

ECOLOGICAL HALF-LIVES OF ¹³⁷CS IN FISHES

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SUMMARY

Ecological half-lives (T_e 's) were estimated for ¹³⁷Cs in fishes from two reservoirs and three streams on the Savannah River Site, a nuclear weapons material production facility in South Carolina. "Ecological half-life" is the time required for a given contaminant concentration to decrease by 50% from combined physical, chemical, and/or biological processes that remove it from an ecosystem or render it biologically unavailable. T_e 's were estimated from ¹³⁷Cs concentrations measured during 1972-1996, following radionuclide releases that occurred primarily during the 1960s and 1970s. T_e 's of centrarchids ranged from 3.2 to 16.7 yr, and all were significantly shorter than expected from the half-life for radioactive decay ($T_p = 30.2$ yr) alone. Smaller data sets for catfishes did not show statistically significant declines in ¹³⁷Cs concentrations. Fishes from the same locations differed in mean ¹³⁷Cs concentrations (highest in largemouth bass and lowest in sunfishes), but in most cases, exhibited similar ¹³⁷Cs T_e 's. The shortest T_e 's occurred in the middle reaches of the streams. T_e 's in lower reaches of the streams were longer, as were T_e 's in one of the reservoirs. T_e 's in Par Pond, which had the shorter mean water residence time of the two reservoirs were nearly comparable to those in the upper portions of the streams. After 1991, however, ¹³⁷Cs concentrations in Par Pond fishes increased following drainage and refilling of the reservoir, which apparently resuspended ¹³⁷Cs buried in the sediments. Recent samples collected during 1997-2000 and presented here demonstrate that ¹³⁷Cs levels in fish have

continued to decline according to our earlier predictions, except for Par Pond and in a stream that drains it, where elevated ¹³⁷Cs levels still persist.

I. BACKGROUND

Environmental releases of cesium-137 (¹³⁷Cs) from nuclear facilities have demonstrated that hydrospheric pathways can be major sources of human exposures. Although ¹³⁷Cs residence times in natural waters are often comparatively short¹, some ¹³⁷Cs enters aquatic food chains and bioaccumulates in the edible skeletal muscle of fishes.² Fishes assimilate ¹³⁷Cs rapidly and eliminate it relatively slowly³, making their consumption a potentially important source of human exposure.

As for any biota, ¹³⁷Cs concentrations in fishes would be expected to decline over time as a result of physical, chemical and biological processes that remove ¹³⁷Cs from the ecosystem or make it biologically unavailable. Such declines, which individually are often single-exponential declines, can often be combined into an effective "ecological half-life" (T_e) that describes the decrease in contaminant concentration in the ecosystem and its biota over time. Such T_e estimates can be useful in predicting how long (and to what degree) contaminated systems may pose an environmental or health risk, and can aid in assessing the success of remediation.⁴

At the Savannah River Site (SRS), a U.S. Department of Energy (DOE) nuclear

production facility in South Carolina, there is concern about possible human health effects resulting from the consumption of fishes contaminated by ^{137}Cs released during the 1960s and 1970s. This concern is unlikely to abate until contamination in fishes reaches levels considered acceptable for human consumption. Environmental monitoring of fishes in SRS streams and reservoirs has produced a unique long-term data set for the estimation of T_e 's for ^{137}Cs in freshwater fishes from warm-water lakes and streams. In contrast, most published studies of ^{137}Cs in fishes are from cold-water, highly acidic, lentic European ecosystems that received fallout from the Chernobyl accident^{4,5} or tropical marine ecosystems⁶.

This paper summarizes previously-published information⁷, together with additional recently-collected data that permits evaluation of the trends and T_e predictions noted in our previous study.

II. METHODS AND MATERIALS

A. Study area

The SRS, located on the Atlantic coastal plain near Aiken (South Carolina, USA), is an 803-km² nuclear weapons materials production site established in 1951. By 1955, the SRS had five operating nuclear production reactors. Water from the Savannah River was pumped through the cooling circuits of these reactors before being released into three tributaries of the Savannah River (Steel Creek, Four Mile Creek, and Lower Three Runs) and two reservoirs (Par Pond and Pond B). Various operational problems resulted in releases of ^{137}Cs into the cooling waters discharged from four of these reactors. It is estimated that releases of ^{137}Cs were approximately 1.220 TBq from C-Reactor into Four Mile Creek; 8.200 TBq from R-Reactor into Pond B, Par Pond, and Lower Three Runs Creek (4.70 TBq to Pond B alone); and 9.435 TBq from P-Reactor into Steel Creek and Par Pond. An additional 1.070 TBq of ^{137}Cs entered Steel Creek from L-Reactor. The vast majority of these ^{137}Cs releases occurred from 1960 through 1970, although minor releases continued until 1985. Additional information on the timing, quantity, and points of release can be found in Paller et al.⁷

B. Sampling and analytical methods

Analysis of ^{137}Cs in fishes of the SRS began in 1961, and many sites were sampled annually or every few years. Collection sites included Steel Creek near SRS Road A (the middle reaches of Steel Creek), the mouth of Steel Creek near its confluence with the Savannah River, Four Mile Creek near Road A (the middle reaches of Four Mile Creek), Par Pond, Pond B, and Lower Three Runs Creek near Patterson Mill (the middle reaches of Lower Three Runs Creek). Means, maximums, and sample sizes were documented in annual reports, and these reports form the source of most of the data in this paper, although additional data concerning ^{137}Cs in Pond B largemouth bass, were from several studies conducted by the University of Georgia, Savannah River Ecology Laboratory. Collection methods generally included angling and the use of traps.

Fishes were separated into several categories, including largemouth bass (*Micropterus salmoides*), sunfishes (primarily redbreast sunfish, *Lepomis auritus*, bluegill, *L. macrochirus*, and spotted sunfish, *L. punctatus*), and bullheads (primarily yellow bullhead, *Ameiurus natalis* and flat bullhead, *A. platycephalus*). Combining related species, as practiced at the SRS, introduced an indeterminate source of variability in the estimates of ^{137}Cs concentration, because closely related species may differ in ^{137}Cs body burdens. Seasonal and age- or size-dependent changes in ^{137}Cs body burdens were also potential sources of unmeasured variability in this study (although all fishes were large enough to be considered potentially edible). However, these sources of variability were likely trivial compared with the large long-term decreases in ^{137}Cs concentrations that were the subject of this study. Potential effects of these were considered in Paller et al.⁷.

Detailed information on analytical procedures and detection limits can also be found in Paller et al.⁷ Briefly, individual whole fish were analyzed prior to 1992. Composite samples, each consisting of fillets from five individuals of a single species, were analyzed in subsequent years. Samples collected after 1989 were counted with a shielded high purity germanium detector. Earlier samples were

counted with NaI(Tl) solid scintillator or Ge(Li) semiconductor detectors.

¹³⁷Cs concentrations were measured for water as well as for fishes. Composited water samples were generally collected with continuous automatic water samplers at the same locations where fish samples were taken. Unfiltered water samples were counted in filled 500-ml containers.⁷

C. Data analysis

Because ¹³⁷Cs concentrations were measured in whole fishes prior to 1992 and fillets afterwards, taxon-specific regression models (based on samples for which both fillets and inedible portions were analyzed) were used to predict concentrations in whole fishes from concentrations in fillets.⁷ The equation describing the loss of ¹³⁷Cs from whole fishes was $dc/dt = -T_e c$, where c was the ¹³⁷Cs concentration (Bq g^{-1}) in whole fish, and T_e was the ecological loss rate constant. Estimates of T_e ($= \log_e 2/k_e$) for each taxonomic group at each location were made from the slopes of \log_e -transformed ¹³⁷Cs concentrations in fishes regressed against year. The ¹³⁷Cs concentrations used in these calculations were sample means, and were weighted by the sample size. Concentrations of ¹³⁷Cs were not corrected for contributions from global fallout, because historical samples collected from local water bodies unaffected by SRS releases indicated that fallout was insignificant compared to reactor releases.⁷

Because T_e represents an exponential decrease only when ¹³⁷Cs inputs are zero, analyses to estimate T_e were restricted to data collected after 1971, when ¹³⁷Cs releases from SRS reactors were negligible or nonexistent. Except for Par Pond, the last year included in the analyses was 1996. In the case of Par Pond, data from 1991-1996 were excluded because Par Pond was partially drained and refilled during these years, resulting in increases in increases in ¹³⁷Cs bioavailability as discussed below.

Analysis of covariance (ANCOVA) was used to determine if rates of ¹³⁷Cs decrease differed among taxa at each sample location. In this analysis \log_e -transformed ¹³⁷Cs body burden

was the dependent variable, time was the covariate, and taxon was the discrete factor.

As described above, the T_e 's reported in our earlier paper⁷ were based on data collected from 1972 to 1996. Since that time, additional data were collected from most locations from 1997-2000. These data have not been included in the T_e computations presented here (which remain the same as in the earlier paper⁷). These data are instead presented (together with the original T_e regressions and their associated 95% prediction intervals⁸) to assess the continuing validity of our previously-documented trends and T_e predictions.

III. RESULTS

Maximum ¹³⁷Cs concentrations in water occurred between 1960 and 1970, and corresponded to the period of maximal ¹³⁷Cs release from the reactors⁷. The highest ¹³⁷Cs concentrations in water, up to $\approx 23 \text{ Bq L}^{-1}$, occurred in Steel Creek. Maximum concentrations in the other water bodies were lower ($\approx 1 \text{ Bq L}^{-1}$). ¹³⁷Cs concentrations in all aquatic systems declined after 1970. These patterns were paralleled by those observed in the fishes, which also reached maximum levels between 1960 and 1970 and then declined. The highest ¹³⁷Cs concentration in fishes, 53.9 Bq g^{-1} , occurred in Steel Creek. Maximum concentrations in Par Pond and Four Mile Creek fishes were lower (6.6 and 5.8 Bq g^{-1} , respectively).

The relationships between ¹³⁷Cs concentration and time were highly predictive for most taxa within most water bodies (Figure 1, Table 1). T_e 's ranged from 3.2 to 16.7 yr (Table 1). The shortest T_e 's occurred in the middle reaches (near Road A) of Steel Creek (3.2-3.5 yr) and Four Mile Creek (4.7-5.0 yr, Table 1). The T_e at the mouth of Steel Creek was longer (7.0 yr), as were the half-lives in Pond B and Lower Three Runs (10.7 to 16.7 yr).

The ANCOVA results indicated that the slopes of the regressions of whole-body ¹³⁷Cs concentration on time did not differ significantly ($P \leq 0.05$) among taxa at each location, i.e., the T_e 's of different taxa within the same water bodies were similar, although

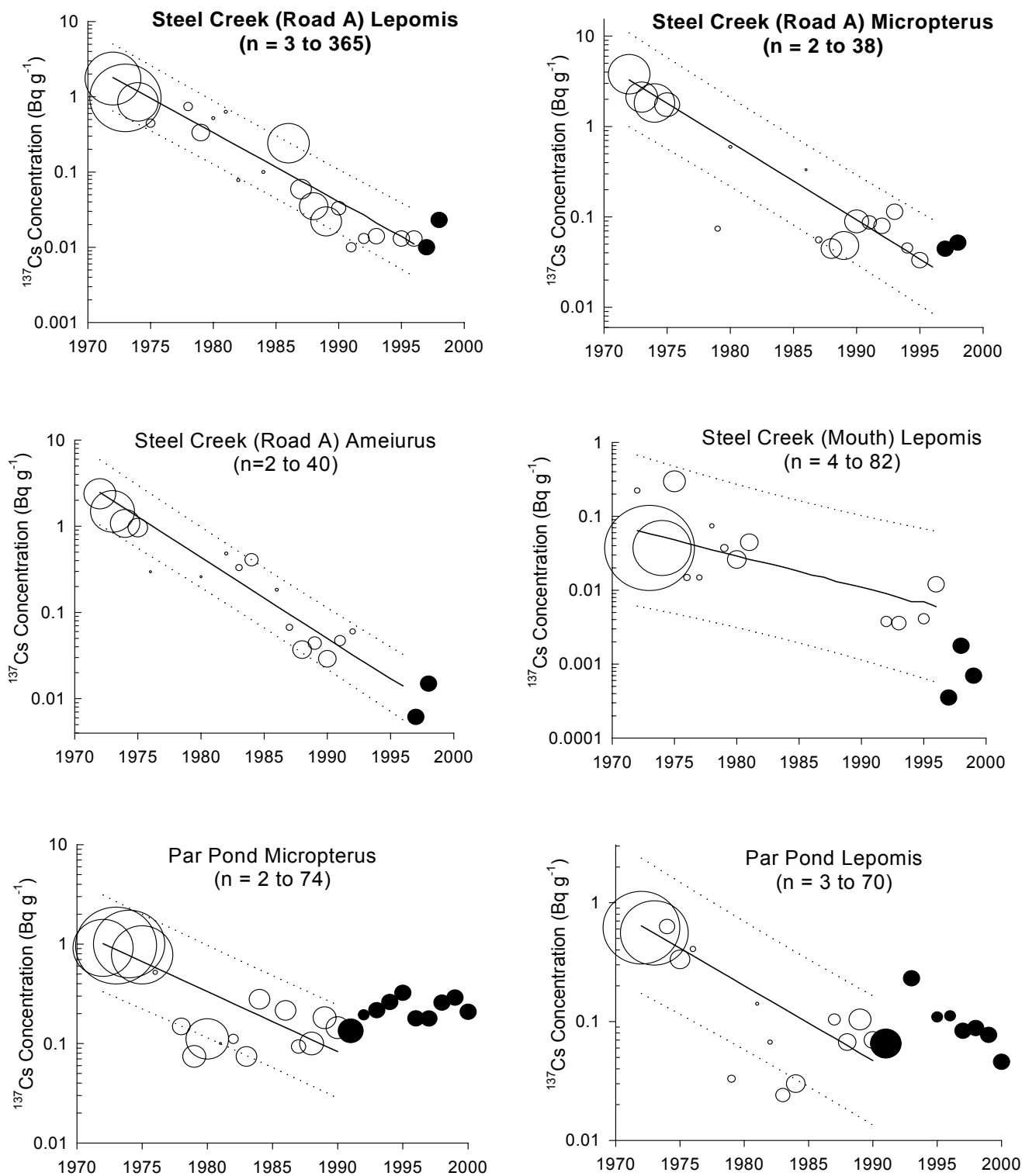


Figure 1. Changes in ^{137}Cs in whole fish regressed against time for fishes at the Savannah River Site. Circles represent data used to compute ^{137}Cs half-lives (T_e 's). Solid circles represent data collected from 1997-2001. Symbol areas are proportional to the number of fish in each sample. Solid lines represent regression lines; dotted lines represent 95% prediction intervals. Statistical parameters for the regression lines appear in Table 1.

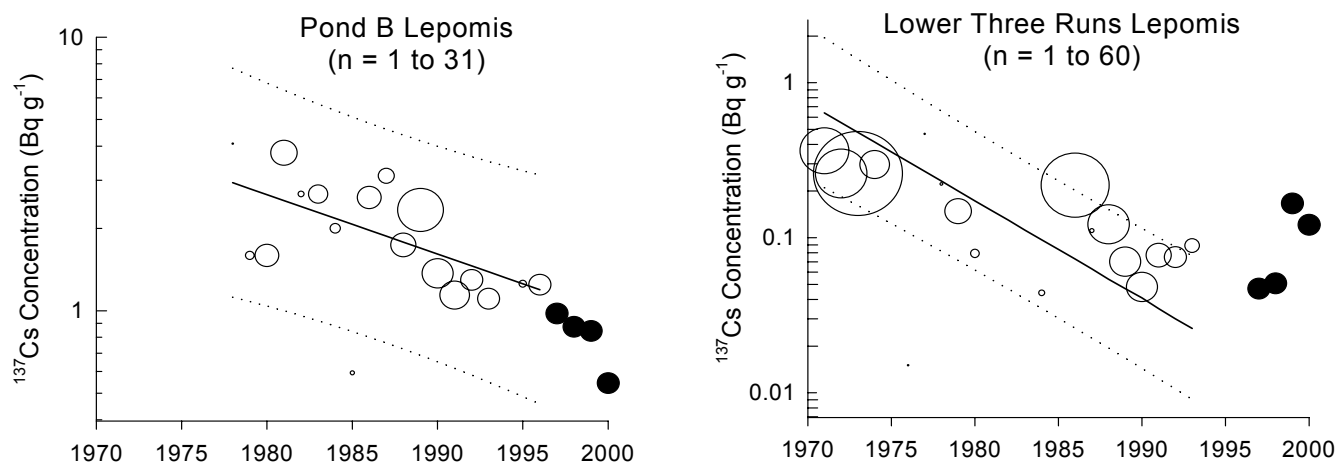


Figure 1. continued.

Table 1. Ecological half-lives (T_e 's) for ^{137}Cs in fish from water bodies on the Savannah River Site. Ecological half-lives were calculated from regression slopes weighted by the number of fish collected each year.

Location	Taxon	Regression Slope (k_e)	r^2	P	Yrs of data	T_e Mean (95% C.I.)
Four Mile Creek	sunfishes	-0.140	0.91	<0.001	17	4.95 (4.63 to 5.32)
Four Mile Creek	largemouth bass	-0.149	0.98	<0.001	13	4.65 (4.31 to 5.05)
Steel Creek (Road A)	sunfishes	-0.211	0.93	<0.001	20	3.29 (2.91 to 3.78)
Steel Creek (Road A)	largemouth bass	-0.198	0.91	<0.001	16	3.50 (3.00 to 4.21)
Steel Creek (Road A)	bullheads	-0.217	0.94	<0.001	15	3.19 (2.81 to 3.69)
Steel Creek (mouth)	sunfishes	-0.099	0.53	0.003	14	7.00 (4.56 to 15.0)
Lower Three Runs	sunfishes	-0.065	0.58	0.001	15	10.7 (7.34 to 19.5)
Par Pond	sunfishes	-0.145	0.72	<0.001	14	4.78 (3.54 to 7.37)
Par Pond	largemouth bass	-0.139	0.65	<0.001	17	4.99 (3.65 to 7.88)
Pond B	sunfishes	-0.050	0.35	0.013	17	13.4 (8.13 to 47.2)
Pond B	largemouth bass	-0.041	0.78	<0.001	22	16.7 (14.3 to 20.3)

mean ^{137}Cs concentrations differed (Table 1). Least-square geometric mean ^{137}Cs levels were 140, 240, and 300 mBq g $^{-1}$ ($P=0.008$) for Steel Creek sunfishes, bullheads, and largemouth bass; 360 and 610 mBq g $^{-1}$ ($P<0.001$) for Four Mile Creek sunfishes and largemouth bass; and 130 and 230 mBq g $^{-1}$ ($P=0.04$) for Par Pond sunfishes and largemouth bass, respectively.

Comparisons of recently collected data with T_e regressions based on 1972-1996 data (1972-1991 for Par Pond) indicated that the previous trend of declining ^{137}Cs concentrations in fishes has continued in most water bodies (Figure 1).

The exceptions are the increases that coincided with the partial draining and refilling of Par Pond (1991 to 1996). ^{137}Cs concentrations in Par Pond fishes remained elevated through 2000, four years after refill was complete. Body burdens of sunfishes from Lower Three Runs, which drains Par Pond (and is thus affected by Par Pond ^{137}Cs concentrations) remain similarly elevated.

IV. DISCUSSION

The T_e 's we observed for ^{137}Cs in fishes were considerably shorter than the 30.2-yr radioactive half-life of ^{137}Cs , and were

presumably the result of various physical and ecological processes that removed ^{137}Cs from the aquatic ecosystems under study and/or made it less bioavailable. However, there were substantial differences among T_e 's for ^{137}Cs in fishes from different water bodies. The comparatively short half-lives in the middle reaches of Steel Creek and Four Mile Creek-Road A (3.2 and 5.0 years) may have resulted from the outflow of sediment-bound and dissolved ^{137}Cs and/or the burial of contaminated sediments by uncontaminated sediments. ^{137}Cs -contaminated sediments removed from the middle reaches of Steel Creek may have been deposited downstream near its mouth, perhaps accounting for the longer T_e for sunfishes at this location. The comparatively longer T_e in Lower Three Runs Creek (10.7 years) may have been due to discharges of dissolved or seston-associated ^{137}Cs from Par Pond.

The T_e 's reported for Par Pond fishes represent a period when changes in ^{137}Cs levels resulted from the processes of physical and biological removal that operated in this ecosystem before disturbance. These T_e 's (4.8 and 5.0 years) were only slightly longer than for Steel Creek or Four Mile Creek fishes (Table 1). In 1992, however, ^{137}Cs levels in Par Pond largemouth bass and sunfish increased, doubtless because of resuspension of sediments and/or changes in water chemistry associated with the draining and subsequent refill of Par Pond. During and following the refill, approximately one-half of the lake bottom was exposed, permitting sediments from shallow areas to erode into the lake basin. Because the pumping of water from the Savannah River into Par Pond was also stopped at this time, the water entering Par Pond was from rainfall and runoff from the local watershed. This water was low in dissolved potassium compared with Savannah River water. The resulting decrease in dissolved potassium levels may have contributed to the increased ^{137}Cs concentrations observed in fishes after 1992.⁹ These same factors are probably also responsible for the increases in ^{137}Cs recently observed in fishes from Lower Three Runs, which received water discharged from Par Pond. The data for *Lepomis* in Par Pond, however, suggest that the T_e for these fishes may be returning to its previously-estimated value.

The longest T_e 's for ^{137}Cs in all fishes were observed in Pond B. In addition to the relatively long T_e 's for largemouth bass and sunfishes observed in this study (16.7 and 13.4 years, respectively), a 50 yr T_e for ^{137}Cs in bullheads from Pond B has been reported by other researchers¹⁰. Factors that may contribute to relatively long T_e 's in Pond B include a relatively low rate of water turnover, low potassium concentrations, and large amounts of rooted aquatic macrophytes (which may translocate sediment-bound cations into the water column¹¹). The exceptionally long T_e reported for Pond B bullheads¹⁰ suggests a unique exposure scenario for this species (due to habitat or feeding preferences) or changes in ^{137}Cs bioavailability in Pond B. Further research will be required to determine causes with certainty.

The results of this study are comparable with those of other studies of ^{137}Cs T_e 's in fishes. An earlier study of Pond B fishes indicated that ^{137}Cs T_e 's ranged from approximately 5 to 19 yr.¹² A 9 to 12 yr T_e range was reported for fishes from lagoons in Bikini and Enewetak atolls.⁶ However, the T_e 's observed at SRS were often longer than those observed for fishes in European water bodies after contamination by Chernobyl fallout. T_e 's for several years after deposition were about 1.0–2.0 yr for fishes at lower trophic levels (perch and roach), 1.0–2.9 yr for brown trout and Arctic charr, and 2–4.9 yr for pike^{3,4,13}. T_e 's for nine species of fishes from the Chernobyl NPP cooling pond were generally <2 yr.¹⁴ The reasons for these relatively short T_e 's are unclear, although removal rates for ^{137}Cs in Steel Creek also appeared to be more rapid in the years initially following release⁷, offering a possible parallel to the rapid T_e 's observed near Chernobyl. In terrestrial ecosystems, some ^{137}Cs is bound loosely to terrestrial soils and may be removed rapidly by flushing¹. Similar phenomena may occur in aquatic ecosystems, resulting in a rapid initial rate of decrease in ^{137}Cs levels in fishes.

V. CONCLUSIONS

Data from streams and reservoirs on the SRS indicated that ^{137}Cs T_e 's in fishes were substantially shorter than expected from the rate of radioactive decay alone. Particularly high rates of removal were observed in areas

characterized by high rates of water turnover, rather than high sedimentation rates. Should current T_e trends continue, fishes from most or all contaminated ecosystems on the SRS will likely reach ^{137}Cs concentrations acceptable for consumption in not more than 50 yr. This consideration is important, because potential consumption of radioactive fishes is considered a principle impediment to unregulated human use of the SRS.

Data from Par Pond indicate that anthropogenic disturbances may result in relatively persistent increases in ^{137}Cs levels in fishes. By inference, it is also possible that invasive cleanup efforts could increase radiological risk to the public by resuspending buried radionuclides, possibly increasing their bioavailability and incorporation into human food webs. In contrast, data from other SRS water bodies suggest that natural succession and concomitant sedimentation processes in relatively undisturbed waters will often lead to burial and sequestration of bioavailable radionuclides until they decay.

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VII. REFERENCES

1. 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," *Environmental Science and Technology* 24: 519-527, (1990).
2. S.E. Kolehmainen, "The balances of Cs-137, stable cesium, and potassium of bluegill (*Lepomis macrochirus*) and other fish in White Oak Lake", *Health Physics* 23: 301-315, (1972).
3. 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", *Journal of Applied Ecology* 28:1053-1067, (1991).

4. J. Brittain, U. Bergström, L. Håkanson, R. Heling, 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, pp 291-298, International Atomic Energy Agency, IAEA-SM-339/58, Vienna, Austria, (1996).
5. L. Håkanson, J. Brittain, L. Monte, R. Heling, U. Bergström, V. Suolanen, "Modeling of radiocesium in lakes—the VAMP model", *Journal of Environmental Radioactivity* 33:255-308, (1996).
6. 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 Physics* 73:49-65, (1997).
7. M.H. Paller, J.W. Littrell, E.L. Peters, "Ecological half-lives of ^{137}Cs in fishes from the Savannah River Site", *Health Physics* 77:392-402, (1999).
8. R.R. Sokal, F.J. Rohlf, *Biometry, second edition*, p. 477, W.H. Freeman and Company, San Francisco, (1981).
9. F.W. Whicker, T.G. Hinton, D.J. Niquette, "Effects of a partial drawdown on the dynamics of ^{137}Cs in an abandoned reactor cooling reservoir", *Studies in Environmental science: Freshwater and Estuarine Radioecology*, Hilton, Desmet, Comans, Blust, deBetencourt, eds., pp.193-202, Elsevier, Amsterdam (1997).
10. C.D. McCreedy, C.H. Jagoe, L.T. Glickman, I.L., Jr. Brisbin, "Bioaccumulation of cesium-137 in yellow bullhead catfish (*Ameiurus natalis*) inhabiting an abandoned nuclear reactor reservoir" *Environmental Toxicology and Chemistry* 16: 328-335, (1997).
11. F.W. Whicker, J. E. III Pinder, J.W. Bowling, J. J. Alberts, I. L. Jr. Brisbin, "Distribution of long-lived radionuclides in an abandoned

reactor cooling reservoir", *Ecological Monographs* 60:471-496, (1990).

12. H.J. Mohler, F.W. Whicker, T.G. Hinton, "Temporal trends of ¹³⁷Cs in an abandoned reactor cooling reservoir", *Journal of Environmental Radioactivity* 37:251-268, (1997).

13. J.E. Brittain, A. Storruste, E. Larsen, "Radiocesium in brown trout (*Salmo trutta*) from a subalpine lake ecosystem after the Chornobyl reactor accident", *Journal of Environmental Radioactivity* 14:181-191, (1991).

14. 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 Chornobyl accident", *The Science of the Total Environment* 139/140:279-285; (1993).