, where c was the ^{137}Cs concentration in soil or vegetation (Bq/g^{-1}). Estimates of λ_e were made from the slopes of $\log_e$-transformed ^{137}Cs activity concentrations regressed on year. The effective half-life was estimated as $Te = \log_e 2/\lambda_e$. Some researchers use the term ecological half-life in lieu of effective half-life, while others define ecological half-life as all processes that reduce activity concentrations exclusive of radioactive decay. Ecological half-life is used in the latter sense in this paper and was computed from the effective half-life with the equation $Tc = TeTr/(Tr-Te)$, where Tc is the ecological half-life and Tr is the radioactive half-life.

Computing effective half-lives for ^{137}Cs as described above will produce accurate results only if additional ^{137}Cs inputs are null. Contamination of the Creek Plantation floodplain occurred primarily before the study began in 1974, but additional inputs of ^{137}Cs may have occurred after 1974 as a result of the downstream displacement of contaminated sediments by flooding. Although such inputs are believed to have been comparatively small, they were not quantified and represent a potential positive bias in the effective half-life computations presented herein. No attempt was made to correct for ^{137}Cs inputs from global fallout because this source was insignificant compared with contamination resulting from reactor releases (Paller et al. 1999).

Two approaches were used to investigate temporal changes in ^{137}Cs activity concentrations on the Creek Plantation floodplain. The first was to compute individual loss rate constants for each sampling location. The second was to combine the data from all sampling locations in analysis of covariance (ANCOVA) models that included time as a covariate. An advantage of the latter approach was that it made it possible to test the significance of spatial variables that could affect rates of ^{137}Cs change over time. Interaction terms were used to test for homogeneity of slopes in the ANCOVA models, and non-significant interaction terms were excluded from final model runs (Engqvist 2005). Variables included in each model will be discussed in detail in the Results. ^{137}Cs data were positively skewed necessitating logarithmic transformation to better adhere to the assumptions of ANCOVA. Means presented in this paper are geometric means with confidence intervals computed as shown in Sokal and Rohlf (1995).

Error variance in successive ^{137}Cs samples from the same location may exhibit a correlation that can affect the accuracy of statistical tests. The assumption of independent errors can be tested with the Durbin-Watson statistic (Montgomery et al. 2001). For the sample size encountered in this study ($n \leq 20$ for each sampling location), Durbin-Watson values under 1.5 are conservatively indicative of positive autocorrelation, which is the type of autocorrelation typically observed among time series data. Only six soil sampling locations and four vegetation sampling locations out of 52 locations in total were characterized by Durbin-Watson statistics ≤ 1.5.

3. RESULTS

3.1 ^{137}Cs in soil

^{137}Cs activity concentrations in Creek Plantation soil samples at a depth of 0-7.6 cm ranged from 0.02 to 19.52 Bq/g with a geometric mean of 0.49 (95% CI = 0.28 – 0.88) Bq/g when the study began in 1974/1975, (Table 1). At the end of the study in 2005, ^{137}Cs concentrations ranged from <0.01 to 2.21 Bq/g with a geometric mean of

0.11 (95% CI = 0.06-0.21) Bq/g. Activity concentrations decreased over time at all sampling locations except two (Table 1). The median percentage decrease for all locations was 81% over the duration of the collection period, which ranged from 16 to 31 years depending upon the sampling location (average of 29 years) (Table 1).

Only about half of the sampling locations (28 of 54) were characterized by statistically significant ($P<0.05$) regressions of log transformed ^{137}Cs activity concentrations on year, despite the fact that ^{137}Cs activity concentrations decreased at nearly all sites (Table 1). Regression slopes for locations with significant regressions ranged from -0.086 to -0.035 (Table 1), yielding ecological half-lives for ^{137}Cs in soil ranging from 8.1 to 19.6 years. Most slopes had large confidence intervals due to high variability among measurements relative to sample size (i.e., low statistical power), a factor that also contributed to the absence of significant regressions at some locations with large overall decreases in ^{137}Cs (Table 1). The significance of regression slope differences among locations was tested using ANCOVA with ^{137}Cs activity concentration as the dependent variable, year as the covariate, and sampling location as the categorical variable. The interaction between sampling location and year (i.e., heterogeneity of slopes) was not significant ($F=0.875$, df = 53, $P=0.718$), indicating that the null hypothesis of equal rates of ^{137}Cs decrease at all locations could not be rejected given the available sample sizes. A follow-up ANCOVA without an interaction term showed that year and location each had highly significant effects on the ^{137}Cs concentration ($F=32.31$, df = 53, $P<0.001$; and $F=141.02$, df = 1, $P<0.001$; respectively).

Spatial effects were explored in greater detail with a more complex ANCOVA that included the independent variables of trail (categorical), year (continuous), distance from the Savannah River (continuous), and quadratic effects for distance. The quadratic term was included to evaluate possible nonlinear effects of distance on ^{137}Cs activity concentrations. Since an objective of this analysis was to determine how ^{137}Cs distribution varied from the river bank to the upper edge of the floodplain, transects 3 and 10 were excluded because of atypical orientation (transect 3) or length (transect 10). Spatial differences in the rate of ^{137}Cs decrease over time would be manifested as significant interactions between trail and year, between distance and year, or among trail, distance, and year in this model. None of these effects were significant (at $P<0.05$), thus agreeing with the results of the previous ANCOVA. Significant factors included year, distance from the river, the quadratic effect for distance, and the interaction between trail and distance (Table 2). The significance effects for distance resulted from higher ^{137}Cs activity concentrations at intermediate distances from the river and lower concentrations near the river and the upper edge of the floodplain (Figure 3). The interaction between trail and distance indicated that differences among trails varied with the distance from the river. The model R^2 was 0.55 indicating that slightly over half of the variance in soil ^{137}Cs activity concentrations was explained by the terms in the model.

Because the preceding ANCOVAs indicated that reductions in ^{137}Cs over time did not differ spatially, ^{137}Cs data from all sampling locations were combined to compute a single overall decay rate for ^{137}Cs in soil for the entire Creek Plantation area. The regression slope of -0.037 was highly significant ($P<0.001$), and the 95% confidence limits for the regression line were narrow reflecting certainty regarding the overall trend of decrease (Figure 4). However, the R^2 was low (0.038) because of uncontrolled spatial variation in the model. Also, the model results were biased because a spatially nonrandom subset of locations was sampled during some years. To correct these problems, residuals were derived from a model with sampling location as the independent variable and $\log_e$ transformed concentration as the dependent variable. These residuals (representing concentrations with location related variance removed) were regressed on year to yield an R^2 of 0.229 ($P<0.001$) and regression coefficient of -0.047 (95% CI = -0.054 - -0.039). The effective half-life for Creek Plantation soil computed from this slope was 14.9 years (95% CI = 12.5 - 17.3 yrs).

ANCOVA of the soil depth data collected at eight sampling locations between 1990 and 2005 indicated that ^{137}Cs concentrations differed significantly with soil depth (Table 3). Concentrations were generally highest in the 0-7.6 cm soil stratum, but depth related differences decreased with time (Figure 5). Additional data indicating a decrease in depth related differences over time was provided by a single set of measurements from 1976. ^{137}Cs activity concentrations at a depth of 0-7.6 cm were 79 times higher than at a depth of 22.9-30.5 cm in 1976 but only 13 times higher in 1990 and three times higher in 2005.

<u>3.2 ^{137}Cs in vegetation</u>

Activity concentrations of ^{137}Cs in vegetation ranged from 0.02 to 8.70 Bq/g with a geometric mean of 0.12 (95% CI = 0.07 – 0.21) Bq/g at the start of the study in 1974 (Table 4). At the end of the vegetation study period in 2004, activity concentrations in vegetation ranged from <0.01 to 0.41 Bq/g with a geometric mean of 0.02 (95% CI = 0.01 – 0.06) Bq/g. Activity concentrations in vegetation decreased at 47 of the 54 sampling stations. The median percentage decrease for all locations was 74% over an average measurement period of 22 years.

Only 13 of the 54 sampling locations had statistically significant regressions of log transformed ^{137}Cs activity concentrations in vegetation on year (Table 4). Slopes for these regressions ranged from -0.26 to 0.06. Most of the half-lives calculated from these slopes ranged from 3.4 to 8.3 years, although one was 23.2 years. Like the soil regressions, the vegetation regression slopes had large confidence intervals as a consequence of high variability among measurements relative to sample size – a problem that also contributed to a lack of significant regressions at many locations. Despite these differences, ANCOVA with ^{137}Cs activity concentrations in vegetation as the dependent variable, year as the covariate and sampling location as the categorical variable indicated that the rate of decrease in ^{137}Cs concentration over time did not differ significantly ($F=1.300$, $df = 53$, $P=0.093$) among sampling locations (i.e., the slopes were not heterogeneous), paralleling results with the soil data.

Like the soil data, spatial differences in the rate of decrease in ^{137}Cs in vegetation over time were explored further with a more complex ANCOVA that included the independent variables of trail (categorical), year (continuous), distance from the Savannah River (continuous), and quadratic effects for year and distance. Interactions between year and trail and between year and distance were not significant showing that the rate of ^{137}Cs decrease in vegetation over time did not significantly differ among sampling locations (Table 5). Significant factors included year, the interaction between trail and distance and distance from the river. These results generally paralleled the results for soil, including the tendency for activity concentrations to be higher at intermediate distances from the river. The R^2 for the vegetation ANCOVA model was 0.39, which was somewhat lower than the R^2 for the soil ANCOVA model (0.55).

Because reductions in ^{137}Cs over time did not differ significantly among sampling locations, ^{137}Cs data from all sampling locations were pooled to compute an overall ecological half-life for vegetation as done with soil. Like the comparable soil model, the regression coefficient of -0.052 was highly significant ($P<0.001$), but the R^2 was low (0.059) because of spatial variations in activity concentrations (Figure 6). As with soil, ^{137}Cs concentrations were replaced with residuals from a model with sampling location as the independent variable and concentration as the dependent variable. These residuals were regressed on year yielding an R^2 of 0.18 ($P<0.001$) and a regression coefficient of -0.060 (95% CI = -0.073 – -0.047). The ecological half-life for Creek Plantation vegetation computed from this slope was 11.6 (95% CI = 9.1 – 14.1) years.

Linear regression showed that ^{137}Cs activity concentrations in soil explained 52% of the variance in ^{137}Cs concentrations in vegetation collected from the same sample sites at the same time (Figure 7). Regressions of vegetation ^{137}Cs concentrations on soil ^{137}Cs concentrations were also computed for individual years to investigate the effects of space alone because the preceding model subsumed spatial and temporal covariance. The average R^2 for these regressions was 0.49 (0.21-0.77), and all years with sample sizes of 20 or greater (10 years) had significant ($P<0.05$) regressions. Concentration ratios (^{137}Cs in plants/^{137}Cs in soil dry weight collected at the same locations and times) averaged 0.45 (95% CI = 0.33-0.57) during the 1974-2005 study period. There was a weak but statistically significant ($P=0.0256$) decrease in CRs with time as indicated by linear regression of $\log_{10}$ transformed CRs on year (log transformation was needed to achieve normality and homogeneity of variance, Figure 8).

4. DISCUSSION

The only prominent and readily interpretable spatial pattern in ^{137}Cs distribution within the Creek Plantation study area was associated with distance from the river. Activity concentrations in both soil and vegetation were comparatively low near the river, rose near the middle of the floodplain, and decreased near the upper edge of the floodplain. Low concentrations near the upper edge of the floodplain likely resulted from reduced deposition of ^{137}Cs due to infrequent inundation. Low concentrations near the river bank, which presumably flooded more frequently than locations farther inland, may reflect the removal of contaminated soil by erosion but could also reflect the deposition of relatively clean sediments upon previously deposited contaminated sediments. Higher levels of ^{137}Cs in the middle of the floodplain presumably resulted from relatively frequent inundation, which allowed for the deposition of contaminated sediments, coupled with the presence of low areas that trapped them. Temporarily flooded wetlands can act as sinks for ^{137}Cs, especially when they have mineral soils with a high clay content (Stark et al. 2006). Linnik et al. (2005) reported that radioactive contamination on the Balchug River floodplain was greater in low-lying areas characterized by frequent flooding and sediment deposition and lower at higher elevations that flooded less.

Much of the variance in ^{137}Cs concentrations in soil was unexplained by year or the spatial variables measured in this study. Some of this unexplained variance was likely the result of small-scale variations in topography, vegetation cover, and soil composition that affected ^{137}Cs deposition and retention. Linnik et al. (2205) reported that the distribution of ^{137}Cs on the Balchug River floodplain was complex and related to the thickness and composition of alluvial deposits, which were affected by small differences in local relief, orientation of the relief, and vegetation cover. Similarly, Gladden et al. (1985) reported that gamma exposure rates on the Steel Creek floodplain were spatially heterogeneous and that high exposure rates were usually restricted to small areas. They attributed this to the morphological complexity of the floodplain on which deposition and chemical sorption potentials varied spatially.

Activity concentrations of ^{137}Cs were higher near the surface in Creek Plantation soils. Examination of contaminated soils from Sweden about 2.5 and 4.5 years after the Chernobyl accident showed that some downward migration of ^{137}Cs occurred in more humus rich and higher pH soils, while almost all ^{137}Cs remained near the surface in humus poor and lower pH soils (Andersson and Roed 1994). Clay, which is widespread in Creek Plantation soils (Wike et al. 2005), can bind ^{137}Cs and slow its downward movement. However, diminishment of the vertical gradient of ^{137}Cs in Creek Plantation soil over time (Figure 5) indicated that some downward migration of ^{137}CS occurred as a likely result of diffusion or leaching. This movement probably contributed to the reduction of ^{137}Cs activity concentrations in shallow soil. Downward migration of ^{137}Cs can affect ^{137}Cs removal rates at different soil depths, and the Creek Plantation data showed that decreases in ^{137}Cs occurred more slowly in deeper soils than in shallow soils. Vertical redistribution of ^{137}Cs implies that effective half-lives calculated for ^{137}Cs (and other radionuclides) in soil are dependent upon the depth of the soil sample. Pröhl et al. (2006) suggested that the ecological half-life concept is questionable for media, such as soils, in which radionuclides are not uniformly distributed and subject to redistribution.

The spatial distribution of ^{137}Cs concentrations in vegetation from the Creek Plantation floodplain was related to the distribution in soil. This relationship supported the use of concentration ratios (CRs) to represent the bioavailability of ^{137}Cs to plants on the Creek Plantation floodplain. The average CR of 0.45 was higher than typical for soils with moderate to high nutrient levels and pHs but not exceptional for other types of soils (Frissel et al. 2002). Considerably higher CRs have been reported for cattails *Sagittaria latifolia* in the Steel Creel delta (3.1) and for wetland (14.9) and terrestrial (8.1) vegetation growing on the exposed lake bed of Par Pond, a contaminated reservoir that was partly drained (Pinder et al. 1980, Hinton et al. 1999). Although the uptake of ^{137}Cs by plants is affected by ^{137}Cs concentrations in soil (Pinder et al. 1980, Absalom et al. 2000), other factors are also important including soil clay and organic matter content, the exchangeable potassium concentration, pH, cation exchange capacity, species-specific physiological differences, and other factors (Hinton et al. 1999, Absalom et al. 2000). About half of the variance in the ^{137}Cs concentrations in Creek Plantation vegetation was not explained by ^{137}Cs concentrations in soil indicating the possible influence of such factors.

The 14.9 year effective half-life of ^{137}Cs in shallow (0-7.6 cm) Creek Plantation soil was about half of what would be expected on the basis of radiological decay alone indicating that environmental processes were important in removing ^{137}Cs. ^{137}Cs removal associated with environmental processes, which can be subsumed under the terms "environmental loss" or "ecological loss", had a half-life of 29.4 years on the Creek Plantation floodplain. This is comparable to the 32.8 year ecological half-life calculated by Pröhl et al. (2006) for ^{137}Cs in shallow (0.0-2.5 cm) clay soils but longer than the ecological half-lives they calculated for ^{137}Cs in shallow sand (8.1 yrs) or shallow loam soils (4.1 yrs). The removal rate of ^{137}Cs from shallow Creek Plantation soils, 81% over an average of 29 years, was also lower than the 36% over three years reported by Hinton et al. (1999) for soils on the Par Pond Lake bed. They suggested that the high removal rate from the recently exposed Par Pond soil was due to erosion, leaching, and/or depletion by rapidly growing vegetation. Such processes may diminish with time as vegetation growth provides greater protection from erosion and as ^{137}Cs binds more tightly to soil particles. Creek Plantation soils have a high clay content and were protected by extensive vegetation cover (Wike et al. 2005), two characteristics that could reduce the rate of ^{137}Cs loss and lengthen the ecological half-life.

The effective half-life of 11.6 years for ^{137}Cs in plants growing on the Creek Plantation floodplain was longer than the half-lives of 4.9 – 8.4 years reported by Peles et al. (2002) for aquatic and terrestrial plants collected from the channel and floodplain of Steel Creek on the SRS. They noted that half-lives for Steel Creek vegetation were shorter than half-lives for macrophytes collected from Pond B (11.0-12.7 years), a contaminated reservoir on the SRS, and attributed the difference to the relatively rapid loss of ^{137}Cs to downstream transport processes such as erosion and sedimentation in Steel Creek. The Creek Plantation floodplain may be more similar to Pond B than to Steel Creek because erosion, especially at higher floodplain elevations is minimized by infrequent flooding and by vegetation cover. The effective half-life for ^{137}Cs in the leaves of trees growing on coral islands in the Pacific that had calcium carbonate soils subject to substantial leaching was 8.5 years (Robison et al. 2003), and a recently reported effective half-life for ^{137}Cs in periphyton collected from the Susquehanna River

(Pennsylvania) was 9.2 years (Cehn 2007). Excluding Steel Creek, where rapid elimination of ^{137}Cs may have been facilitated by high water flow, half-lives in the preceding environments, including Creek Plantation, were fairly similar (8.5-12.7 years). However, much longer and shorter half-lives have been reported for vegetation collected from soils in Europe that were contaminated by fallout from Chernobyl (Pröhl et al. 2006), indicating that differing environmental conditions can result in large differences in the rate at which ^{137}Cs is removed from ecosystems. Another factor that can affect effective half lives is the time since contamination. Processes that determine radionuclide transfer shortly after contamination has occurred result in more rapid losses than those that prevail later (Smith et al. 1999).

The effective half-life of ^{137}Cs in Creek Plantation vegetation (11.6 years) was somewhat shorter than in Creek Plantation soil (14.9 years). This could have resulted from decreases in the bioavailability of ^{137}Cs as a result of stronger binding in the soil over time, a hypothesis supported by decreases in soil to plant CRs over time, although CR variability was substantial and the rate of decrease slow (Figure 8). A reduction in the availability of ^{137}Cs and other radionuclides to plants over time has been observed in other studies (Noordijk et al. 1992). Hinton et al. (1999) showed that ^{137}Cs activity concentrations in plants growing on the exposed sediments of Par Pond decreased more rapidly than activity concentrations in soil. Extractions of sediment samples with ammonium acetate indicated a decrease in the availability of ^{137}Cs during their three year study. Knox et al. (2001) performed sequential extractions on wetland sediments from the SRS contaminated over 30 years earlier and found that 50 – 85% of the ^{137}Cs was strongly bound to the soil and likely of limited availability to plants.

The relatively short effective half-life of ^{137}Cs in the Creek Plantation floodplain implies that potential health risks associated with radiation exposure have declined more quickly than expected. These findings parallel those from other ecosystems and show that natural processes of recovery have the potential to restore radionuclide contaminated ecosystems more rapidly than can radioactive decay alone.

ACKNOWLEDGEMENTS

We thank the many individuals that collected and analyzed soil and vegetation samples during this study. This report was developed during US Department of Energy Contract No. DE-AC09-96SR18500.

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Table 1. Temporal changes in ^{137}Cs activity concentrations in soil at different distances from the Savannah River along 10 trails in Creek Plantation. Also shown are the results for regressions of $\log_e$ transformed ^{137}Cs on year and the half-lives for ^{137}Cs in soil at locations with significant (P<0.05) regression results.

Trail	Dist (m)	Initial conc. (Bq/g)	Final conc. (Bq/g)	% change	Initial/ final (years)	n	P	Slope (95% CI)	$T_{1/2}$
1	0	1.15	0.11	-90	74/05	6	0.003	-0.082 (-0.116 to-0.048)	8.5
1	178	0.85	0.27	-69	74/05	14	0.004	-0.037 (-0.059 to-0.014)	18.9
1	358	4.67	0.47	-90	74/05	6	0.024	-0.063 (-0.113 to-0.014)	10.9
1	550	12.78	1.47	-88	74/05	14	<0.001	-0.054 (-0.079 to-0.029)	12.9
1	655	7.19	2.11	-71	74/05	11	0.089		
1	805	0.11	0.01	-88	74/05	14	0.019	-0.035 (-0.064 to-0.007)	19.6
2	0	0.04	0.02	-54	74/05	6	0.090		
2	207	0.07	0.04	-52	74/05	10	<0.001	-0.045 (-0.065 to-0.025)	15.5
2	405	0.15	0.01	-90	74/05	5	<0.001	-0.071 (-0.087 to-0.056)	9.7
2	597	0.33	0.03	-92	74/05	5	0.024	-0.085 (-0.148 to-0.021)	8.2
2	799	1.74	0.35	-80	74/05	6	0.357		
2	945	4.52	1.23	-73	74/05	15	0.241		
2	975	0.11	0.01	-88	74/05	10	0.013	-0.077 (-0.133 to-0.022)	9.0
3	0	0.02	0.02	-3	74/00	8	0.121		
3	280	0.19	0.03	-83	74/04	15	0.204		
3	626	0.04	0.00	-92	74/00	7	0.054		
4	0	0.04	0.01	-65	74/05	6	0.070		
4	293	0.48	0.22	-54	74/05	10	0.011	-0.051 (-0.086 to-0.016)	13.6
4	379	3.19	0.66	-79	74/05	6	0.149		
4	515	2.67	0.91	-66	74/05	6	0.090		
4	579	6.93	1.86	-73	74/05	15	0.004	-0.047 (-0.076 to-0.017)	14.9
4	728	0.07	0.02	-69	74/05	9	0.053		
5	0	0.04	0.01	-85	74/05	5	0.007	-0.074 (-0.110 to-0.038)	9.3
5	533	1.26	0.17	-86	74/05	13	0.006	-0.049 (-0.080 to-0.017)	14.2
5	573	5.19	0.44	-92	74/05	5	0.003	-0.074 (-0.099 to-0.049)	9.4
5	640	9.63	1.50	-84	74/05	18	0.017	-0.072 (-0.130 to-0.015)	9.6

Table 1. continued.

5	773	0.04	0.02	-58	74/05	13	0.008	-0.059 (-0.099 to-0.019)	11.7
6	0	0.04	0.01	-67	74/05	6	0.045	-0.040 (-0.078 to-0.001)	17.5
6	549	1.85	0.18	-90	74/05	10	<0.001	-0.057 (-0.075 to-0.039)	12.1
6	701	5.93	2.09	-65	74/90	3	0.115		
6	771	11.11	1.31	-88	74/96	11	0.001	-0.081 (-0.120 to-0.042)	8.5
6	817	0.15	0.03	-78	74/05	10	0.023	-0.036 (-0.065 to-0.006)	19.4
7	0	0.07	0.01	-85	74/00	5	0.021	-0.058 (-0.100 to-0.017)	11.9
7	579	0.22	0.05	-78	74/00	9	0.019	-0.041 (-0.073 to-0.009)	16.9
7	792	19.52	1.14	-94	74/00	7	0.121		
7	823	0.11	0.72	550	74/02	13	0.139		
7	945	0.56	0.11	-81	82/04	6	0.450		
7	975	0.07	0.04	-50	82/00	5	0.297		
8	0	0.44	0.01	-99	74/00	5	0.061		
8	168	0.15	0.03	-81	74/00	9	0.046	-0.038 (-0.075 to-0.001)	18.2
8	279	0.04	0.04	-1	74/00	5	0.963		
8	445	0.19	0.05	-76	74/00	5	0.122		
8	611	0.22	0.03	-87	74/00	4	0.141		
8	814	2.33	0.44	-81	74/04	10	0.140		
8	884	4.22	0.04	-99	74/02	8	0.587		
8	914	0.19	0.03	-85	74/05	10	0.035	-0.062 (-0.118 to-0.005)	11.2
9	0	0.07	0.01	-81	74/05	7	0.025	-0.045 (-0.082 to-0.009)	15.3
9	512	4.44	0.64	-86	74/05	6	0.006	-0.058 (-0.088 to-0.028)	12.0
9	620	4.96	0.30	-94	74/05	10	<0.001	-0.086 (-0.099 to-0.073)	8.1
9	671	0.04	0.98	2540	74/05	15	0.801		
9	770	0.04	0.01	-71	75/05	9	0.095		
10	0	0.89	0.27	-69	75/05	13	0.349		
10	30	1.89	0.03	-98	74/05	20	<0.001	-0.058 (-0.087 to-0.029)	12.0
10	73	0.19	0.15	-21	74/05	14	0.110		

Table 2. ANCOVA table for factors affecting concentrations of ^{137}Cs in soil at different distances from the Savannah River along 10 trails in Creek Plantation (n=420). Non-significant interaction terms were excluded from the model.

Source of variation	Sum-of-Squares	df	Mean-Square	F-ratio	P
Trail	32.84	7	4.69	2.40	0.021
Distance	483.41	1	483.41	246.93	<0.001
Distance squared	426.18	1	426.18	217.70	<0.001
Year	77.93	1	77.93	39.81	<0.001
Trail-distance interaction	103.20	7	14.74	7.53	<0.001
Trail-distance squared interaction	240.36	7	34.34	17.54	<0.001
Error	773.28	395	1.96		

Table 3. ANCOVA table for activity concentrations of ^{137}Cs at different soil depths in Creek Plantation (n=186). Non-significant interaction terms were excluded from the model.

Source of variation	Sum-of-Squares	df	Mean-Square	F-ratio	P
Year	2.56	1	2.56	2.74	0.100
Sampling location	25.36	7	3.62	3.89	0.001
Depth	46.36	3	15.45	16.58	<0.001
Trail-year interaction	25.17	7	3.60	3.86	0.001
Error	155.66	167	0.93		

Table 4. Temporal changes in ^{137}Cs activity concentrations in vegetation at different distances from the Savannah River along 10 trails in Creek Plantation. Also shown are the results for the regression of $\log_e$ transformed ^{137}Cs on year and half-lives for ^{137}Cs in vegetation at locations with significant ($P<0.05$) regression results.

Trail	Dist (m)	Initial conc. (Bq/g)	Final conc. (Bq/g)	% change	Duration (years)	n	P	Slope (95% CI)	$T_{1/2}$
1	0	0.07	0.01	-88	74-96	3	0.426		
1	178	0.74	0.02	-98	74-00	12	0.006	-0.172 (-0.281 to-0.063)	4.0
1	358	0.11	0.14	28	74-96	4	0.905		
1	550	4.52	0.09	-98	74/00	13	0.017	-0.162 (-0.288 to-0.036)	4.3
1	655	0.81	0.06	-93	74-04	9	0.015	-0.092 (-0.160 to-0.025)	7.5
1	805	0.07	<0.01	-97	74-00	12	0.01	-0.107 (-0.182 to-0.032)	6.5
2	0	0.02	0.01	-47	74-96	4	0.007	-0.030 (-0.041 to-0.019)	23.2
2	207	0.11	<0.01	-99	74-00	7	0.053		
2	405	0.04	0.01	-63	74-00	5	0.183		
2	597	0.02	0.01	-59	74-00	5	0.313		
2	799	0.07	0.01	-90	74-96	4	0.391		
2	945	5.33	0.03	-99	74-04	12	<0.001	-0.152 (-0.216 to-0.088)	4.6
2	975	0.04	0.01	-74	74-00	8	0.734		
3	0	0.02	0.01	-68	74-90	6	0.151		
3	280	0.02	<0.01	-74	76-04	11	0.207		
3	626	0.02	<0.01	-94	74-00	8	0.027	-0.130 (-0.239 to-0.021)	5.3
4	0	0.07	<0.01	-96	74-96	4	0.073		
4	293	0.19	0.08	-57	74-96	8	0.554		
4	379	0.56	0.10	-81	74-96	4	0.518		
4	515	0.70	0.11	-85	74-90	3	0.153		
4	579	3.63	0.05	-99	74-04	12	0.64		
4	728	0.04	<0.01	-98	74-90	6	0.019	-0.204 (-0.354 to-0.054)	3.4
5	0	0.04	0.01	-62	74-96	3	0.469		
5	533	0.04	0.10	163	74-96	11	0.444		
5	573	0.56	0.18	-68	74-90	3	0.82		

Table 4. continued.

5	640	1.33	0.41	-69	74-04	16	0.366		
5	773	0.02	0.16	782	74-00	11	0.927		
6	0	0.04	0.01	-77	74-96	4	0.175		
6	549	1.74	0.10	-94	74-96	7	0.081		
6	701	0.67	0.96	44	74-75	2			
6	771	8.70	0.03	-100	74-04	12	0.178		
6	817	0.04	0.01	-75	74-96	7	0.087		
7	0	0.04	<0.01	-98	74-90	3	0.13		
7	579	0.04	0.02	-50	74-/96	7	0.373		
7	792	2.81	2.36	-16	74-96	7	0.882		
7	823	0.02	0.01	-64	74-04	9	0.286		
7	945	0.07	0.01	-87	90-96	2			
7	975	0.01	<0.01		90-96	2			
8	0	0.15	<0.01	-98	74-96	4	0.091		
8	168	0.04	<0.01	-88	74-96	7	0.048	-0.123 (-0.244 to-0.002)	5.6
8	279	0.02	0.01	-24	74-96	4	0.538		
8	445	0.04	0.01	-69	74-96	4	0.081		
8	611	0.04	<0.01	-87	74-96	5	0.253		
8	814	0.41	0.04	-89	74-96	7	0.026	-0.084 (-0.153 to-0.015)	8.3
8	884	1.59	0.01	-100	74-/04	8	0.003	-0.169 (-0.257 to-0.081)	4.1
8	914	0.04	0.01	-74	74-96	7	0.068		
9	0	0.02	<0.01	-95	74-90	3	0.036	-0.193 (-0.330 to-0.056)	3.6
9	512	0.11	0.04	-67	74-75	2			
9	620	0.04	0.01	-73	74-96	6	0.813		
9	671	0.04	0.01	-63	74-04	13	0.297		
9	770	0.04	0.12	233	75-/96	5	0.956		
10	0	0.11	0.10	-13	75-96	11	0.043	-0.103 (-0.201 to-0.005)	6.7
10	30	0.04	0.02	-46	74-04	16	0.279		
10	73	0.02	0.02	1	74-96	11	0.7		

Table 5. ANCOVA table for factors affecting concentrations of ^{137}Cs in plants at different distances from the Savannah River along 10 trails in Creek Plantation (n=324). Non-significant interaction terms were excluded from the model.

Source of variation	Sum-of-Squares	df	Mean-Square	F-ratio	P
Trail	2.78	7	0.40	0.15	0.994
Distance	151.11	1	151.11	56.71	<0.001
Distance squared	127.02	1	127.02	47.67	<0.001
Year	79.20	1	79.20	29.72	<0.001
Trail-distance interaction	69.73	7	9.96	3.74	0.001
Trail-distance squared interaction	133.48	7	19.07	7.16	<0.001
Error	796.73	299	2.67		

Figure titles

Figure 1. Map of the Savannah River Site showing the source of contamination to Creek Plantation.

Figure 2. Sampling transects through the floodplain swamp in Creek Plantation.

Figure 3. Relationship between ^{137}Cs activity concentrations in soil and distance from the Savannah River at Creek Plantation

Figure 4. a) Changes in ^{137}Cs activity concentrations over time in soil at the Creek Plantation sampling sites. b) Plot of ^{137}Cs residuals (with sampling location variance removed from $\log_{10}$ transformed soil ^{137}Cs activity concentration) against time. Also shown are regression lines (solid) and 95% confidence intervals (dashed).

Figure 5. Geometric mean ^{137}Cs activity concentrations at different soil depths.

Figure 6. a) Changes in ^{137}Cs activity concentrations over time in vegetation at the Creek Plantation sampling sites. b) Plot of ^{137}Cs residuals (with sampling location variance removed from $\log_{10}$ transformed vegetation ^{137}Cs activity concentration) against time. Also shown are regression lines (solid) and 95% confidence intervals (dashed).

Figure 7. Regression of ^{137}Cs in soil on ^{137}Cs in vegetation from Creek Plantation. Also shown are the regression line (solid) and 95% confidence interval (dashed).

Figure 8. Changes in ^{137}Cs concentration ratios (^{137}Cs in plants/^{137}Cs in soil) over time on the Creek Plantation floodplain. Also shown are the regression line (solid) and 95% confidence interval (dashed).

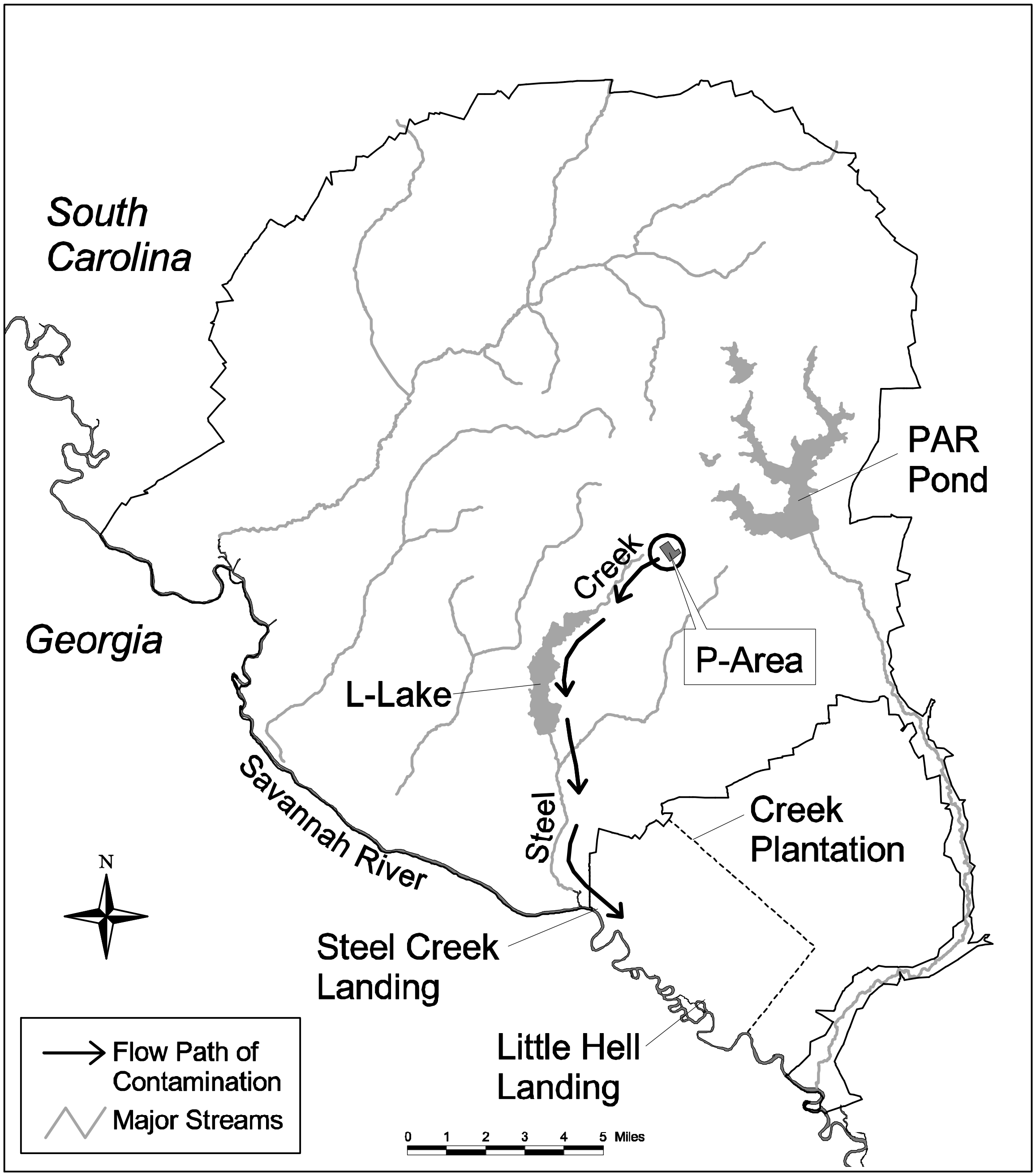
South
Carolina
Georgia
PAR
Pond
Creek
P-Area
L-Lake
Steel
Savannah River
Creek
Plantation
N
Steel Creek
Landing
Little Hell
Landing
Flow Path of
Contamination
Major Streams
0 1 2 3 4 5 Miles

Figure 1.

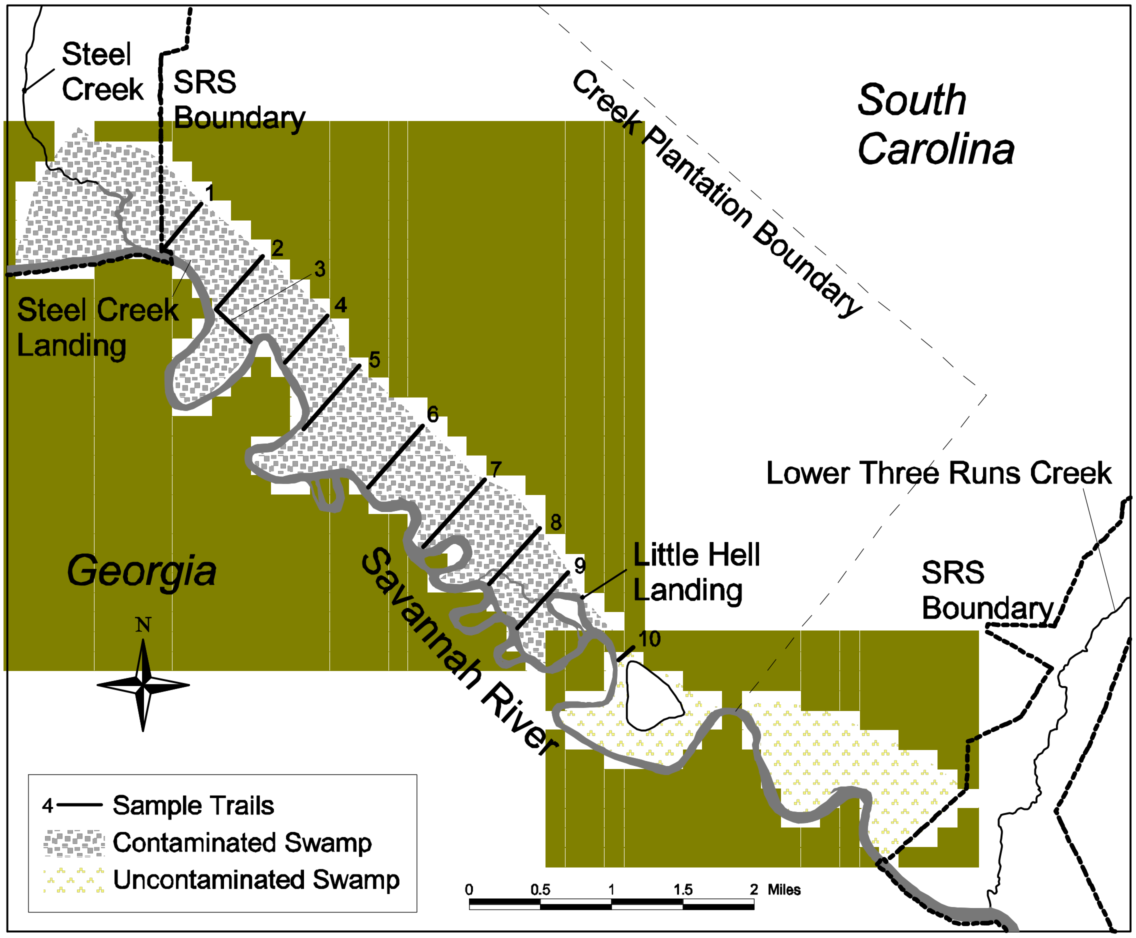
Steel
Creek
SRS
Boundary
Creek Plantation Boundary
South
Carolina
1
2
3
4
5
6
7
8
9
10
Steel Creek
Landing
Lower Three Runs Creek
Little Hell
Landing
SRS
Boundary
Georgia
Savannah River
N
4 Sample Trails
Contaminated Swamp
Uncontaminated Swamp
0
0.5
1
1.5
2 Miles

Figure 2

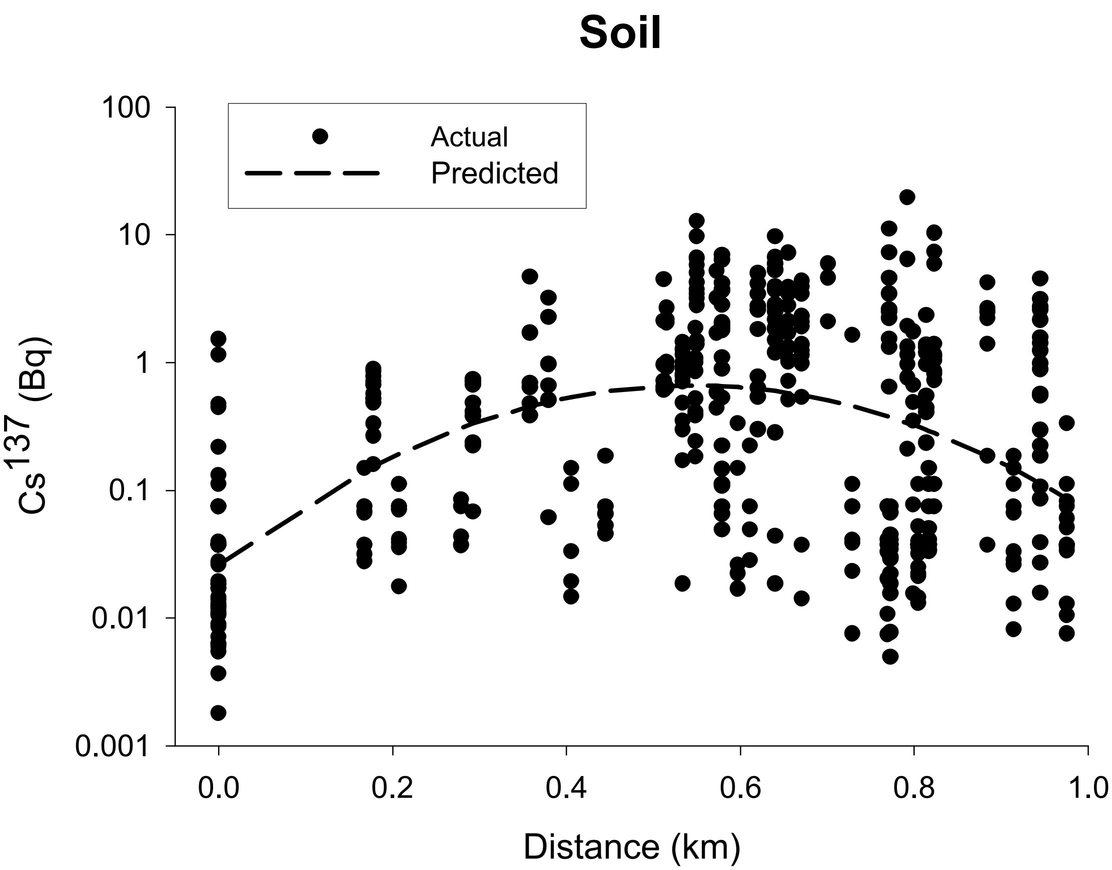
Soil
Actual
Predicted
100
10
1
0.1
0.01
0.001
Cs137 (Bq)
0.0
0.2
0.4
0.6
0.8
1.0
Distance (km)

Figure 3.

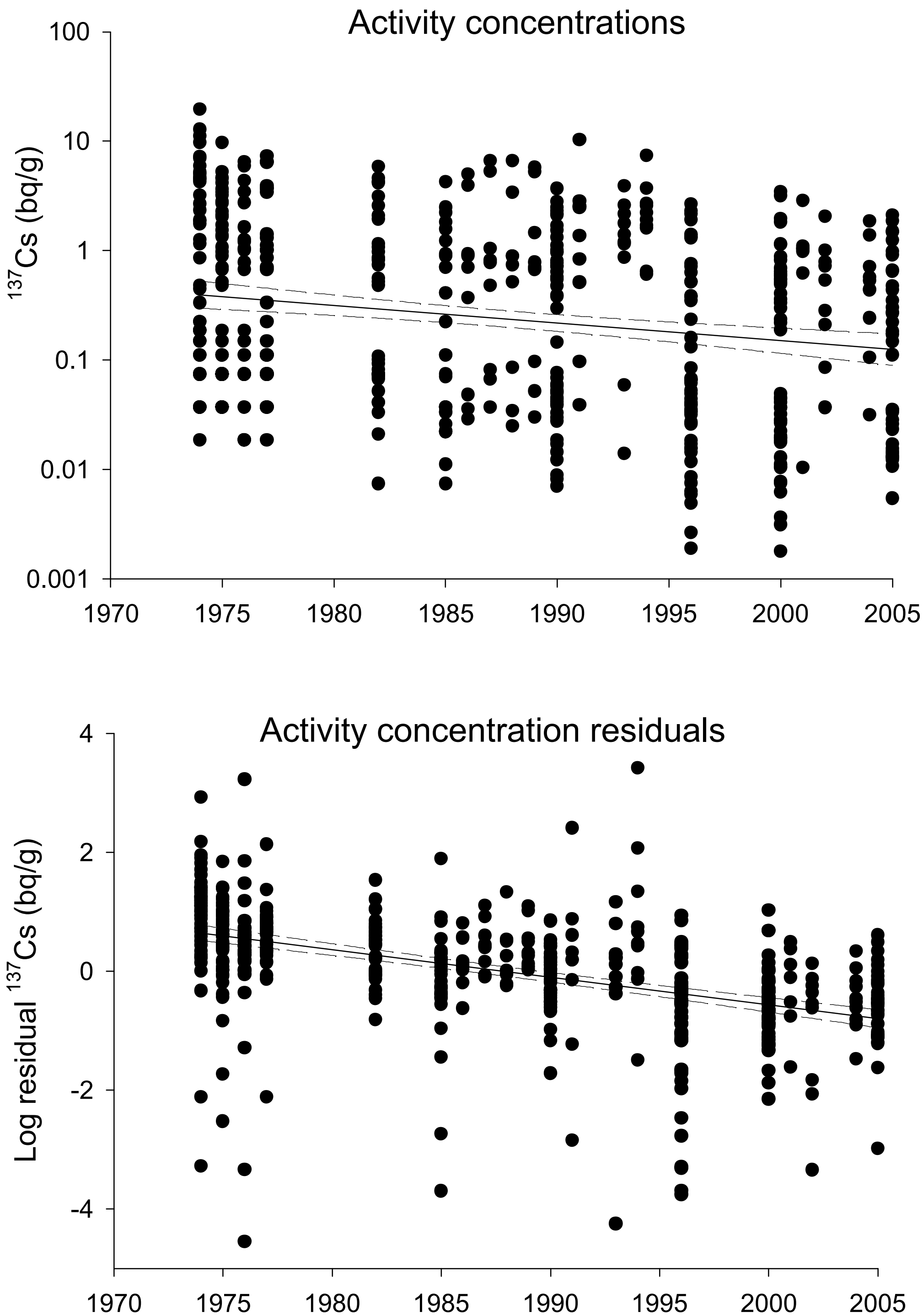
Activity concentrations
^{137}Cs (bq/g)
100
10
1
0.1
0.01
0.001
1970
1975
1980
1985
1990
1995
2000
2005
Activity concentration residuals
Log residual ^{137}Cs (bq/g)
4
2
0
-2
-4
1970
1975
1980
1985
1990
1995
2000
2005

Figure 4.

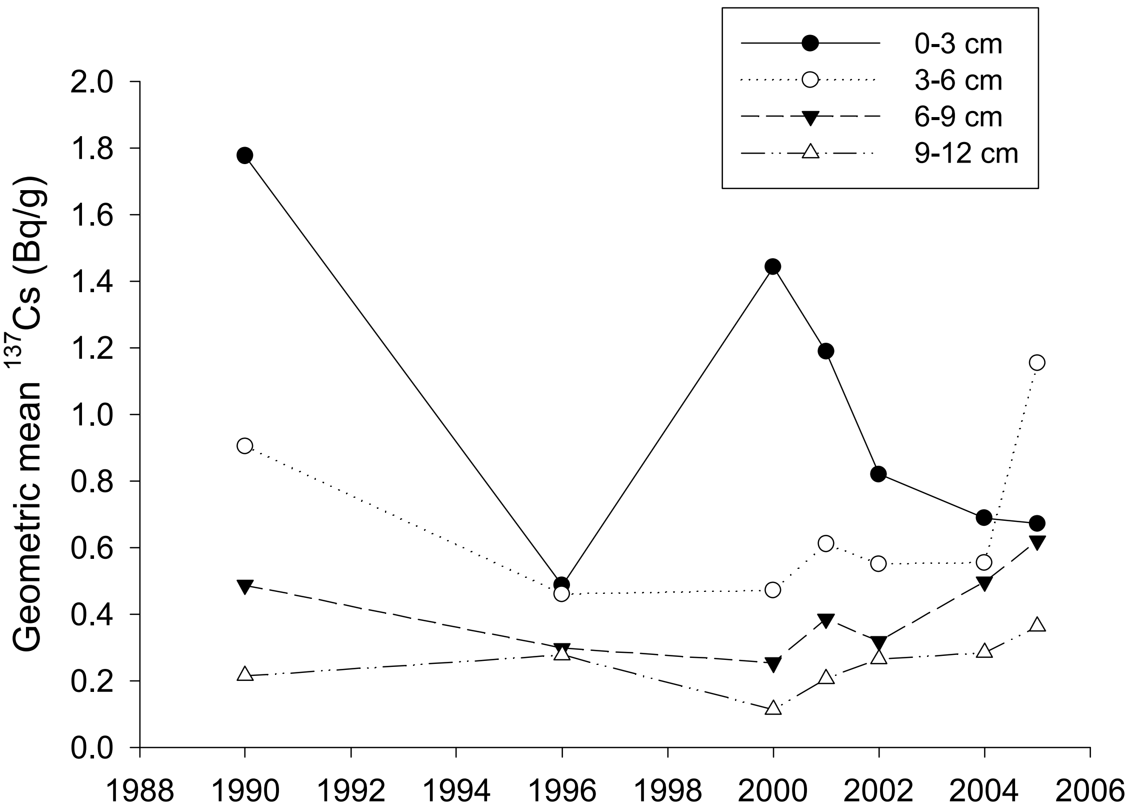
0-3 cm
3-6 cm
6-9 cm
9-12 cm
Geometric mean ^{137}Cs (Bq/g)
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
1988
1990
1992
1994
1996
1998
2000
2002
2004
2006

Figure 5.

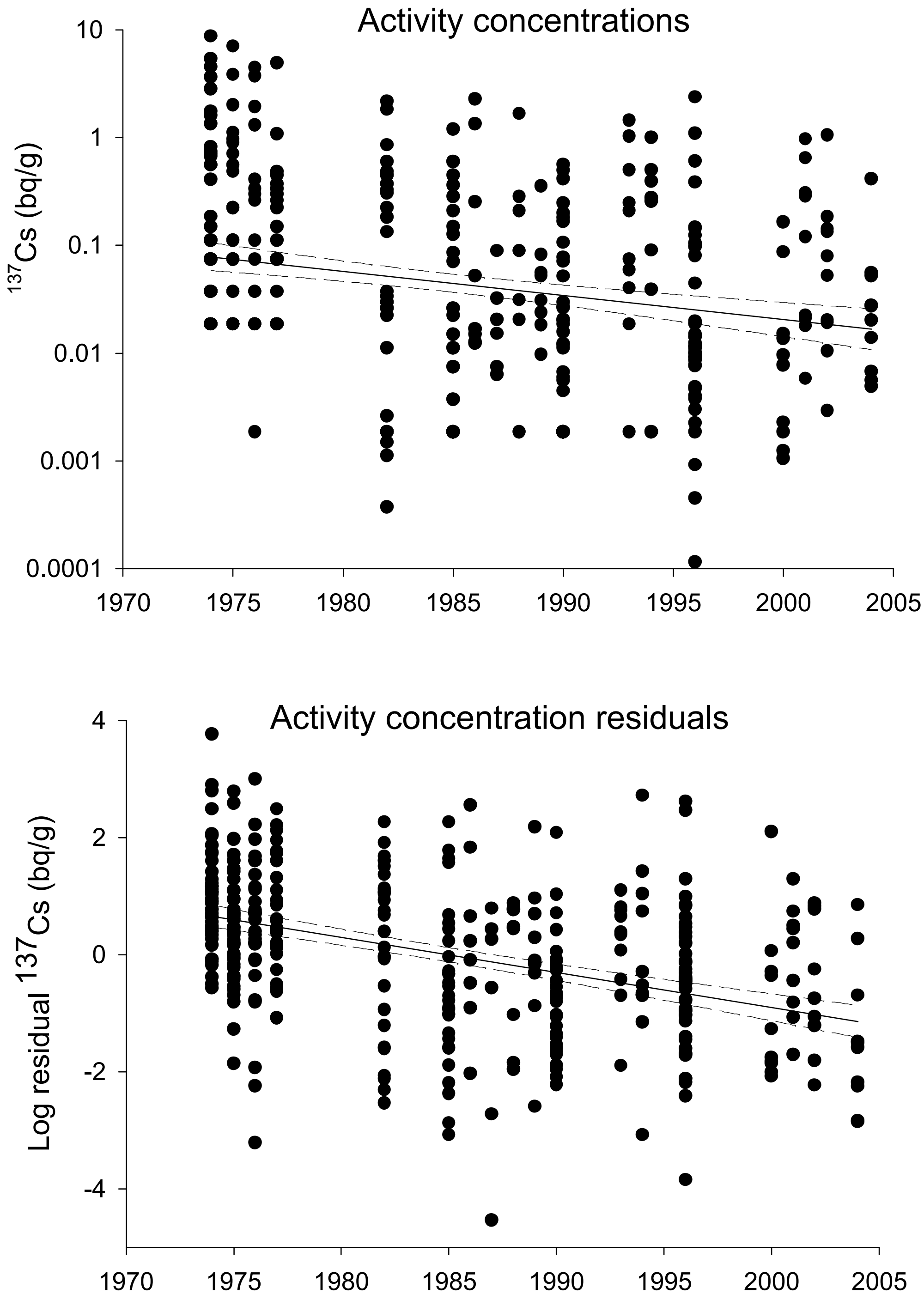
Activity concentrations
10
1
0.1
0.01
0.001
0.0001
137Cs (bq/g)
1970
1975
1980
1985
1990
1995
2000
2005
Activity concentration residuals
4
2
0
-2
-4
Log residual 137Cs (bq/g)
1970
1975
1980
1985
1990
1995
2000
2005

Figure 6.

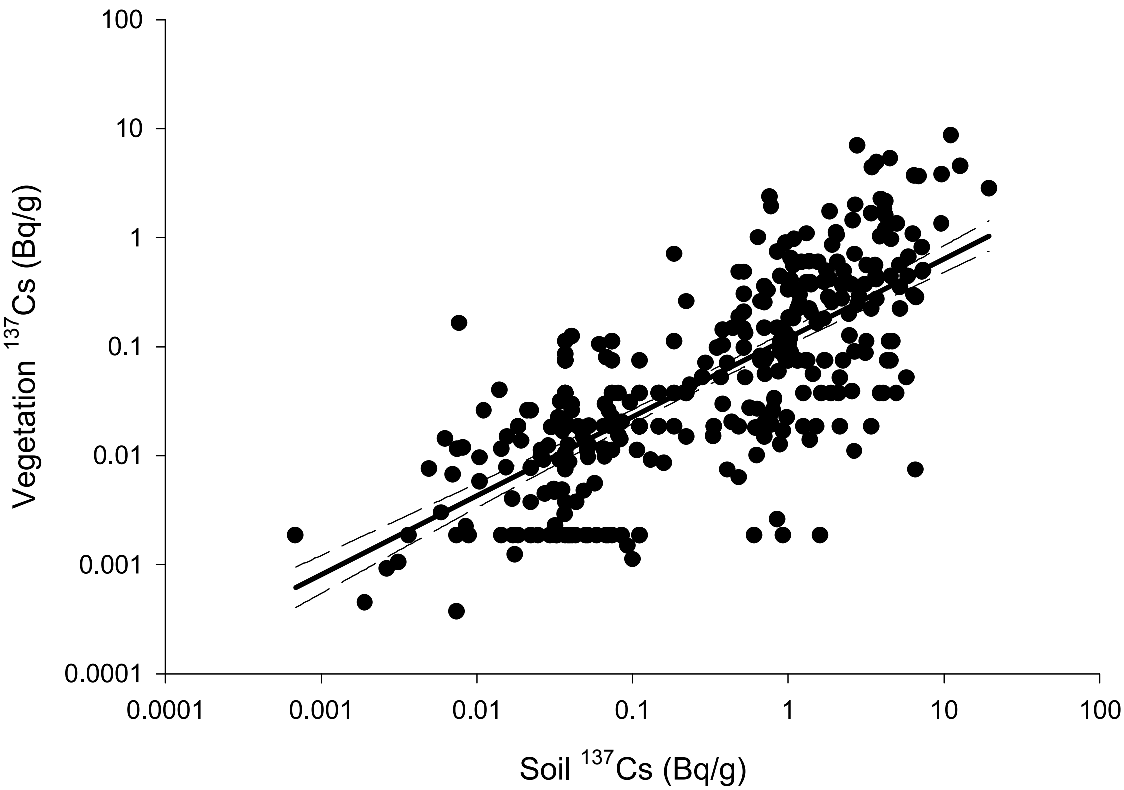
100
10
1
0.1
0.01
0.001
0.0001
Vegetation ^{137}Cs (Bq/g)
0.0001
0.001
0.01
0.1
1
10
100
Soil ^{137}Cs (Bq/g)

Figure 7.

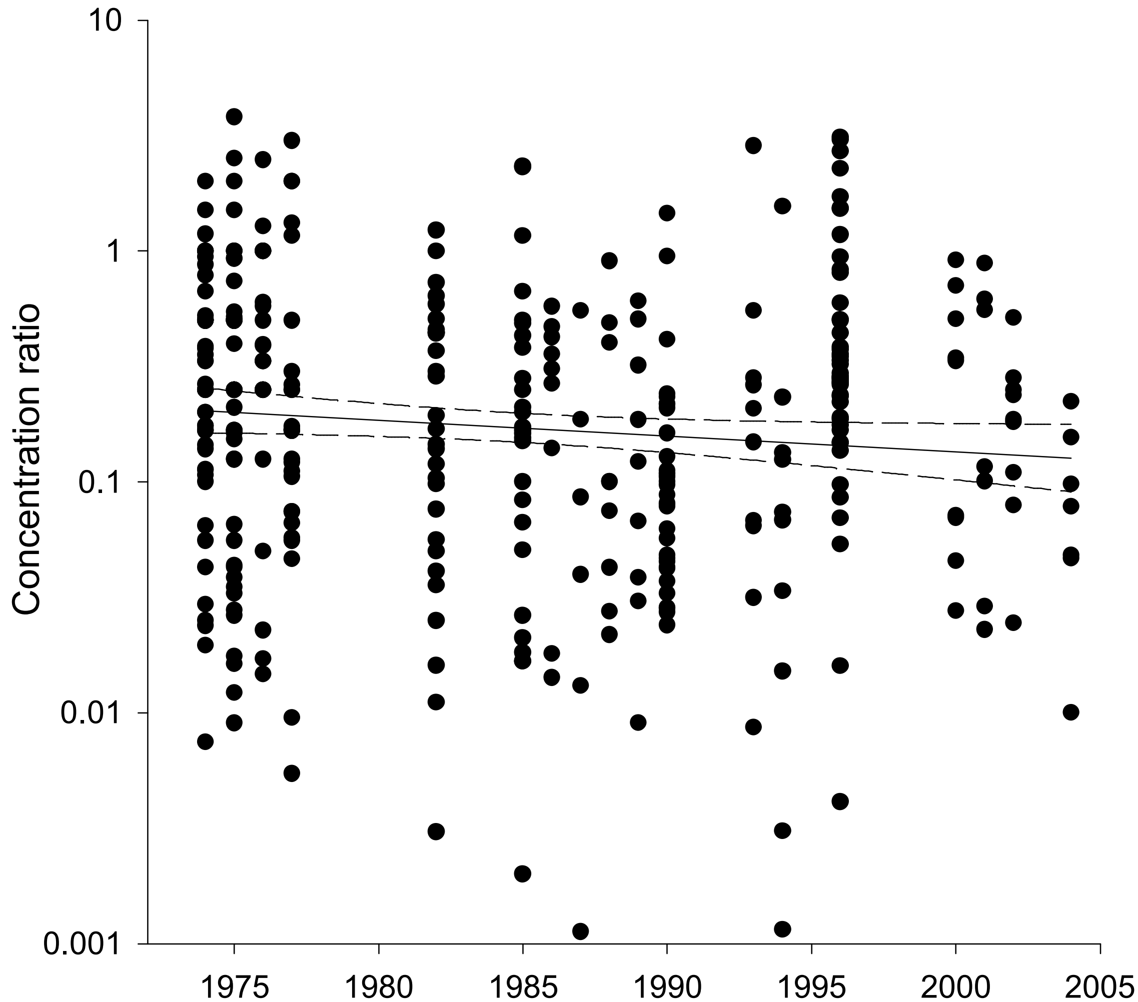
10
1
0.1
0.01
0.001
Concentration ratio
1975
1980
1985
1990
1995
2000
2005

Figure 8.