

Effective half-life of ^{137}Cs in various environmental media at the Savannah River Site

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Abstract

During the operational history of the Savannah River Site (SRS), many different radionuclides have been released from site facilities into the SRS environment. However, only a relatively small number of pathways, most importantly ^{137}Cs in fish and deer, have contributed significantly to doses and risks to the public. The “effective” half-lives (T_e) of ^{137}Cs (which include both physical decay and environmental dispersion) in Savannah River floodplain soil and vegetation and in fish and white-tailed deer from the SRS were estimated using long-term monitoring data. For 1974–2011, the T_e s of ^{137}Cs in Savannah River floodplain soil and vegetation were 17.0 years (95% CI = 14.2–19.9) and 13.4 years (95% CI = 10.8–16.0), respectively. These T_e s were greater than in a previous study that used data collected only through 2005 as a likely result of changes in the flood regime of the Savannah River. Field analyses of ^{137}Cs concentrations in deer collected during yearly controlled hunts at the SRS indicated an overall T_e of 15.9 years (95% CI = 12.3–19.6) for 1965–2011; however, the T_e for 1990–2011 was significantly shorter (11.8 years, 95% CI = 4.8–18.8) due to an increase in the rate of ^{137}Cs removal. The shortest T_e s were for fish in SRS streams and the Savannah River (3.5–9.0 years), where dilution and dispersal resulted in rapid ^{137}Cs removal. Long-term data show that T_e s are significantly shorter than the physical half-life of ^{137}Cs in the SRS environment but that they can change over time. Therefore, it is desirable have a long period of record for calculating T_e s and risky to extrapolate T_e s beyond this period unless the processes governing ^{137}Cs removal are clearly understood.

Keywords

- Cesium;
- Effective half-life;
- Ecological half-life
- Long-term change

1. Introduction

^{137}Cs has a physical half-life of 30.2 years, a high fission yield, and a high bioavailability due to its physiological similarity to potassium. The latter factor results in its movement through aquatic and terrestrial food chains where it concentrates in soft tissues such as edible skeletal muscle and bioaccumulates in higher trophic level consumers, including humans. At the Savannah River Site (SRS), a US Department of Energy nuclear materials production site in South Carolina, dose and risk assessments indicate that ^{137}Cs in deer, fish, soil, and vegetation has been, and will continue to be, the critical radionuclide/pathway with the highest potential human exposure ([Jannik and Scheffler, 2011](#)).

Research shows that the rate of ^{137}Cs elimination in the environment is often different from the physical decay rate of ^{137}Cs because of a variety of factors that remove ^{137}Cs from ecosystems, sequester it, or reduce its biological availability ([Whicker and Schultz, 1982](#)). The “effective half-life” or T_e of a radionuclide includes the physical decay half-life and the ecological (environmental dispersion/dilution) half-life ([Whicker and Schultz, 1982](#)). It is the time required for the radionuclide to decrease by 50% in the environment as a result of physical (e.g. radioactive decay, sedimentation, and washout), chemical (e.g. change in oxidation state and adsorption), and biological (e.g. changes in food web and biota translocations) factors. The T_e is determined by the equation $T_e = \log_e 2 / \lambda_e$, where λ_e is estimated from the slopes of \log_e -transformed ^{137}Cs activity concentration regressions.

Knowledge of the environmental fate of radionuclides is important because it contributes to the projection of long-term risks resulting from radionuclide releases and the selection of cost-effective remediation strategies. There has been substantial research on the fate of ^{137}Cs and other radionuclides released by nuclear weapons testing, the Chernobyl accident, and other incidents in temperate zone ecosystems of Europe ([Pröhl et al., 2006](#)). Radionuclide fate in the warm temperate climate of the SRS has been studied by [Pinder et al., 1980](#), [Gladden et al., 1985](#), [Whicker et al., 1990](#) and [Hinton et al., 1999](#) and others, although most of these studies did not concern long-term rates of ^{137}Cs elimination from the environment. More recently, [Paller et al. \(2008\)](#) analyzed long-term (1974–2005) trends in ^{137}Cs concentrations in Savannah River floodplain soil and vegetation. They showed that the T_e s in this soil and vegetation were 14.9 years (95% CI = 12.5–17.3) and 11.6 years (95% CI = 9.1–14.1), respectively. T_e s of ^{137}Cs have also been calculated for fish from various aquatic ecosystems on the SRS ([Paller et al., 1999](#) and [Paller et al., 2002](#)). The objectives of the current study are to 1) extend the evaluations of Savannah River floodplain soils and vegetation and fish through 2011 to determine if the T_e s remained stable and 2) add evaluations of the T_e of ^{137}Cs in onsite SRS deer to provide a more complete assessment of the fate of ^{137}Cs in SRS ecosystems.

2. Materials and methods

2.1. Study area

The SRS is an 800 km² reservation established in 1951 for the production of nuclear material. It is located on the upper coastal plain of South Carolina near Aiken, South Carolina (USA) ([Fig. 1](#)). By 1955 there were five functioning nuclear reactors on the SRS (all of which have since been deactivated) as well as various facilities for processing nuclear materials. Throughout the past 60 years, measurable releases of over 50 radionuclides and various non-radiological contaminants into the atmosphere, onsite streams, and seepage basins have occurred as a result of operations at SRS. However, only a relatively small number of the released radionuclides have been significant contributors to doses and risks to the public ([Jannik and Scheffler, 2011](#)).



Fig. 1. Location of the Savannah River Site in the southeastern United States.

2.2. ¹³⁷Cs in the SRS environment

Sources of residual radioactivity on the SRS include global fallout and releases associated with SRS operations. Global fallout of fission products from above ground weapons testing is a major source of residual radioactivity at the SRS. A worldwide total of about 480 megatons of nuclear weapons were detonated above ground between 1945 and 1980 introducing a total of about 1.3×10^6 TBq into the atmosphere ([Eisenbud, 1987](#)). The deposition density of ¹³⁷Cs in the southeastern US (including the SRS area) ranged between 4000 and 6000 Bq/m², with some localized areas receiving even greater amounts ([CDC/NCI, 2002](#)) ([Fig. 2](#)). Because of anthropogenic activities such as agriculture and suburban and urban developments, much of this ¹³⁷Cs has been dispersed and is no longer bioavailable. However, at the SRS, less than 10% of the site has been impacted by industrial activities and the rest is forested land that has been managed by the US Forest Service since the early 1950's. Because of this protection, a much larger fraction of the ¹³⁷Cs deposited on the SRS during the 1950's and early 1960's remains bioavailable to higher trophic level animals, such as deer. This phenomenon has also been reported from other large protected land areas such as military bases and National/State Forests ([Gaines and Novak, 2011](#)).

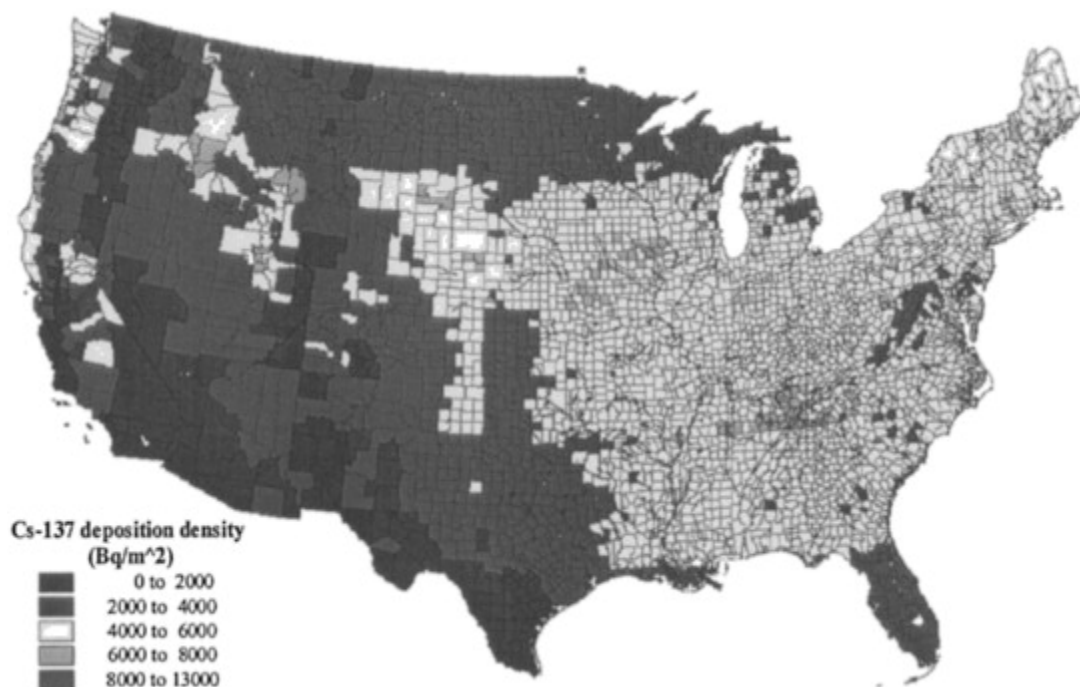


Fig. 2.

¹³⁷Cs deposition density (Bq/m²) in the United States ([CDC/NCI, 2002](#)).

SRS operations have resulted in the release of ¹³⁷Cs to the air and to site streams and other water bodies ([Jannik and Scheffler, 2011](#)) (Figs. 3 and 4). During 1995–2010, the total measured atmospheric release of ¹³⁷Cs from the SRS was about 0.12 TBq. Most of this was released in 1955 and 1987. Operational problems at four of five onsite reactors resulted in aqueous releases that contaminated several streams on the SRS and their contiguous floodplains ([Paller et al., 1999](#)). These releases were associated with the withdrawal of reactor cooling water from the Savannah River, which was subsequently discharged into streams and cooling reservoirs that returned the water to the Savannah River. The total liquid release of ¹³⁷Cs to SRS streams was about 21 TBq. An additional 50 TBq was discharged to onsite seepage basins. Residual contamination from the early aqueous releases of ¹³⁷Cs remains in SRS stream sediments and floodplain soils. Overflight data showed that three onsite streams (Fourmile Branch, Steel Creek, and Lower Three Runs) and their floodplains still had residual ¹³⁷Cs contamination in their sediments/soils more than 40 years after the releases.

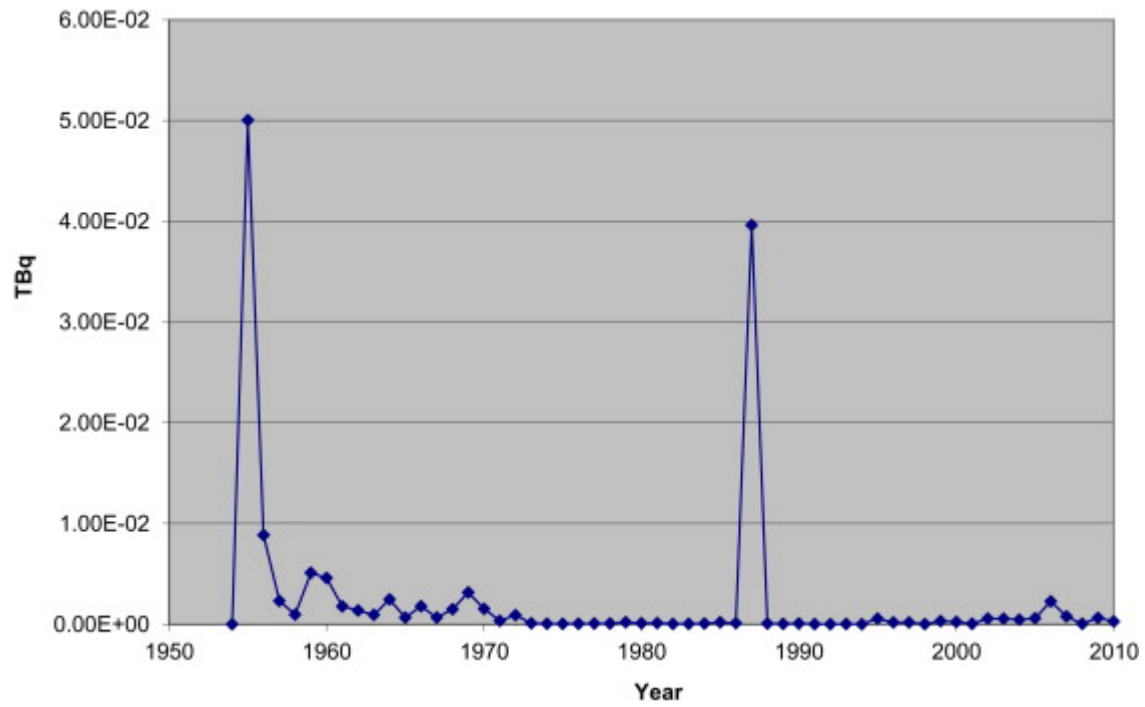


Fig. 3.

Airborne releases of ^{137}Cs from the Savannah River Site ([Jannik and Scheffler, 2011](#)).

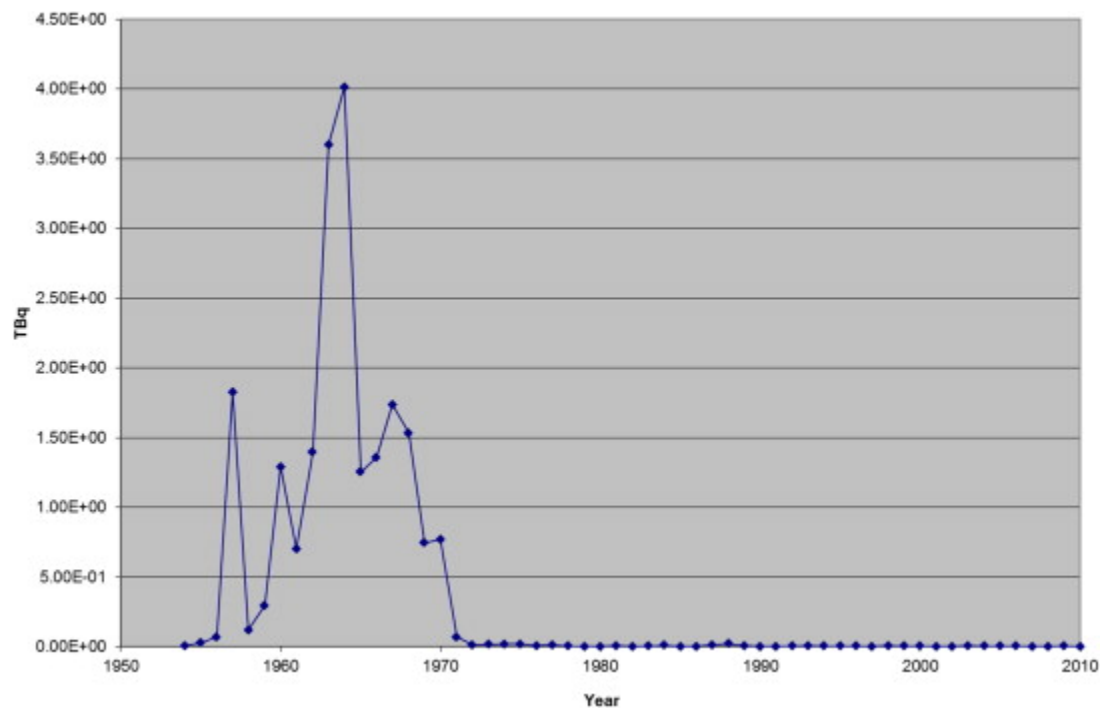


Fig. 4.

Liquid releases of ^{137}Cs from the Savannah River Site ([Jannik and Scheffler, 2011](#)).

Steel Creek, a Savannah River tributary, received approximately 10.35 TBq of ^{137}Cs during 1954–1974 ([Fledderman et al., 2007](#)). Water from Steel Creek backed up and overflowed into the Savannah River floodplain swamp when Savannah River levels were high resulting in the deposition of radioactive materials on a privately owned, uninhabited portion of the Savannah River floodplain known as Creek Plantation. This area subsequently became the focus of a monitoring program that documented levels of ^{137}Cs in floodplain soils and vegetation beginning in 1974 and continuing through the present. More information about the Creek Plantation environment can be found in [Fledderman et al. \(2007\)](#) and [Paller et al. \(2008\)](#).

2.3. Field and laboratory methods

Data presented herein are from four environmental media that have been sampled as part of the SRS environmental monitoring program: floodplain soils, floodplain vegetation, fish tissue, and white-tail deer (*Odocoileus virginianus*) tissue.

Floodplain soil and vegetation data were obtained from Creek Plantation, where 10 trails were established through the swamp in the early 1970s for sample collection. With the exception of Trail 10, which was 60 m long, the trails were 626–975 m long and had five to eight fixed sampling locations extending from near the Savannah River channel to the upper boundary of the swamp. Shallow (to a depth of about 7.6 cm) soil samples were collected from each sampling location as were samples of living, low-growing herbaceous vegetation. Soil and vegetation samples were dried for radiological analysis; processed samples were counted on an HPGe detector of approximately 35% efficiency. Sampling was initiated in 1974 and efforts were made to sample most sites every few years. [Fledderman et al. \(2007\)](#) and [Paller et al. \(2008\)](#) can be consulted for more details concerning sampling methods, sampling design, and sample sizes.

Fish have been collected from a number of sites on and near the SRS including the Savannah River upstream and downstream from the SRS and several Savannah River tributary streams that received reactor cooling water. Most sites were sampled annually or every few years. Mean ^{137}Cs activity concentrations in fish from each site were documented in annual reports. Several types of fish were collected including largemouth bass (*Micropterus salmoides*), sunfishes (*Lepomis* spp.), and bullheads (*Ameiurus* spp.); however, we averaged all species to provide an overview of temporal trends. Results for individual taxonomic groups and more information on analytical and statistical procedures are presented in [Paller et al. \(1999\)](#). Samples collected after 1989 were counted with a shielded high purity germanium detector while earlier samples were counted with NaI(Tl) solid scintillator or Ge(Li) semiconductor detectors.

Since 1965, annual game animal hunts, open to the general public, have been conducted at the SRS to control deer and feral hog populations. SRS personnel use portable sodium iodide detectors to perform field analyses for ^{137}Cs prior to the release of any animals to hunters. The estimated dose from the consumption of harvested deer or hog meat is determined from the field measured ^{137}Cs concentrations to ensure that the hunter's dose does not exceed the SRS administrative game limit of 0.22 millisievert (22 mrem). The maximum annual dose from the onsite-hunter deer/hog consumption pathway typically exceeds all standard maximally exposed individual pathways combined and all other SRS sportsman dose scenarios ([Jannik and Scheffler, 2011](#)).

2.4. Data analysis

Effective loss rate constants (λ_e) for each environmental medium were estimated from the slopes of \log_e -transformed ^{137}Cs activity concentrations regressed on year. These values 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 (Bq/g^{-1}). The effective half-life was estimated as $T_e = \log_e 2 / \lambda_e$. Computing effective half-lives for ^{137}Cs in this manner will produce accurate results only if additional ^{137}Cs inputs are minimal or null. Therefore, T_e estimates for fish were restricted to after 1971 when ^{137}Cs releases from SRS reactors were negligible. Contamination of the Creek Plantation floodplain occurred primarily before sampling 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. Such inputs were likely small but may represent a potential positive bias in the effective half-life computations. Corrections were not made for ^{137}Cs inputs from global fallout. In the aquatic ecosystems under study this source was insignificant compared with contamination from the SRS (Paller et al., 1999).

An objective of this study was to determine if loss rate constants remained stable or if they decreased or increased with time. Stability would be indicated by a consistent linear decrease in \log_e -transformed ^{137}Cs activity concentrations and change by an upward or downward deflection of the ^{137}Cs trend line; i.e., a curvilinear trend. Curvilinear trends were assessed by including second degree (quadratic) terms in polynomial regressions models relating ^{137}Cs concentration and time and testing them for statistical significance ($P \leq 0.05$).

Statistical models for Creek Plantation soil and vegetation included terms for sample site location so that this source of variation could be separated from variation in ^{137}Cs activity concentrations that were associated with time. T_e s for soil and vegetation from Creek Plantation were calculated from model residuals that represented ^{137}Cs activity concentrations with location related variance removed. This step, which was also performed in previous analyses, was needed to control potential biases associated with nonrandom spatial sampling during some years (Paller et al. (2008).

3. Results

3.1. Effective half-life (T_e) for ^{137}Cs in Savannah River floodplain soil and vegetation

Statistical models for ^{137}Cs activity concentrations in Savannah River floodplain soil and vegetation were significant ($P < 0.001$) but characterized by low coefficients of determination (R^2 s of 0.22 and 0.19, respectively). This was likely because of small-scale spatial variation in ^{137}Cs concentration (see Paller et al., 2008). Previous analysis of data collected from 1974 to 2005 indicated T_e s of 14.9 years (95% CI = 12.5–17.3) and 11.6 years (95% CI = 9.1–14.1), respectively (Paller et al., 2008). When floodplain soil and vegetation data from the 10 sites sampled from 2006 through 2011 were added to the analysis, the T_e for ^{137}Cs in soil increased to 17.0 years (95% CI = 14.2–19.9), and the T_e for ^{137}Cs in vegetation increased to 13.4 years (95% CI = 10.8–16.0) (Fig. 5 and Fig. 6). These increases suggest that rates of ^{137}Cs elimination decreased over time. Decreases could be manifested as a gradual slope decline that resulted in a curvilinear trend. This possibility was tested by fitting the ^{137}Cs soil data to a polynomial quadratic model. The model produced only a trivial increase in R^2 (from 0.223 to 0.228), and the quadratic term was not significant at $P \leq 0.05$. Other models (e.g., exponential decay) were also tested; however, none fit the data better than a linear model. Similar results were obtained with the floodplain vegetation data.

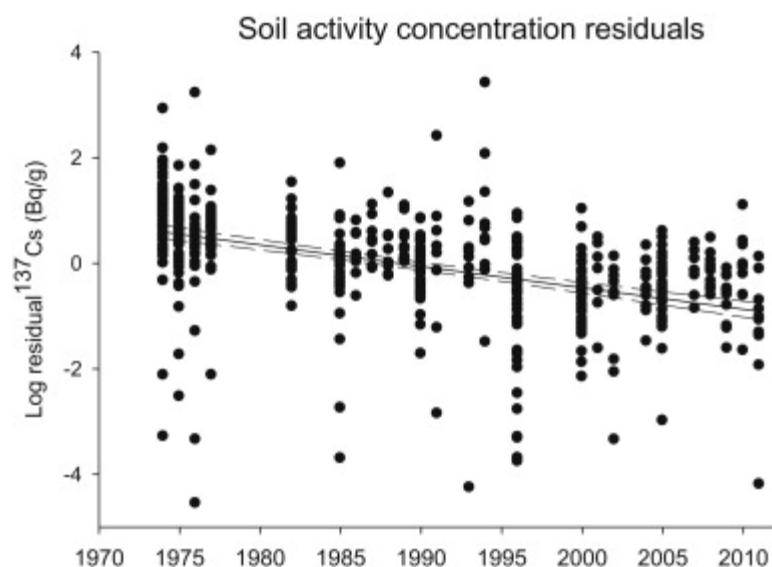


Fig. 5.

Changes in ^{137}Cs in Savannah River floodplain soil during 1974–2011. T_e for ^{137}Cs = 17.0 (95% CI = 14.2–19.9) years. Plotted data are ^{137}Cs general linear model residuals with sampling location variance removed from \log_{10} transformed activity concentrations. Also shown are regression lines (solid) and 95% confidence intervals (dashed).

Another possibility is that changes in the rate of ^{137}Cs decline were sudden rather than gradual resulting in an inflection point (or points) rather than a systematic trend. Plotting ^{137}Cs activity concentrations in soil against time suggested intermittent periods of decline and stability but was complicated by the wide dispersion of the individual data points (Fig. 5). Plotting the yearly arithmetic means of the individual points eliminated this problem and revealed that ^{137}Cs activity concentrations showed little consistent change from 1974 to 1995, then declined abruptly, and subsequently entered a second period from 1996 to 2011 characterized by relatively low levels and little consistent change (Fig. 7). This second period followed two years of extremely high Savannah River discharge (1993 and 1998) in which the entire Creek Plantation floodplain was inundated for several months each spring (Fig. 7). Statistical testing confirmed that significant ($P \leq 0.05$) linear trends were lacking when the 1974–1995 and the 1996–2011 periods were analyzed separately. Therefore, inclusion of the new (2006–2011) data in the T_e calculations reduced the overall T_e for floodplain soils because these data represented a period of little ^{137}Cs decline. Examination of annual means for the vegetation data (not shown) indicated a more regular decline and an absence of obvious temporal inflection points as observed in the soil data.

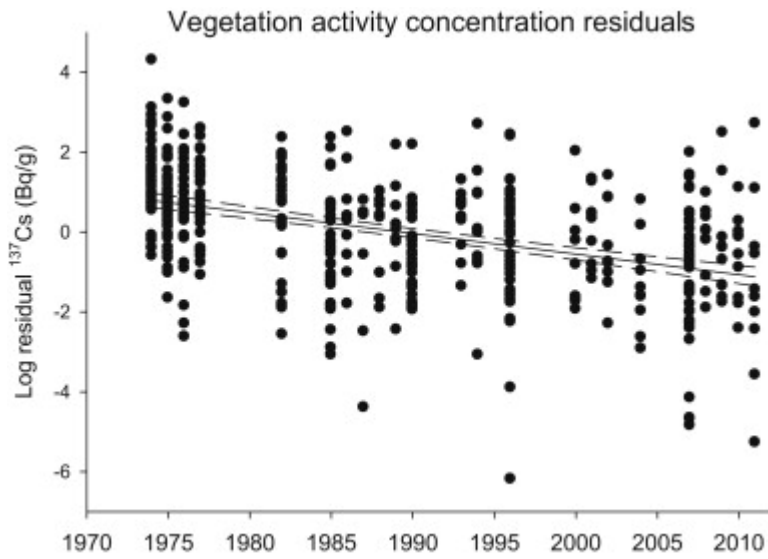


Fig. 6.

Changes in ^{137}Cs in Savannah River floodplain vegetation during 1974-2011 T_e for ^{137}Cs = 13.4 (95% CI = 10.8-16.0) years. Plotted data are ^{137}Cs general linear model residuals with sampling location variance removed from \log_{10} transformed activity concentrations. Also shown are regression lines (solid) and 95% confidence intervals (dashed).

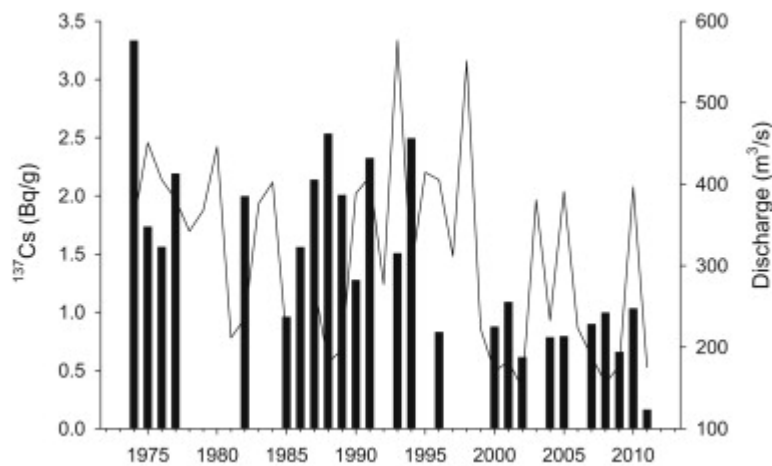


Fig. 7.

Yearly arithmetic mean ^{137}Cs concentrations in Savannah River floodplain soils and yearly arithmetic mean Savannah River discharge near Augusta, Georgia.

3.2. Effective half-life (T_e) for ^{137}Cs in SRS white-tailed deer

From 1965 to 2011, the natural log of the mean ^{137}Cs concentration measured in white-tailed deer at the SRS ranged from about 0.67 Bq/g to about 0.037 Bq/g. During the past five years, the range has been from 0.093 Bq/g to 0.044 Bq/g. Linear regression of these data produced a T_e of 15.9 years (95% CI = 12.3–19.6); however examination of the residuals from this regression suggested the possibility that the trend line was curvilinear rather than linear. A significant quadratic term in a polynomial regression model confirmed this observation and indicated that the log transformed ^{137}Cs loss rate in deer has declined more rapidly in recent years (Fig. 8). Thus, it may be more appropriate to divide the deer data into two sets: old (1965–1989) and new (1990–2011) and calculate individual T_e s for each. Linear regression of the old data failed to produce a statistically significant ($P \leq 0.05$) loss term. Analysis of the new data resulted in a T_e of 11.8 years (95% CI = 4.8–18.8).

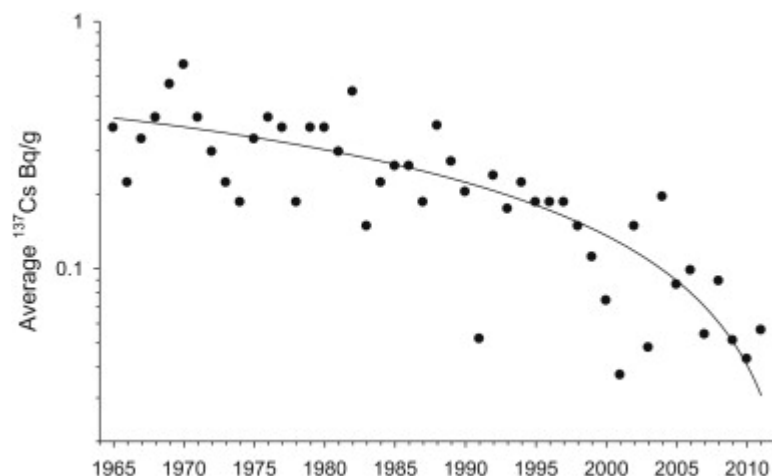


Fig. 8.

Yearly arithmetic mean ^{137}Cs activity concentrations measured in deer collected during SRS hunts from 1965–2011. Also shown is the quadratic polynomial regression line.

3.3. Effective half-life (T_e) for ^{137}Cs in SRS fish

The Augusta Lock and Dam is located on the Savannah River more than 25 river-miles upstream of the SRS. The maximum natural log of the mean ^{137}Cs activity concentration in fish composites from this background location was 0.078 Bq/g, which occurred in 1971 (Fig. 9). This value largely reflects the effects of global fallout of ^{137}Cs (although small numbers of fish from near the SRS may move to this area, Paller et al., 2005). Mean values over the last five years have generally been near 0.001 Bq/g. Regression of these data produced a T_e of 7.43 years, much shorter than the effective half-life of ^{137}Cs in soil and vegetation.

The US Highway 301 Bridge over the Savannah River is about 10 river-miles downstream of the SRS. ^{137}Cs activity concentrations at this site over the last five years have generally been around 0.001 Bq/g. Regression analysis for this site included only 1972–2011 data because aqueous releases of ^{137}Cs were still occurring before this (Fig. 4). Linear regression of these data produced a T_e of 8.1 years (95% CI = 5.7–9.6) (Fig. 9). This is about the same as the upriver background location and indicates that the potential dose from consumption of offsite fish downriver of SRS has declined more rapidly than expected on the basis of radioactive decay alone.

Fish samples have also been collected through 2011 from other locations on the SRS, including Steel Creek, Fourmile Creek, and Lower Three Runs (Fig. 9). T_{res} for these sites were 3.6 years (95% CI = 3.0–4.2), 3.8 years (95% CI = 3.4–4.2), and 4.9 years (95% CI = 3.7–4.2), respectively. ^{137}Cs loss rates from these sites were consistent and rapid as shown by strong linear trends (R^2 values of 0.69–0.92) and relatively little dispersion of individual data points around the regression lines.

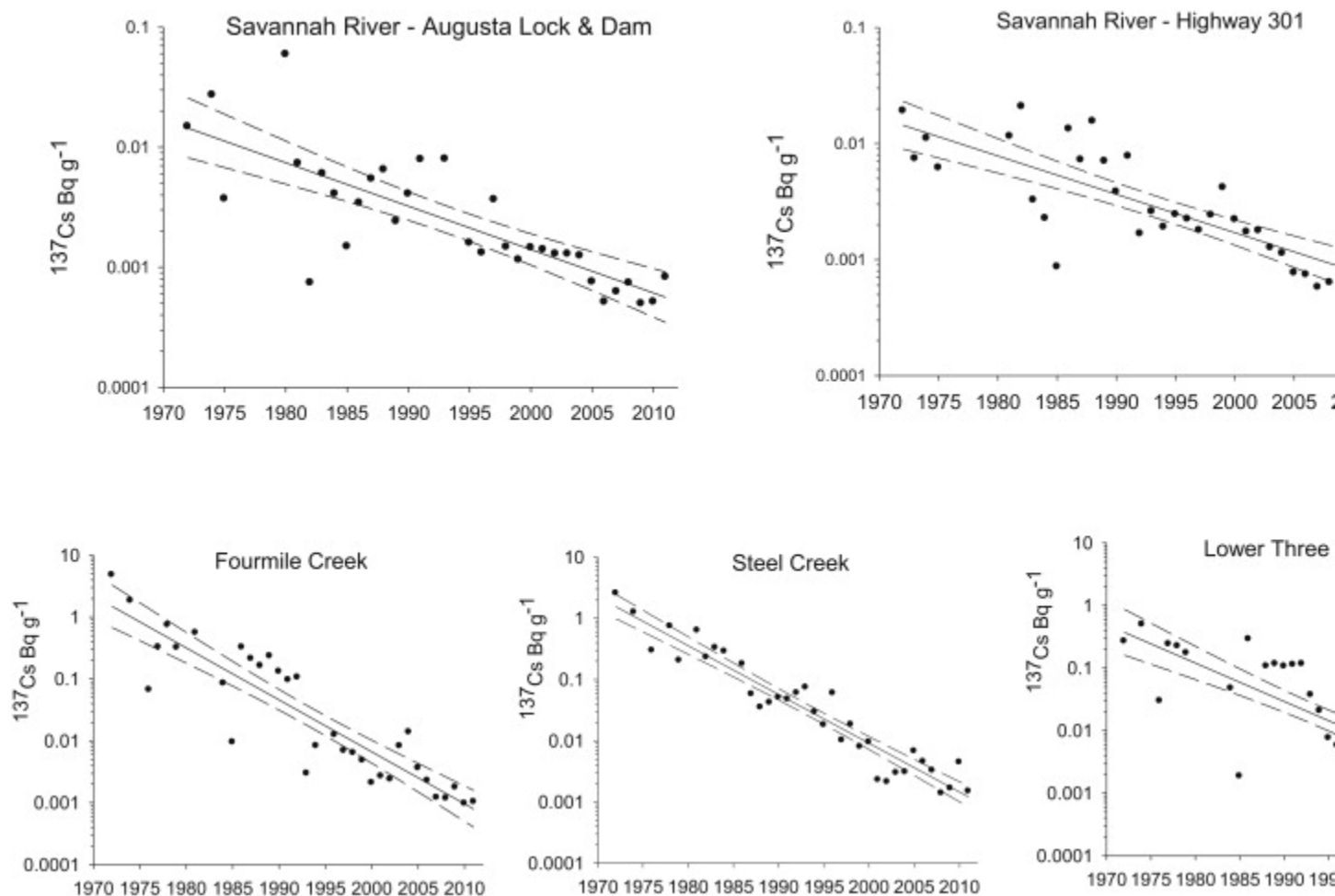


Fig. 9.

Changes in arithmetic mean ^{137}Cs in whole fish over time in the Savannah River upstream (Augusta Lock & Dam) and downstream (Highway 301) from the Savannah River Site (SRS) and in three SRS streams. Solid lines represent regression lines, and dotted lines represent 95% prediction intervals.

4. Discussion

^{137}Cs removal varied in rate and consistency among SRS environmental media as a result of the action of different environmental processes that contributed to the dispersion and sequestration of ^{137}Cs . ^{137}Cs activity concentrations in floodplain surface (0–7.6 cm) soils appeared to decline rapidly when inundated by Savannah River floodwaters but change little at other times. ^{137}Cs may have been mobilized from the floodplain soil into the floodwater when the latter became anoxic, a likely possibility in relatively stagnant backwaters given the large amount of organic matter in the swamp and the duration of flooding (months). Remobilization of ^{137}Cs into anoxic waters may result from the displacement of ^{137}Cs by NH_4^+ ions (Evans et al. 1983). The Savannah River Swamp is seldom flooded during dry periods (e.g., 2006–2011), which eliminates a major factor in the dispersion and dilution of ^{137}Cs at these times. In any case, ^{137}Cs *Tes* for the floodplain soils appeared to be time-scale dependent, with the overall *Te* of 17.0 years for the 1974–2011 study period representing an average that deviates from the *Tes* that could be obtained by dividing the study period into smaller blocks.

The rate of removal of ^{137}Cs from the floodplain herbaceous vegetation was somewhat more rapid than the overall rate of removal from the floodplain soil and lacked the period of abrupt floodwater associated decline observed with the latter. ^{137}Cs availability to the floodplain vegetation was likely determined by both the amount of ^{137}Cs in the root zone and its bioavailability, the latter being influenced by the chemical speciation of the ^{137}Cs . Earlier research showed that ^{137}Cs concentration ratios (i.e., ^{137}Cs dry mass in plants/ ^{137}Cs dry mass in soil) declined slowly over time suggesting a progressive decrease in ^{137}Cs bioavailability (Paller et al., 2008). Similarly, sequential extraction studies indicate that much of the ^{137}Cs in aged SRS floodplain soils is present in mineralogical forms that are likely unavailable to plants (Knox et al., 2001). Therefore, ^{137}Cs removal from the floodplain plants may have differed from shallow floodplain soils because it was governed by changes in the speciation of ^{137}Cs as well as by changes in its quantity.

^{137}Cs removal from SRS white-tailed deer is likely the summation of several complex processes that may include spatial heterogeneity in the distribution of atmospherically deposited ^{137}Cs , differences in the uptake of ^{137}Cs by different types of deer forage plants (and changes in the availability of these plants), temporal changes in the use of forage areas by deer (e.g., uplands versus riparian areas), changes in deer demography (e.g., changes in age distribution), and changes in the locations from which deer were harvested. Because of these unknowns, it is difficult to speculate on the reasons for the observed decrease in the ^{137}Cs *Te* for SRS deer over time, a phenomenon not observed in other SRS environmental media.

^{137}Cs removal rates from SRS stream fish were more consistent over time and far more rapid than from other SRS environmental media. This likely reflected the activity of strongly acting dispersal and sequestration processes including dilution, erosion and transport of contaminated sediments from areas of scouring, and burial of contaminated sediments in areas of deposition. Unlike floodplain soils, *Tes* for fish from SRS streams were relatively time-scale independent (i.e., the removal rates were fairly constant), probably because the processes that governed ^{137}Cs removal from SRS streams were relatively consistent compared with the processes that governed the removal of ^{137}Cs from the surface floodplain soils. Additionally, unlike floodplain soils, where ^{137}Cs can be redistributed vertically and recycled to the soil surface by various processes (Paller et al., 2008), much of the ^{137}Cs in SRS streams was permanently removed by discharge into the Savannah River.

The ^{137}Cs removal rates reported herein can be compared with removal rates reported by other studies from the SRS and from other geographic areas. *Tes* for ^{137}Cs in fish from Par Pond, a reservoir on the SRS with a high water turnover due to its use for once-through reactor cooling, were generally comparable to those from SRS streams ([Table 1](#)). In contrast, Pond B, a comparatively stagnant reservoir without cooling water throughput, was characterized by substantially longer *Tes* and, in the case of Pond B bullheads (*Ameiurus* spp), a *Te* that exceeded the physical half-life of ^{137}Cs . The long *Te* for bullheads, a benthic feeding fish, may reflect the recycling of ^{137}Cs from the sediments by aquatic plants or other processes. *Tes* for marine fish collected at the Bikini and Enewatak Atolls ([Noshkin et al., 1997](#)) after nuclear testing are slightly longer than for SRS streams and Par Pond but still comparatively short, likely because of ^{137}Cs dispersal by water currents from these comparatively open ecosystems. The shortest *Tes* for fish were from European water bodies soon after the Chernobyl incident ([Table 1](#)). [Santschi et al. \(1990\)](#) reported that loosely bound ^{137}Cs may be rapidly removed from terrestrial soils by flushing soon after deposition. Rapid initial removal may also be characteristic of aquatic ecosystems followed by slower rates of removal after the easily removed ^{137}Cs has been dispersed.

Table 1.

Comparison of ^{137}Cs effective half-lives in various fish species collected from different locations.

| Taxon | Location | Notes | <i>Te</i> (yrs) |
|---|--------------------------|-------------------------------|-----------------|
| Mostly bass & sunfishes (Paller et al., 2008 and Paller et al., 2002) | SRS streams | Flowing | 3.5-9.0 |
| Bass, sunfishes & catfishes (Paller et al., 2008 and Paller et al., 2002) | Savannah River | Flowing | 7.6-8.1 |
| Bass & sunfishes (Paller et al., 2008 and Paller et al., 2002) | Par Pond (SRS reservoir) | Fast water exchange | 4.8-5.0 |
| Bass & sunfishes (Paller et al., 2008 and Paller et al., 2002) | Pond B (SRS reservoir) | Slow water exchange | 13.4-16.7 |
| Bullheads (McCreedy et al., 2009) | Pond B (SRS reservoir) | Slow water exchange | 50 |
| Various (Mohler et al., 1997) | Pond B (SRS reservoir) | Slow water exchange | 5-19 |
| Various (Noshkin et al., 1997) | Bikini & Enewetak atolls | Marine | 9-12 |
| Perch, roach, trout, and charr (Forseth et al., 1991 , Brittain et al., 1991 and Brittain et al., 1996) | European water bodies | Soon after Chernobyl accident | 1.0-2.9 |
| Pike (Forseth et al., 1991 , Brittain et al., 1991 and Brittain et al., 1996) | European water bodies | Soon after Chernobyl accident | 2.0-4.9 |
| Nine species (Kryshev et al., 1993) | Chernobyl cooling ponds | Soon after Chernobyl accident | <2 |

As previously discussed, T_{es} for fish from most SRS water bodies have remained relatively constant over time. However, this was not the case in Par Pond when it was partially drained and refilled during 1991–1996 ([Paller et al., 2002](#)). During and following the refill, about one-half of the lake bottom was exposed, permitting the erosion of contaminated sediments from exposed areas and their transport into the lake basin. ^{137}Cs levels in largemouth bass *M. salmoides* and bluegill *Lepomis macrochirus* increased markedly at this time before stabilizing and subsequently declining. More details concerning the effects of the Par Pond drawdown on ^{137}Cs in fish can be found in [Paller et al. \(2002\)](#). T_{es} shown in [Table 1](#) were calculated from data collected before the drawdown.

Comparisons with other studies can also be made for ^{137}Cs removal rates in soil and vegetation ([Table 2](#)). These have been expressed in the literature as both T_{es} and ecological half-lives (T_{cs}). The latter were determined for the Creek Plantation data using the equation $T_c = T_e * Tr / (Tr - T_e)$ to facilitate comparisons with researchers who reported T_{cs} rather than T_{es} . The T_c for ^{137}Cs in SRS floodplain soils was comparable to T_{cs} for similar soils in Europe composed mostly of clay but longer than for sandy and loamy soils, which may have different drainage and sorption properties than clay soils. ^{137}Cs removal rates from Creek Plantation soils were substantially slower than from Par Pond sediments that were freshly exposed as a result of the previously described Par Pond drawdown. The exposure of these formerly submerged sediments may have resulted in comparatively rapid ^{137}Cs removal processes more characteristic of newly contaminated soils (e.g., erosion and leaching) than older contaminated soils ([Hinton et al., 1999](#)). Generally, T_{es} and T_{cs} for Creek Plantation floodplain soil and vegetation were somewhat longer than observed at most other locations. Contributing factors may have been the flat topography and extensive vegetation cover on the floodplain, which reduced erosion, and the relatively tight chemical bonds that may form between ^{137}Cs and soil particles after long periods of time.

Table 2. Comparison of ^{137}Cs loss rates in soil and vegetation from different locations.

| Medium | Location | Notes | T_e | T_c^a | Decrease |
|------------|---|--------------------------------|-----------|---------|-----------------|
| Soil | SRS Floodplain (this study) | Mostly clay | 17.0 | 38.9 | 81% over 29 yrs |
| Soil | Europe (Pröhl et al., 2006) | Clay | | 32.8 | |
| Soil | Europe (Pröhl et al., 2006) | Sand | | 8.1 | |
| Soil | Europe (Pröhl et al., 2006) | Loam | | 4.1 | |
| Soil | Par Pond, SRS (Hinton et al., 1999) | Exposed sediment | | | 36% over 3 yrs |
| Vegetation | Creek Plantation | Herbaceous | 13.4 | 24.2 | |
| Vegetation | Steel Creek (Peles et al., 2002) | Herbaceous & aquatic | 4.9–8.4 | | |
| Vegetation | Pond B (Peles et al., 2002) | Aquatic | 11.0–12.7 | | |
| Vegetation | Pacific Islands (Robinson et al., 2003) | Trees in CaCO_3 soils | 8.5 | | |
| Vegetation | Susquehanna River (Cehn, 2007) | Periphyton | 9.2 | | |

a

$T_c = T_e Tr / (Tr - T_e)$, where T_c is the ecological half-life and Tr is the radioactive half-life.

Long-term trending of ^{137}Cs in shallow floodplain soil, vegetation, and deer from the SRS indicated T_e 's of about 13–17 years, which are about one half the ^{137}Cs physical half-life of 30.2 years. The T_e 's for ^{137}Cs in stream and river fish were shorter, about 3.5–9.0 years. These results indicate that natural environmental processes have the potential to restore ecosystems contaminated with ^{137}Cs more rapidly than expected on the basis of radioactive decay. However, the long-term SRS data also indicate that natural and anthropogenic factors can significantly affect T_e 's for ^{137}Cs . The extent of these effects will be determined by the degree to which the factors influence ^{137}Cs activity concentrations, the time-scales on which they operate, and their consistency of operation. Therefore, it is desirable have as long a period of record as possible for calculating T_e s and risky to extrapolate T_e s beyond the period of record unless the processes governing ^{137}Cs removal are clearly understood.

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References

[Brittain et al., 1991](#)

J.E. Brittain, A. Storruste, E. Larsen

Radiocesium in brown trout (*Salmo trutta*) from a subalpine lake ecosystem after the Chornobyl reactor accident

J. Environ. Radioact., 14 (1991), pp. 181–191

[Brittain et al., 1996](#)

J. Brittain, U. Bergström, L. Håkanson, R. Helling, L. Monte, V. Suolanen

Estimation of ecological half-lives of caesium-137 in lakes contaminated by Chernobyl fallout

Proceedings of the International Symposium on Environmental Impact of Radioactive Releases, Vienna, Austria, May 8–12, 1995, International Atomic Energy Agency, IAEA-SM-339/58, Vienna, Austria (1996), pp. 291–298

[CDC/NCI, 2002](#)

CDC/NCI

Progress Report to Congress: A Feasibility Study of the Health Consequences to the American Population of Nuclear Weapons Test Conducted by the United States and Other Nations

Prepared by the Centers for Disease Control and Prevention and the National Cancer Institute, Washington, DC (2002)

[Cehn, 2007](#)

J.I. Cehn

Decay of environmental ¹³⁷Cs

Radiat. Saf. J., 93 (2007), p. 325

[Eisenbud, 1987](#)

M. Eisenbud

Environmental Radioactivity

(third ed.) Academic Press, San Diego, CA (1987), pp. 272–280

[Evans et al., 1983](#)

D.W. Evans, J.J. Alberts, R.A. Clark

Reversible ion-exchange fixation of cesium-137 leading to mobilization from reservoir sediments

Geochim. Cosmochim. Acta, 47 (1983), pp. 1041–1049

|

[Fledderman et al., 2007](#)

P.D. Fledderman, G.T. Jannik, M.H. Paller

An overview of ¹³⁷Cs contamination in a southeastern swamp environment

Radiat. Saf. J., 93 (2007), pp. 160–164

T. Forseth, O. Ugedal, B. Jonsson, A. Langeland, O. Njastad

Radiocaesium turnover in arctic charr (*Salvelinus alpinus*) and brown trout (*Salmo trutta*) in a Norwegian lake

J. Appl. Ecol., 28 (1991), pp. 1053–1067

[Gaines and Novak, 2011](#)

K.F. Gaines, J.M. Novak

Spatiotemporal-toxicodynamic Modeling of Cs-137 to Estimate White-tailed Deer Background Levels for the Department of Energy's Savannah River Site

SRNS-RP-2009-01283, Revision 1.3 Savannah River Nuclear Solutions, Aiken, SC (2011)

[Gladden et al., 1985](#)

J.B. Gladden, K.L. Brown, M.H. Smith, A. Towns

Distribution of g exposure rates in a reactor effluent stream flood plain system

Health Phys., 48 (1985), pp. 49-59

[Hinton et al., 1999](#)

T.G. Hinton, C.M. Bell, F.W. Whicker, T. Philippi

Temporal changes and factors influencing ¹³⁷Cs concentration in vegetation colonizing an exposed lake bed over a three-year period

J. Environ. Radioact., 44 (1999), pp. 1-19

[Jannik and Scheffler, 2011](#)

G.T. Jannik, R. Scheffler

Critical Radionuclide and Pathway Analysis for the Savannah River Site

SRNL-STI-2011-0503 Savannah River National Laboratory, Aiken, SC (2011)

[Knox et al., 2001](#)

A.S. Knox, T.G. Hinton, D.I. Kaplan

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

WSRC-TR-2001-00455, Rev. 0 Savannah River National Laboratory, Aiken, SC (2001)

[Kryshev et al., 1993](#)

I.I. Kryshev, I.N. Ryabov, T.G. Sazykina

Using a bank of predatory fish samples for bioindication of radioactive contamination of aquatic food chains in the area affected by the Chernobyl accident

Sci. Total Environ., 139/140 (1993), pp. 279-285

[McCreedy et al., 2009](#)

C.M. McCreedy, C.H. Jagoe, L.T. Glickman, I.L. Brisbin

Bioaccumulation of cesium-137 in yellow bullhead catfish (*Ameiurus natalis*) inhabiting an abandoned nuclear reactor reservoir

Environ. Toxicol. Chem., 16 (2009), pp. 328-335

[Mohler et al., 1997](#)

H.J. Mohler, F.W. Whicker, T.G. Hinton

Temporal trends of ¹³⁷Cs in an abandoned reactor cooling reservoir

J. Environ. Radioact., 37 (1997), pp. 251-268

[Noshkin et al., 1997](#)

V.E. Noshkin, W.L. Robison, K.M. Wong, J.L. Brunk, R.J. Eagle, H.E. Jones

Past and present levels of some radionuclides in fish from Bikini and Enewetak atolls

Health Phys., 73 (1997), pp. 49-65

[Paller et al., 1999](#)

M.H. Paller, J.W. Littrell, E.L. Peters

Ecological half-lives of ^{137}Cs in fishes from the Savannah River Site

Health Phys., 77 (1999), pp. 392-402

[Paller et al., 2002](#)

M.H. Paller, J.W. Litrell, E.L. Peters

Ecological half-lives of ^{137}Cs in fishes

Proceedings of the 12th Biennial RPSD April Topical Meeting, Santa Fe, USA (April 14-18, 2002)

[Paller et al., 2005](#)

M.H. Paller, D.E. Fletcher, T. Jones, S.A. Dyer, J.J. Isely, J.W. Littrell

Potential of largemouth bass as vectors of ^{137}Cs dispersal

J. Environ. Radioact., 80 (2005), pp. 27-43

[Paller et al., 2008](#)

M.H. Paller, P.D. Fledderman, G.T. Jannik

Changes in Cs-137 concentrations in soil and vegetation on the floodplain of the savannah river over a 30 year period

J. Environ. Radioact., 99 (2008), pp. 1302-1310

[Peles et al., 2002](#)

J.D. Peles, M.H. Smith, I.L. Brisbin Jr.

Ecological half-life of ^{137}Cs in plants associated with a contaminated stream

J. Environ. Radioact., 59 (2002), pp. 169-178

[Pinder et al., 1980](#)

J.E. Pinder III, C.T. Garten, D. Paine

Factors affecting radiocesium uptake by plants inhabiting a contaminated floodplain

Acta Oecologia, 1 (1980), pp. 3-10

[Pröhl et al., 2006](#)

G. Pröhl, S. Ehlken, I. Fiedler, G. Kirchner, E. Klemm, G. Zibold

Ecological half-lives of ^{90}Sr and ^{137}Cs in terrestrial and aquatic ecosystems

J. Environ. Radioact., 91 (2006), pp. 41-72

[Robinson et al., 2003](#)

W.L. Robinson, C.L. Conrado, K.T. Bogen, C.A. Stoker

The effective and environmental half-life of ^{137}Cs at coral islands at the former US nuclear test site

J. Environ. Radioact., 69 (2003), pp. 207-223

[Santschi et al., 1990](#)

P.H. Santschi, S. Bollhalder, S. Zingg, A. Lück, K. Farrenkothen

The self-cleaning capacity of surface waters after radioactive fallout. Evidence from European waters after Chernobyl, 1986-1988

Environ. Sci. Technol., 24 (1990), pp. 519-527

[Whicker and Schultz, 1982](#)

F.W. Whicker, V. Schultz

Radioecology: Nuclear Energy and the Environment, vol. IICRC Press, Inc, Boca Raton, FL (1982)

[Whicker et al., 1990](#)

F.W. Whicker, J.E. Pinder III, J.W. Bowling, J.J. Alberts, I.L. Brisbin Jr.

Distribution of long-lived radionuclides in an abandoned reactor cooling reservoir

Ecol. Monogr., 60 (1990), pp. 471-496