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## **Long-Term Assessment of Critical Radionuclides and Associated Environmental Media at the Savannah River Site – 13038**

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### **ABSTRACT**

During the operational history of the Savannah River Site (SRS), many different radionuclides have been released from site facilities. However, only a relatively small number of the released radionuclides have been significant contributors to doses and risks to the public. At SRS dose and risk assessments indicate tritium oxide in air and surface water, and Cs-137 in fish and deer have been, and continue to be, the critical radionuclides and pathways. In this assessment, in-depth statistical analyses of the long-term trends of tritium oxide in atmospheric and surface water releases and Cs-137 concentrations in fish and deer are provided. Correlations also are provided with 1) operational changes and improvements, 2) geopolitical events (Cold War cessation), and 3) recent environmental remediation projects and decommissioning of excess facilities. For example, environmental remediation of the F- and H-Area Seepage Basins and the Solid Waste Disposal Facility have resulted in a measurable impact on the tritium oxide flux to the onsite Fourmile Branch stream. Airborne releases of tritium oxide have been greatly affected by operational improvements and the end of the Cold War in 1991. However, the effects of SRS environmental remediation activities and ongoing tritium operations on tritium concentrations in the environment are measurable and documented in this assessment. Controlled hunts of deer and feral hogs are conducted at SRS for approximately six weeks each year. Before any harvested animal is released to a hunter, SRS personnel perform a field analysis for Cs-137 concentrations to ensure the hunter's dose does not exceed the SRS administrative game limit of 0.22 millisievert (22 mrem). However, most of the Cs-137 found in SRS onsite deer is not from site operations but is from nuclear weapons testing fallout from the 1950's and early 1960's. This legacy source term is trended in the SRS deer, and an assessment of the "effective" half-life of Cs-137 in deer (including the physical decay half-life and the environmental dispersion half-life) is provided. The "creek mouth" fisherman is the next most critical pathway at SRS. On an annual basis, three species of fish (panfish, catfish, and bass) are sampled from the mouths of the five SRS streams. Three composites of up to five fish of each species are analyzed from each sampling location. Long-term trending of the Cs-137 concentrations in fish and the subsequent doses from consumption of SRS fish is provided.

### **INTRODUCTION**

SRS is a large (over 800 km<sup>2</sup>) DOE facility located in the upper coastal plain of South Carolina near Aiken, South Carolina, and it shares a border with the Savannah River for approximately 44 km (see Fig. 1).



Fig. 1 SRS Location

The U.S. Atomic Energy Commission, a DOE predecessor agency, established SRS in the early 1950s, with its primary mission to produce special nuclear materials (SNM) (such as Pu-239 and tritium) used in the production of nuclear weapons. Five SNM-production reactors, along with support facilities to separate and purify the reactor products, were constructed and operated until 1988. With the end of the Cold War in 1991, waste management, environmental remediation, tritium reprocessing, plutonium disposition, and facility deactivation and decommissioning activities became primary missions for SRS.

## DISCUSSION

Throughout the past 60 years, measurable releases of over 50 radionuclides and various non-radiological contaminants into the atmosphere, onsite streams and seepage basins 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 [1].

The major source of residual radioactivity at SRS is global fallout of fission products from above ground weapons testing. As shown in Table I, a worldwide total of about 480 megatons of nuclear weapons were detonated above ground.

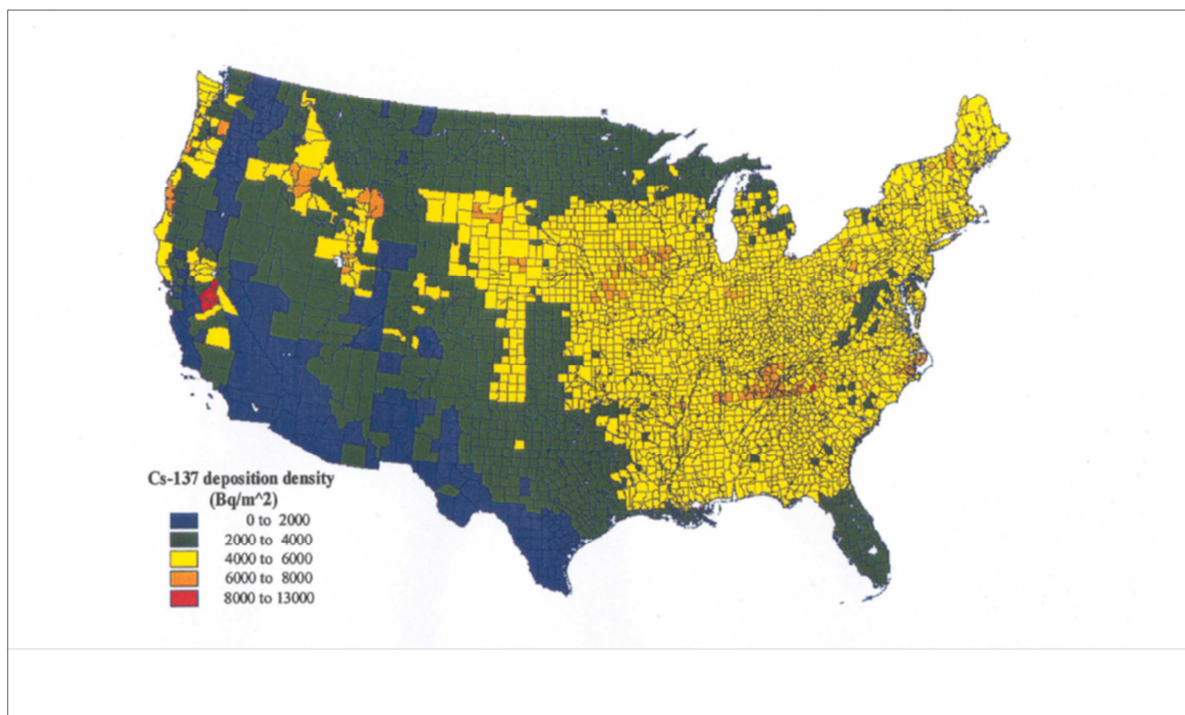
### Cs-137 in the SRS Environment

Due to relatively wetter weather conditions in the eastern US, the wet deposition of fairly long-lived radionuclides (such as Cs-137 with a 30 y half-life) was enhanced in this region of the US during the time of above ground weapons testing. A total of about  $1.3 \times 10^6$  TBq ( $3.5 \times 10^7$  Ci) was introduced to the atmosphere during above ground testing [2].

TABLE I. Worldwide Above Ground Nuclear Weapons Tests

Nation	Number of Above Ground Detonations	Years	Total Yield
United States	216	1945-1962	153.8 mt
U.S.S.R.	214	1949-1962	281.6 mt
United Kingdom	21	1952-1958	10.8 mt
France	46	1960-1974	11.4 mt
P.R.China	23	1964-1980	21.5 mt
South Africa	1	1979	0.003 mt

As shown in Fig. 2 [3], the deposition density of Cs-137 in this region of the US (including the SRS area) ranges between 4,000 and 6,000 Bq/m<sup>2</sup>, with some localized areas receiving even higher deposition. Because of anthropogenic activities such as agriculture and suburban and urban developments, much of this cesium has been dispersed and is no longer bioavailable in the environment. However, at SRS, less than 10% of the site has been impacted by industrial activities and the rest has remained as managed forested areas by the US Forest Service since the early 1950's. Because of this protection, a much larger fraction of the Cs-137 deposited on SRS during the 1950's and early 1960's remains bioavailable to higher trophic level animals, such as deer on the site. This phenomenon also is observed at other large protected land areas such as military bases and National/State Forests [4].

Fig. 2 Cs-137 Deposition Density (Bq/m<sup>2</sup>) in the United States



SRS operational releases of Cs-137 to the air and site streams are shown in Fig. 3 and Fig. 4, respectively [1]. For the years 1995 to 2010, the total measured atmospheric release of Cs-137 from SRS was about 0.12 TBq (3.7 Ci). Most of this was released in 1955 during H-Area startup (0.052 TBq - primarily from leakage at the sand filter) and in 1987 (0.041 TBq - from a failed steam valve at a waste evaporator in H-Tank Farm). The total amount of liquid releases of Cs-137 to SRS streams was about 21 TBq (568 Ci). An additional 50 TBq (1,340 Ci) were discharged to onsite seepage basins.

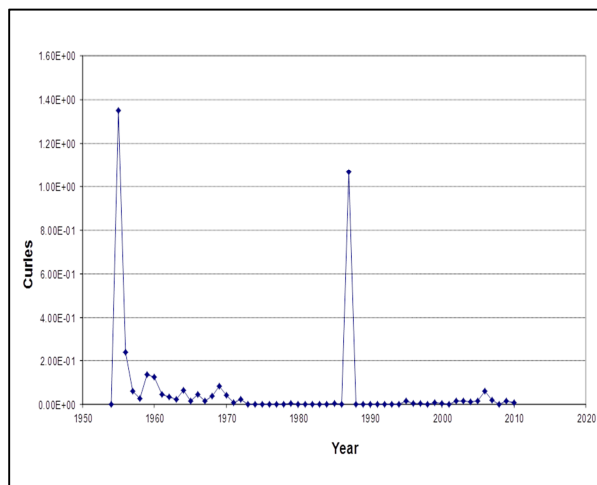


Fig. 3. Airborne Releases of Cs-137 (Ci¹)

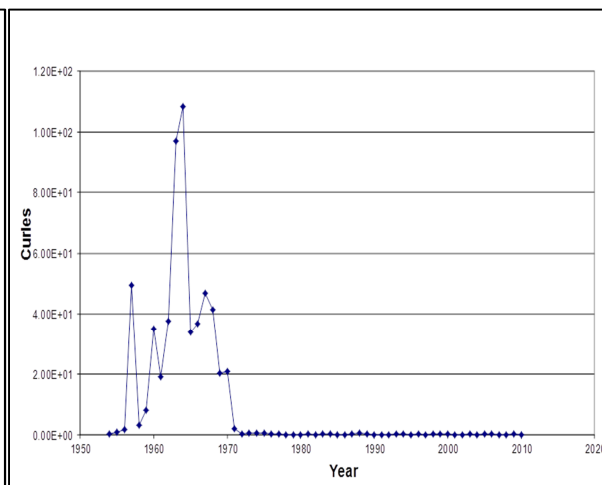


Fig. 4. Liquid Releases of Cs-137 (Ci¹)

<sup>1</sup> 1 Ci = 3.7x10<sup>10</sup> Bq

### Cs-137 in Soil and Vegetation

In the early 1960's, an area of the Savannah River Swamp located downstream of the SRS was contaminated with Cs-137 during periods of high river water. In 1974, a thorough assessment of radionuclide levels in various environmental media was performed in this area. During this assessment, a series of 10 ground survey transects were established ranging from 240 to 3,200 feet in length as shown in Fig. 5. Fifty-two monitoring locations were designated on these transects to allow for continued monitoring at consistent locations. Following the original assessment, a series of surveys were performed which evolved into a long-term surveillance program that continues today. The extensive data collected in these assessments provided a unique long-term record that was used to assess and document the fate and transport of radiocesium in a river swamp environment [5].

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. Cs-137 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.

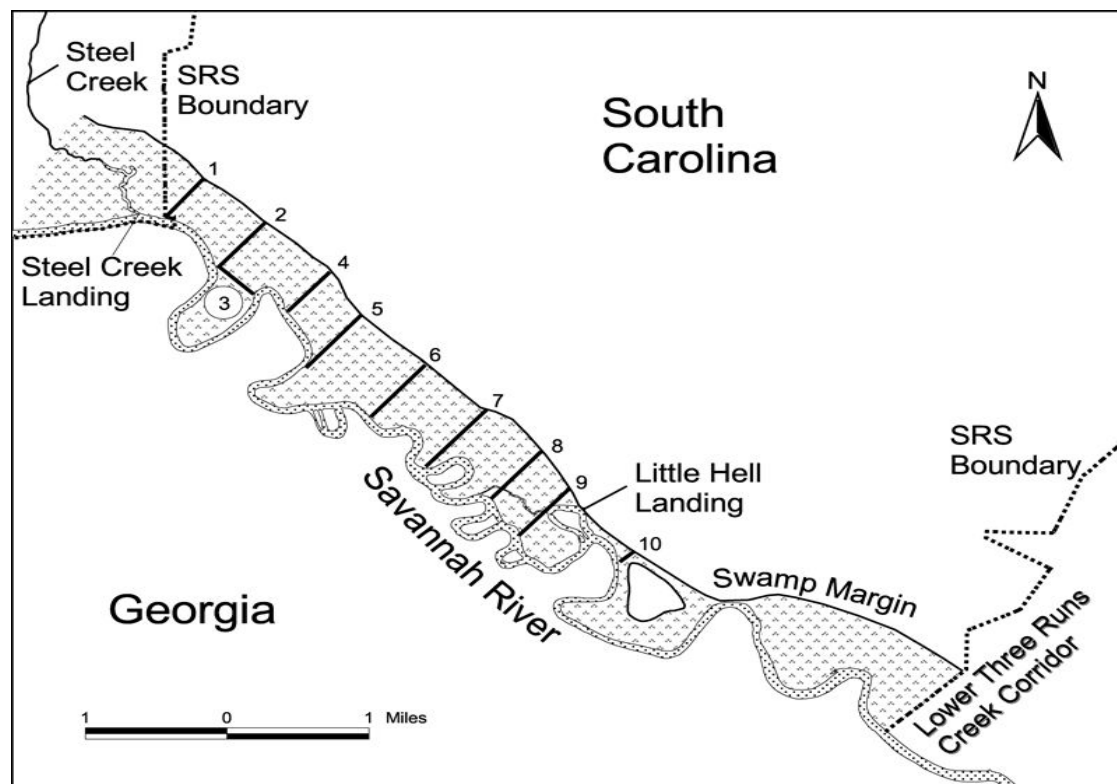


Fig. 5. Survey Transects in the Savannah River Swamp on Creek Plantation

The effective half-lives for Cs-137 in shallow (0-7.6 cm) soil and in vegetation were 14.9 (95% CI = 17.3 - 12.5) years and 11.6 (95% CI = 14.1 – 9.1 yrs) years, respectively, and rates of Cs-137 removal from shallow soil and vegetation did not differ significantly among sample sites. Fig. 6 shows these changes for shallow soil and the associated regression lines. Therefore, potential doses from exposure to the soil and vegetation have declined more rapidly than expected on the basis of radioactive decay (30.17 y half-life for Cs-137) alone because of the relatively short effective half-life of Cs-137 on the Creek Plantation floodplain [5].

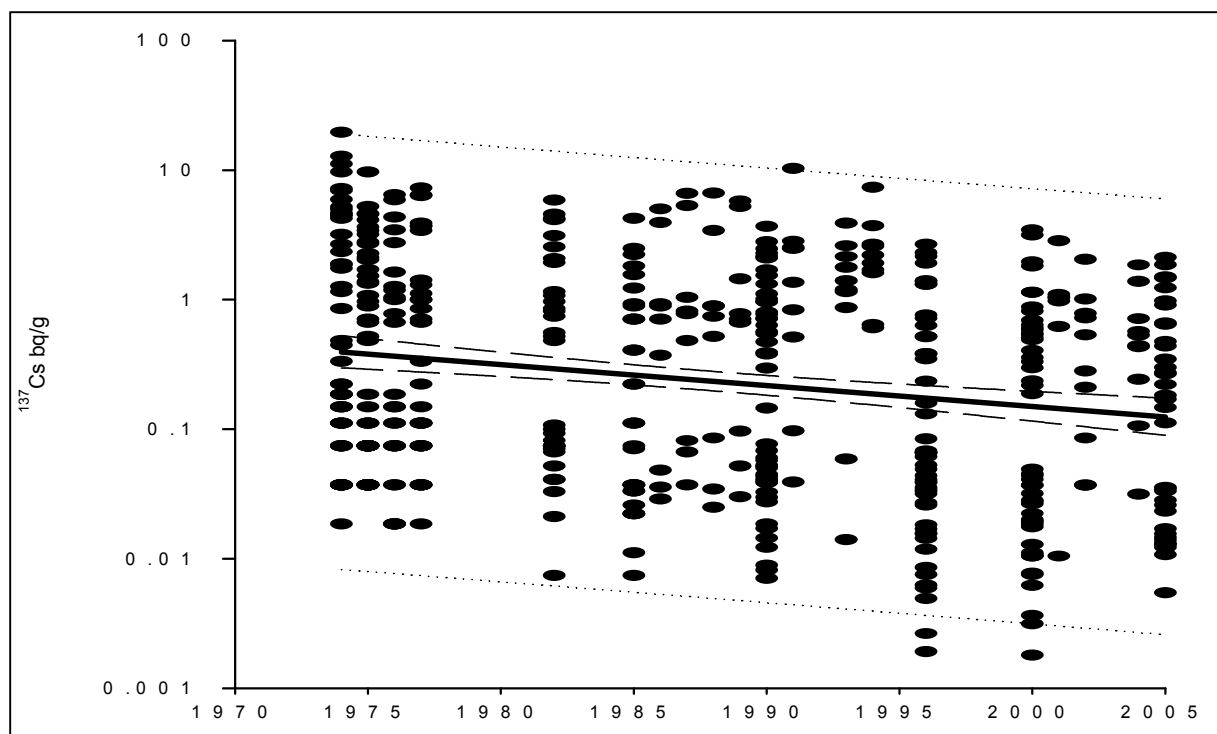


Fig. 6 Changes in Cs-137 concentrations over time in shallow soil (0-7.6 cm) at the Creek Plantation sampling sites. Also shown are regression lines (solid) and 95% confidence intervals (dashed) [5].

### Cs-137 in Deer

Since 1965, annual game animal hunts, open to members of the general public, are conducted at SRS to control the Site's deer and feral hog populations and to reduce animal-vehicle accidents. Prior to release of any animal to a hunter, SRS personnel use portable sodium iodide detectors to perform field analyses for Cs-137. The estimated dose from the consumption of harvested deer or hog meat is determined for every onsite hunter based on the field measured Cs-137 concentration to ensure 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 it exceeds all other SRS sportsman dose scenarios [1].

Fig. 7 provides the long term trend of the mean Cs-137 concentration measured in deer at SRS. From 1965 to 2011, the mean has ranged from a high of about 0.67 Bq/g (18 pCi/g) to a low of 0.037 Bq/g (1 pCi/g). During the past 5 y, the range has been from 0.093 Bq/g (2.4 pCi/g) to 0.044 Bq/g (1.2 pCi/g). Fig. 8 provides these same data on a natural log scale. Also shown in Fig. 8 is the regression line of the data equating to an effective half-life of 15.9 y as compared to the Cs-137 physical half-life of 30.17 y. This effective half-life is similar to Creek Plantation soil and vegetation. Again, the potential dose from consumption of deer meat has declined more rapidly than expected on the basis of radioactive decay alone.

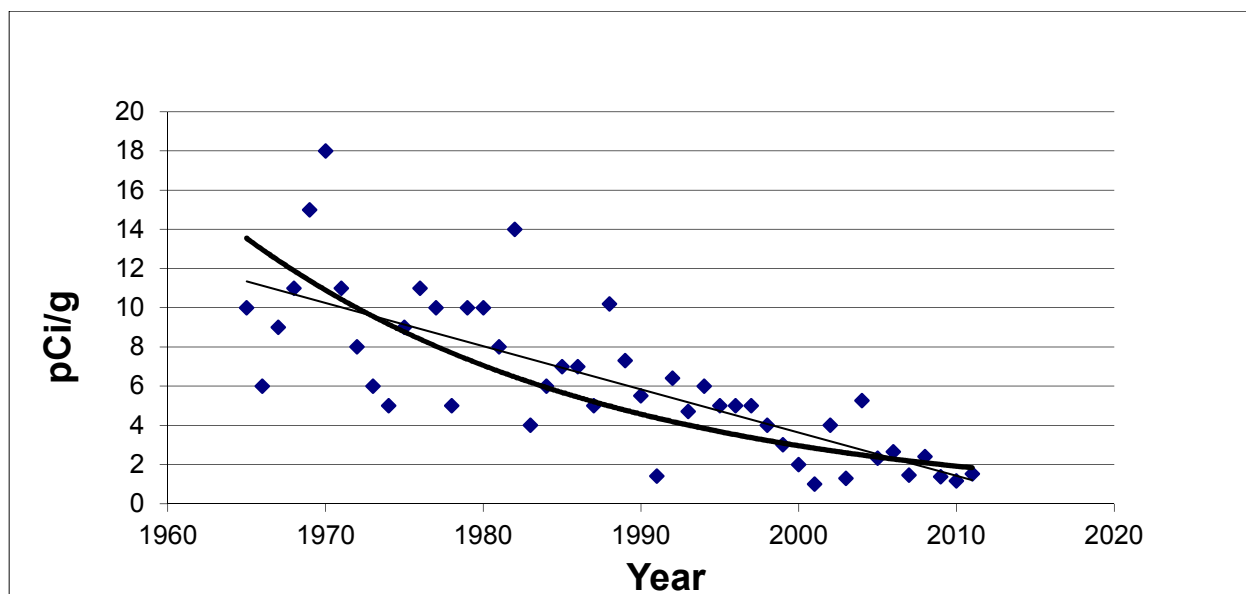


Fig. 7. Mean Cs-137 concentrations (pCi/g) measured in deer during SRS hunts.

1 pCi/g = 0.037 Bq/g

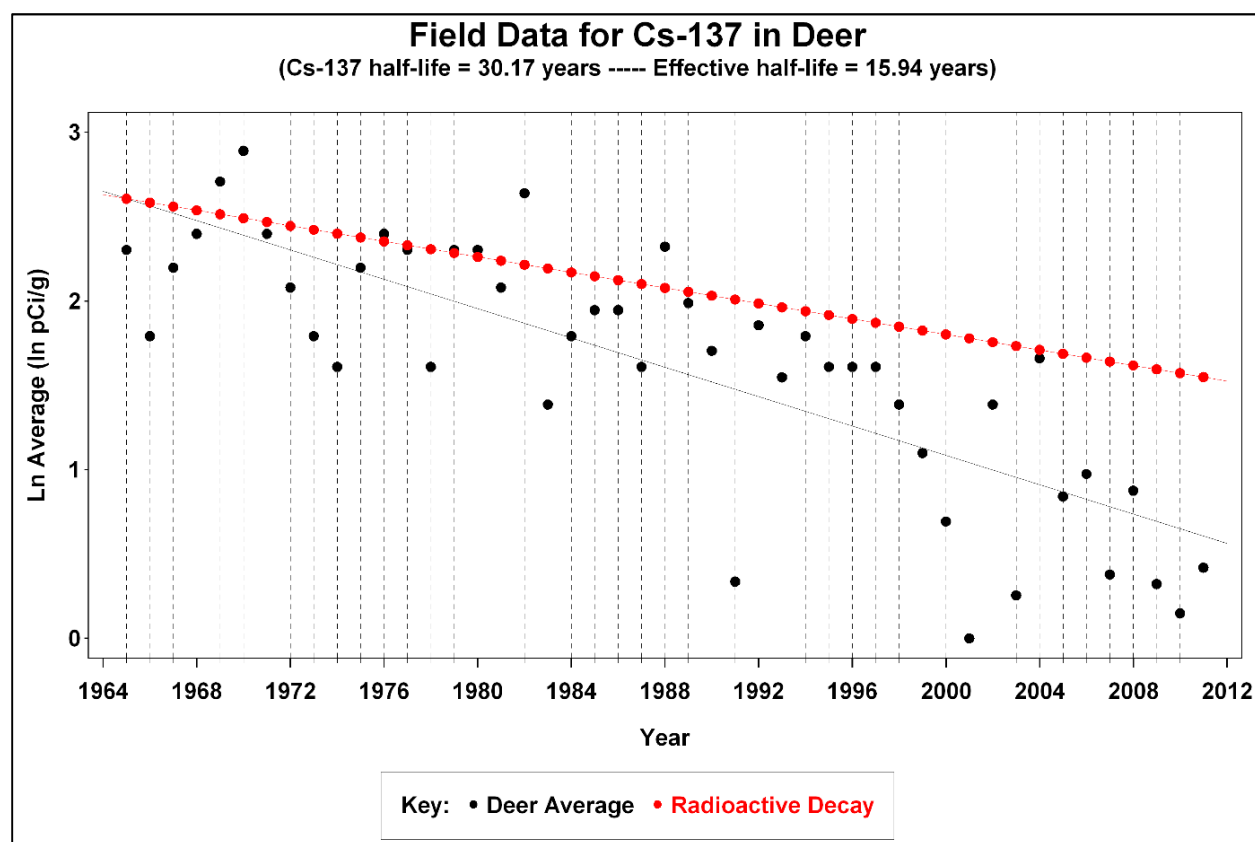


Fig. 8. Ln of the mean Cs-137 concentrations (pCi/g) measured in deer during SRS hunts. Also shown are the regression lines for the data equating to an effective half-life of 15.9 y as compared to the Cs-137 physical half-life of 30.17 y.

## Cs-137 in Fish

Since the 1960's, Cs-137 in fish has been measured in the environs of SRS. Fig. 9 shows the natural log of the mean Cs-137 in fish composites from near the Augusta Lock and Dam, which is more than 25 river-miles upstream of SRS. The maximum mean value measured at this background location was 0.078 TBq/g (2.1 pCi/g), which occurred in 1971 and reflects the effects of global fallout of Cs-137. The values measured over the last 5 y have generally been slightly less than 0.00075 TBq/g (0.02 pCi/g). As shown in Fig. 9, the regression of these data equate to an effective half-life of 7.43 y. This is much shorter than the effective half-life of Cs-137 in soil and vegetation due to larger amounts of dilution and dispersion in the riverine ecosystem.

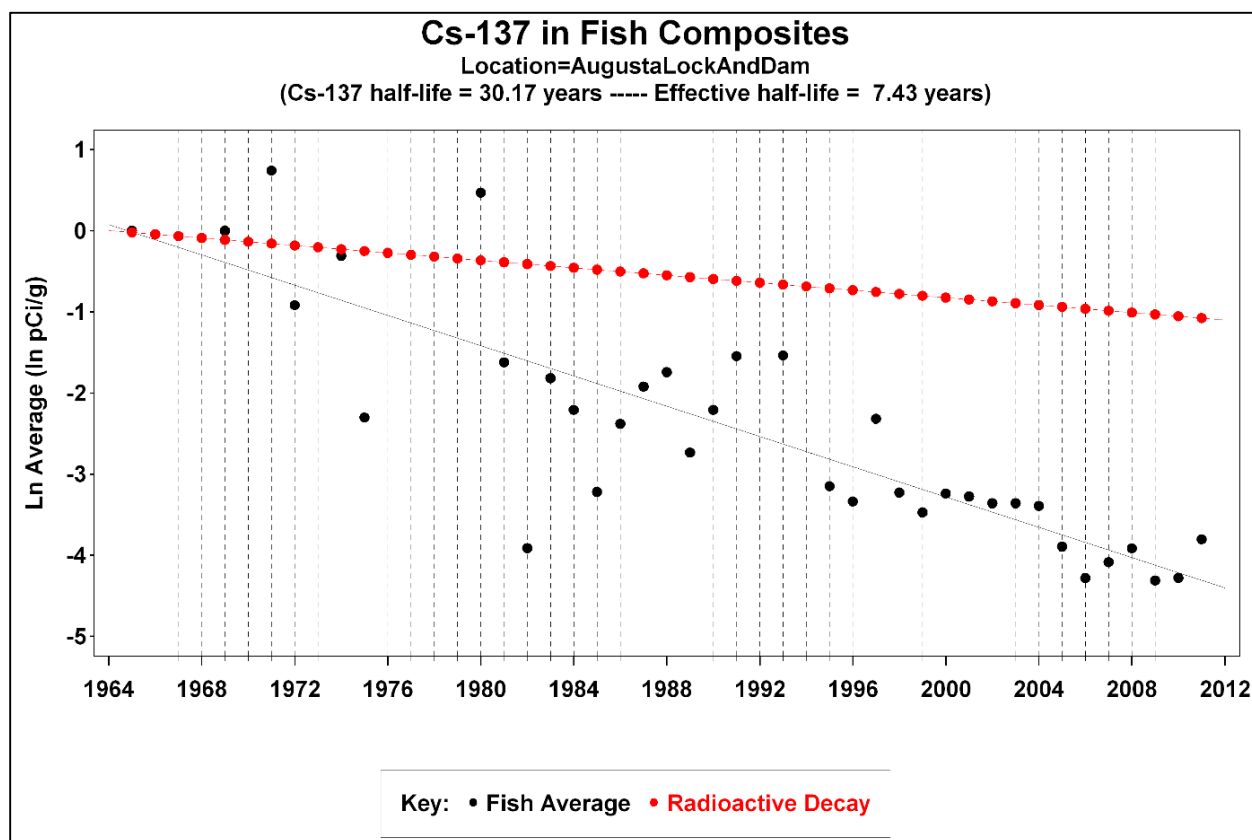


Fig. 9. Ln of the mean Cs-137 concentrations (pCi/g) measured in fish at the Augusta Lock and Dam upriver of SRS. Also shown are the regression lines for the data equating to an effective half-life of 7.43 y as compared to the Cs-137 physical half-life of 30.17 y.

Fig. 10 shows the natural log of the mean Cs-137 in fish composites from near the US Highway 301 Bridge, which is about 10 river-miles downstream of SRS. The maximum mean value measured at this location was 0.296 TBq/g (8.0 pCi/g), which occurred in 1965 and reflects the impact of SRS releases in the early 1960's (Fig. 4). The values measured over the last 5 y have generally been slightly more than 0.00075 TBq/g (0.02 pCi/g).

As shown in Fig. 10, the regression of these data equate to an effective half-life of 6.51 y. This is about the same as the upriver background location and indicates that the potential dose from consumption of fish downriver of SRS has declined more rapidly than expected on the basis of radioactive decay alone.

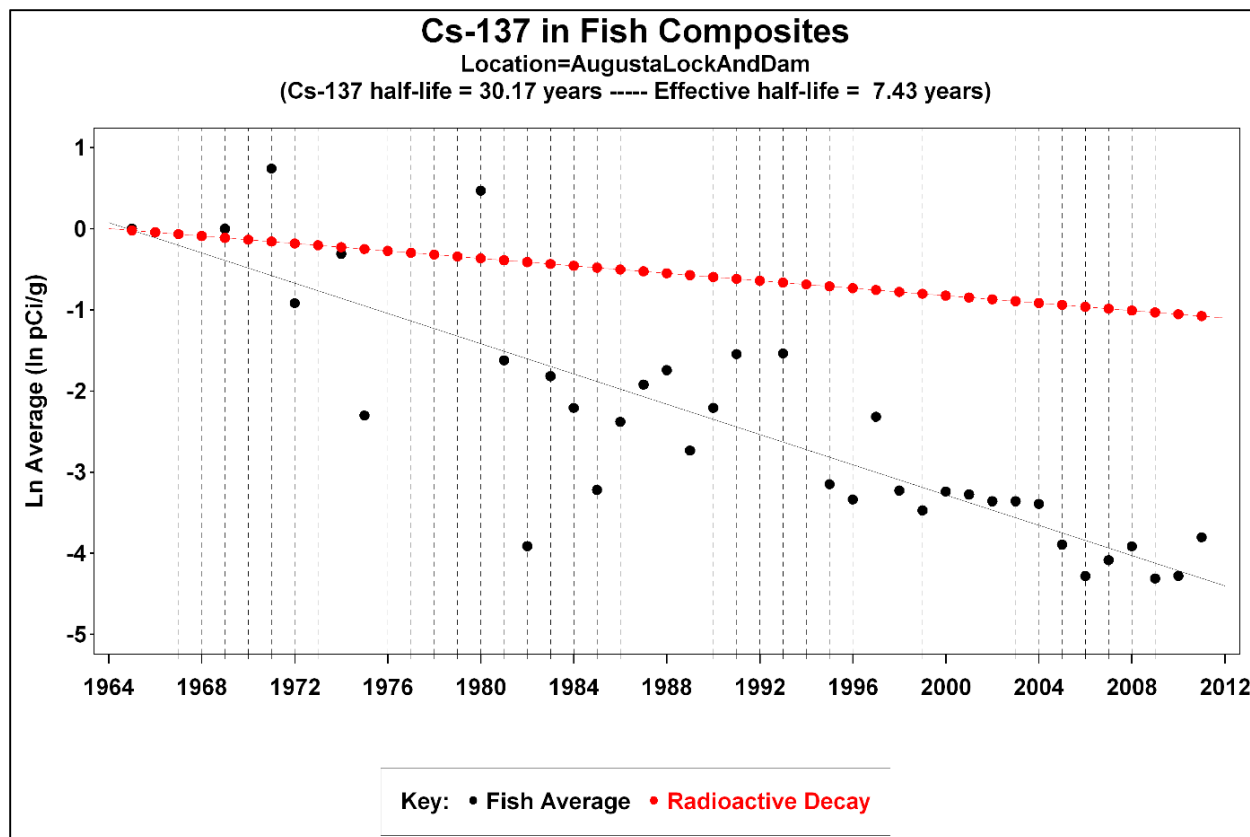


Fig. 10. Ln of the mean Cs-137 concentrations (pCi/g) measured in fish at the Hwy 301 Bridge downriver of SRS. Also shown are the regression lines for the data equating to an effective half-life of 6.51 y as compared to the Cs-137 physical half-life of 30.17 y.

## Tritium in Air

Atmospheric tritium releases at SRS reflected the Site's production of SNM. As shown in Fig. 11, most of the total ( $9.6 \times 10^5$  TBq ( $2.6 \times 10^7$  Ci)) airborne releases of tritium occurred during the height of the Cold War - from 1955 to 1988. These releases were approximately evenly split between the Site's reactor areas and the separations areas [1].

After the shutdown of all SRS reactors in 1988, airborne tritium releases dropped off quickly. The most recent 10 y history of SRS atmospheric tritium releases is provided in Fig. 12. As shown, releases have remained relatively stable reflecting the ongoing tritium processing activities in support of the current US nuclear weapons stockpile.

It is projected that atmospheric tritium releases at SRS will remain about the same as the current levels as long as there is a continuing need for tritium processing.

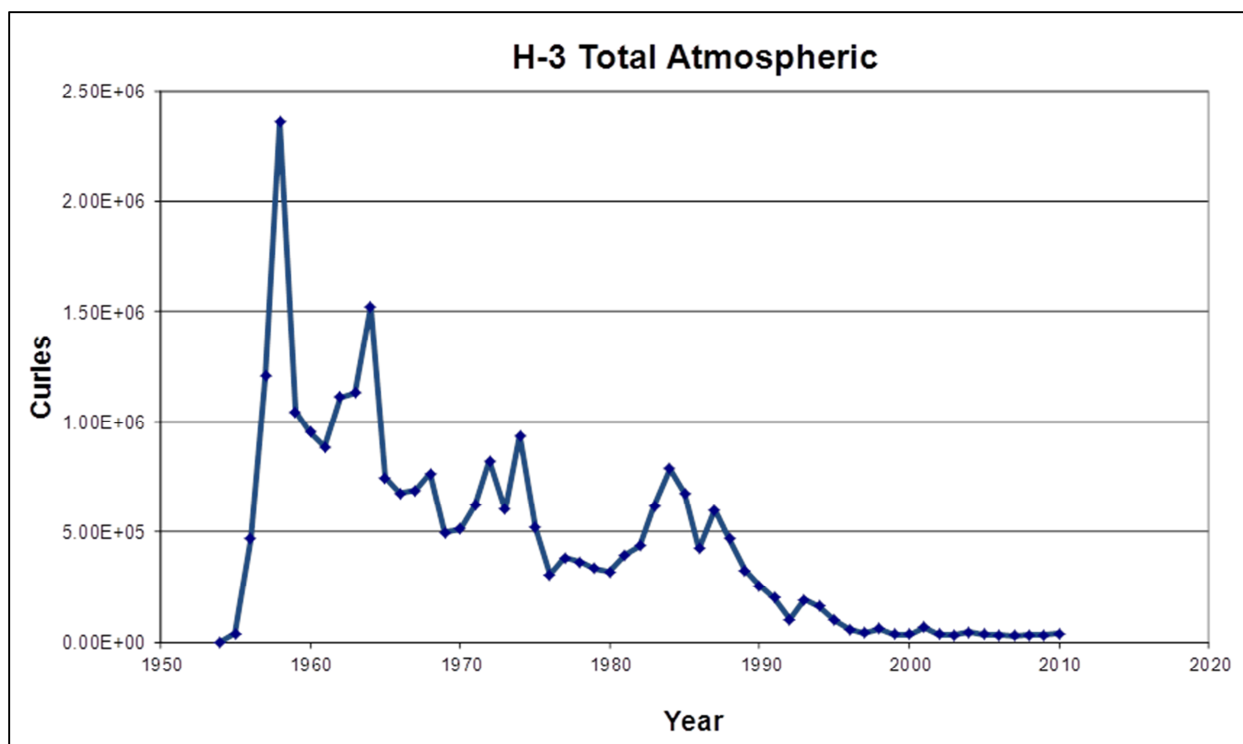


Fig. 11. SRS atmospheric tritium releases (1954- 2010).

1 Ci =  $3.7 \times 10^{10}$  Bq.

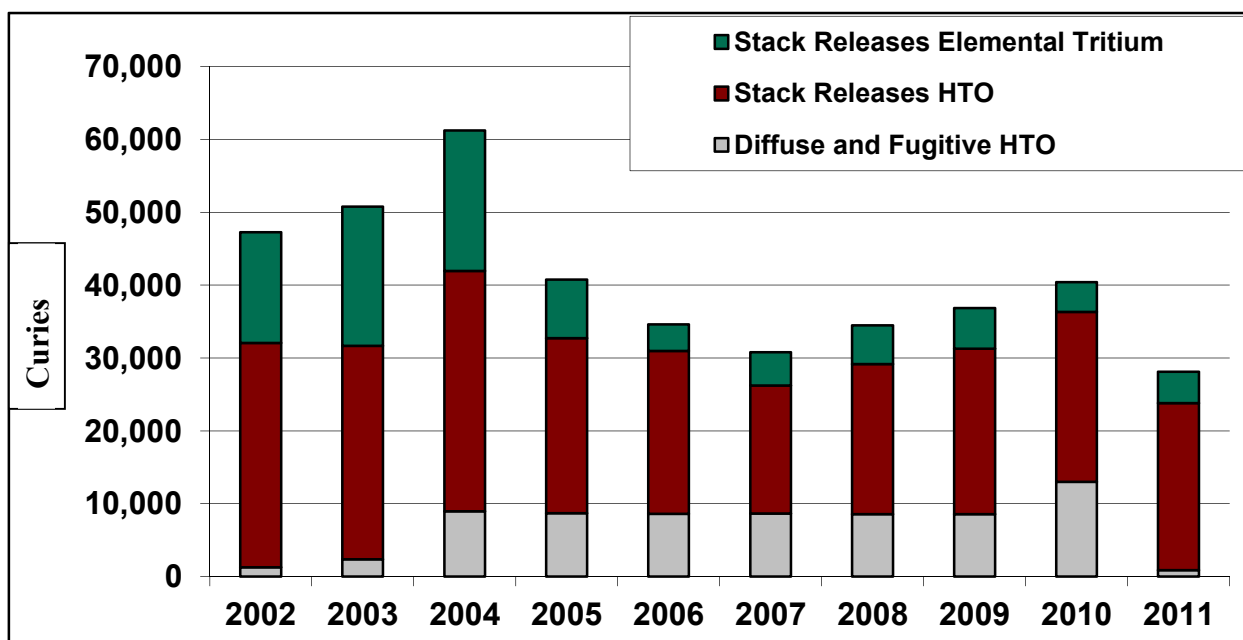


Fig. 12. Ten year history of SRS atmospheric tritium releases. 1 Ci =  $3.7 \times 10^{10}$  Bq.

HTO = tritium oxide

## Tritium in Water

Aqueous tritium releases at SRS also reflected the Site's production of SNM. As shown in Fig. 13, most of the total ( $6.3 \times 10^4$  TBq ( $1.7 \times 10^6$  Ci)) liquid releases of tritium occurred prior to the end of the Cold War in 1988. During this time, an additional  $4.4 \times 10^4$  TBq ( $1.2 \times 10^6$  Ci) were discharged to onsite seepage basins.

Tritium is introduced into SRS streams and the Savannah River from current operational effluents and from legacy discharges to Site seepage basins and waste disposal areas. The legacy discharges migrate into site streams from shallow groundwater and, therefore, are dependent on the amount of rainfall each year. Because of the mobility of tritium in water and the quantities of the radionuclides released during the years of SRS operations, a tritium balance has been performed annually since 1960. SRS tritium releases and transport data for 1960–2010 are depicted in Fig. 13, which shows the history of direct plus migration releases, stream transport, and river transport.

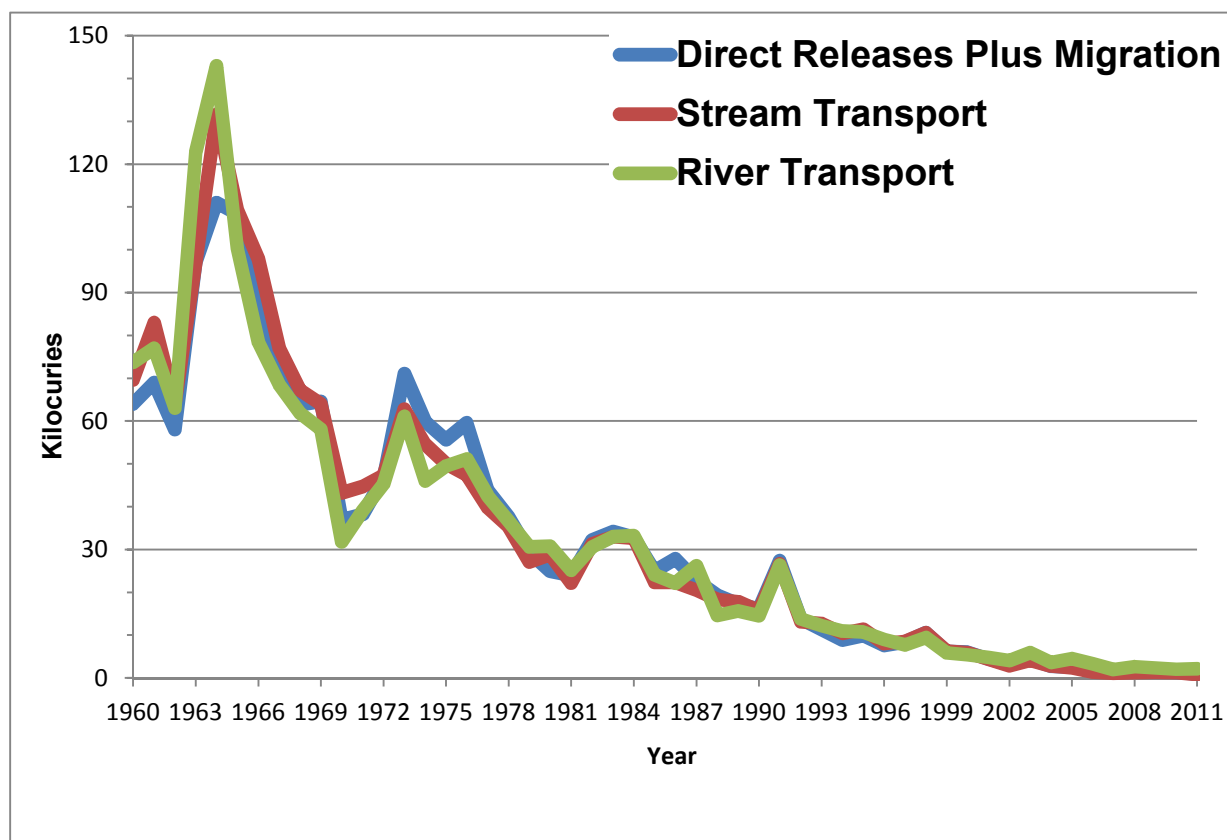


Fig. 13. SRS tritium transport summary, 1960-2011.

1 Kilocurie =  $3.7 \times 10^{13}$  Bq.



The SRS tritium balance includes correlation of:

- total direct tritium releases, including releases from (1) facility effluent discharges and (2) measured migration of tritium from site seepage basins and waste disposal area migration (direct releases plus migration)
- tritium transport in SRS streams, measured at the last sampling point before entry into the Savannah River (stream transport)
- tritium transport in the Savannah River, measured downriver of SRS (near RM 118.8) after subtraction of any measured contribution above the site (river transport)

After the shutdown of all SRS reactors in 1988, the majority (typically over 75%) of the aqueous tritium releases from the Site were legacy migration releases from closed seepage basins and waste disposal areas. The most recent 10 y history of SRS aqueous tritium releases is provided in Fig. 14. As shown, operational releases of tritium have continued to decline, and legacy migration releases average about 80% to 90% of the total.

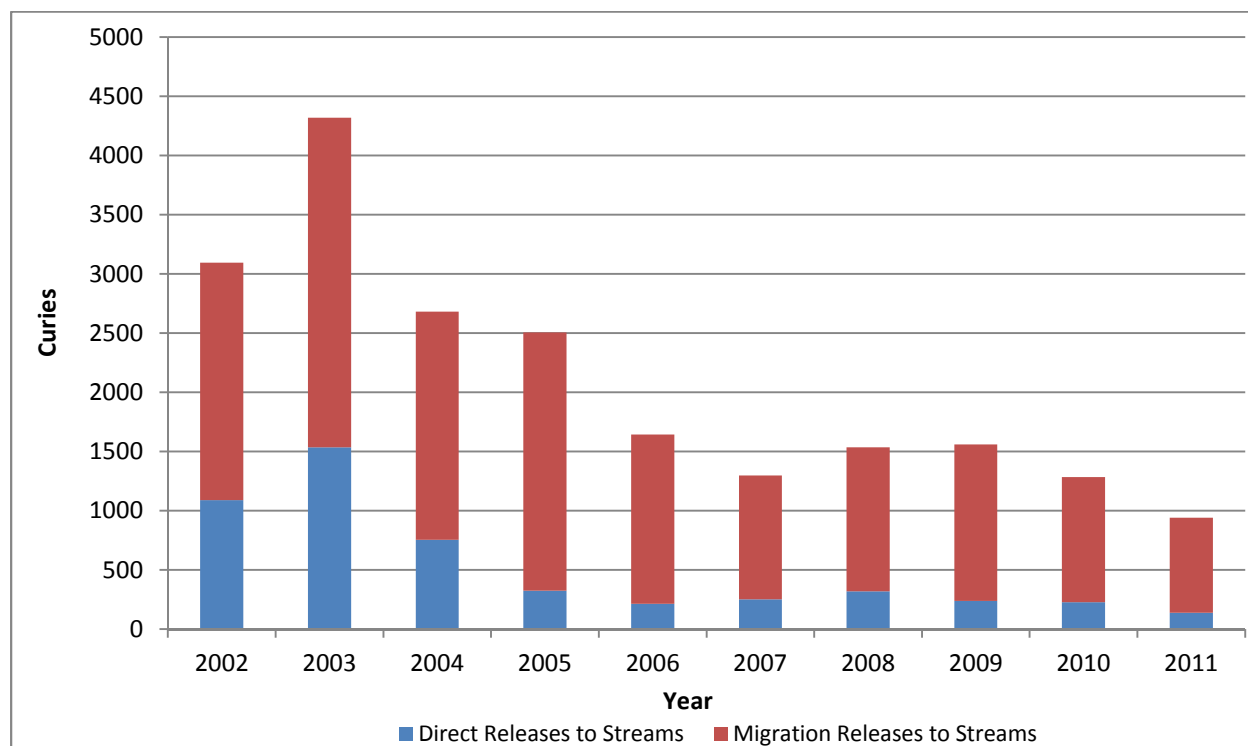


Fig. 14. Ten year history of SRS direct and migration releases of aqueous tritium releases.

1 Ci =  $3.7 \times 10^{10}$  Bq.

Because of the importance of legacy tritium migration discharges, especially from the H- and F-Area seepage basin tritium plumes and the Mixed Waste Management southwest tritium plume, focused attention was placed on environmental remediation efforts to reduce the tritium flux to the onsite stream called Fourmile Branch (FMB). FMB is the SRS stream that these tritium plumes discharge into.

There are no viable removal technologies for tritium in water; however, a RCRA permit goal was established in the 1990's to achieve a 70% reduction in the tritium flux to FMB. Fig. 15 shows the time table and effects of various remediation projects that have been completed to date. In 1997, the H- and F-Area pump and treat projects were started to remove various contaminants, but these had little effect on the tritium flux and were stopped in 2003 and 2005, respectively. In late 2000, a dam was built on a small tributary of FMB (Castor Creek) and an associated phytoremediation project (pine forest irrigation) was started. This project was very effective and it reduced the tritium flux to FMB by about 65%. In 2005, subsurface barriers were completed and in 2008 the capping of the Old Radioactive Waste Burial Ground was completed. These two projects helped to further reduce the tritium flux to FMB so that by 2011, the 70% reduction goal was achieved.

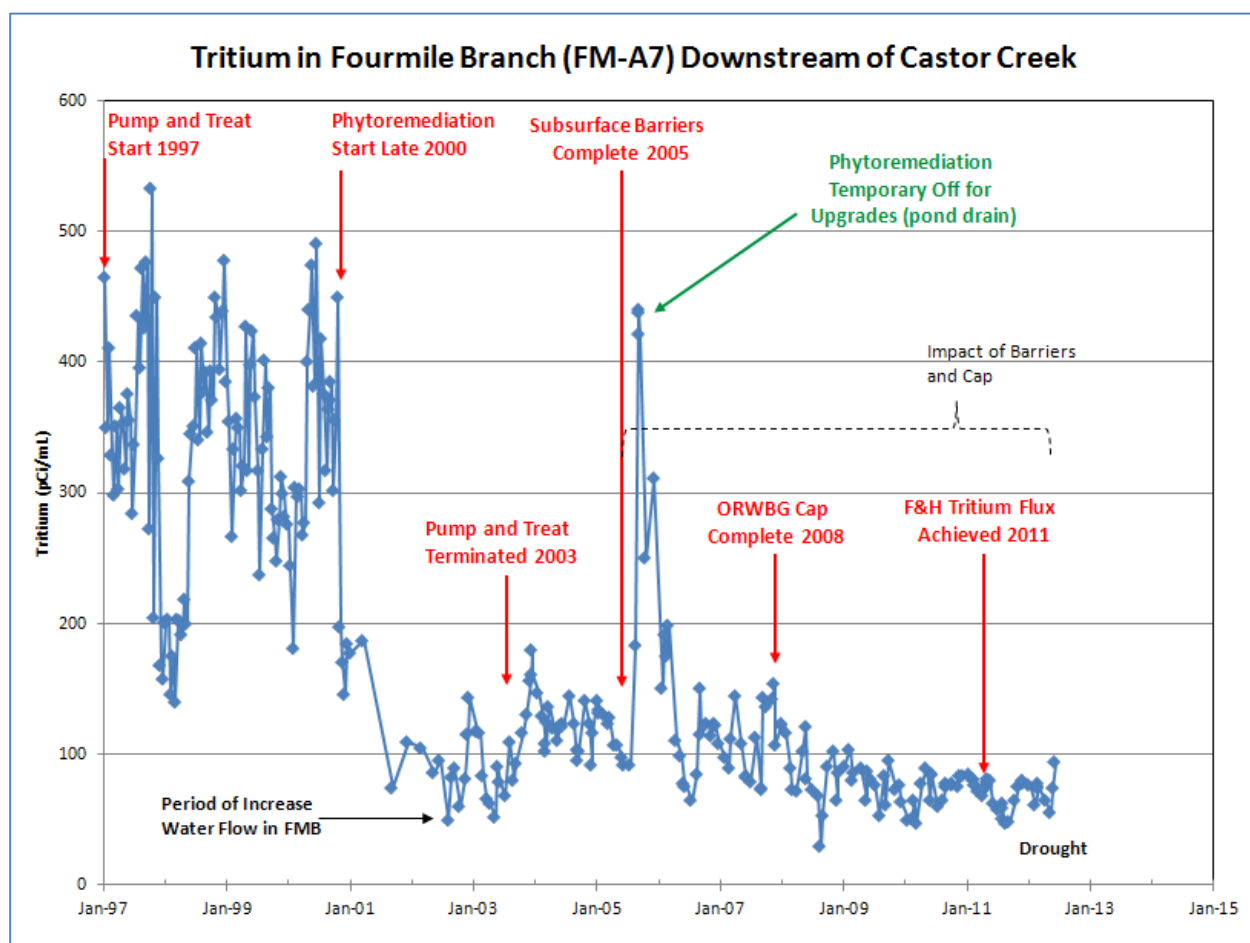


Fig. 15. Tritium concentrations in Fourmile Branch and the effects of various remediation projects. 1 pCi/mL = 0.037 Bq/mL

In Fig. 16, the projected tritium concentrations in FMB are shown. Based on continuation of the current tritium management practices and on radioactive decay (12.3 y half-life), the tritium concentrations in FMB are expected to go below the EPA maximum concentration level (MCL) of 20 pCi/mL (0.74 Bq/mL) by about the year 2037.

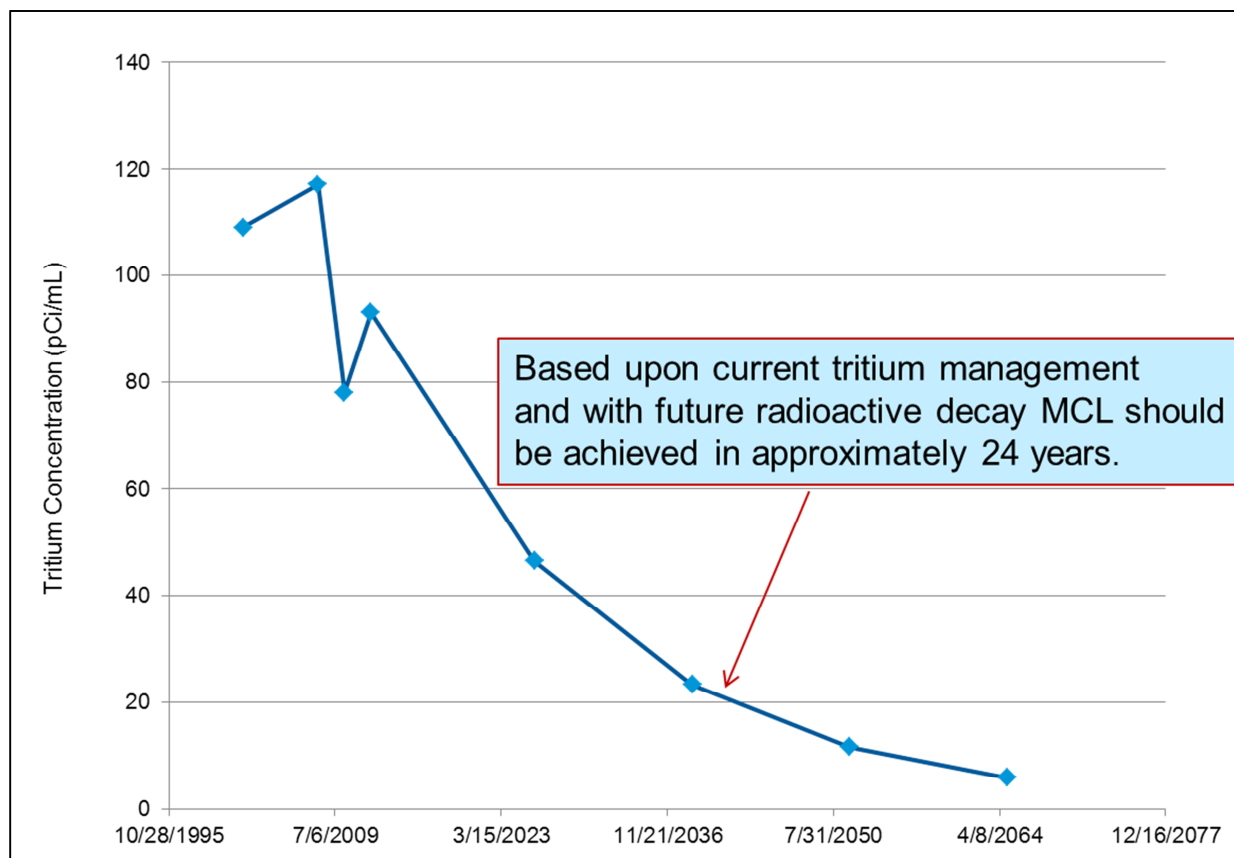


Fig. 16. Projected tritium concentrations in Fourmile Branch.

1 pCi/mL = 0.037 Bq/mL

## CONCLUSIONS

During the operational history of SRS, radiological releases to the environment have been directly related to the production of SNM with a majority of the releases occurring prior to the end of the Cold War in 1991. A major source of Cs-137 on SRS was (and remains to be) global fallout from above ground testing of nuclear weapons.

In the SRS environs, Cs-137 in soil, vegetation, and deer has shown an “effective” half-life of about 14 – 16 y, which is about one half the Cs-137 physical half-life of 30.17 y. The effective half-life of Cs-137 in fish has been even shorter at about 6 - 7 y. However, as shown in the various graphs depicting long-term trends, the effects of environmental dispersion and dilution

are expected to continue to slow down and the observed effective half-life of Cs-137 will begin to approach its physical half-life.

Atmospheric tritium releases at SRS dropped off quickly after all Site reactors were shutdown in 1988. However, they will continue into the future and remain relatively stable reflecting the Site's ongoing tritium processing activities in support of the current US nuclear weapons stockpile.

Since 1991, the majority of aqueous tritium releases at SRS has been legacy discharges to site streams from onsite seepage basins and waste management areas. However, focused efforts to reduce the tritium flux in FMB have been successful. It is now projected that the tritium concentrations in FMB will go below the EPA MCL of 20 pCi/mL (0.74 Bq/mL) in about 24 y.

## REFERENCES

1. G.T. JANNIK AND R. SCHEFFLER, *Critical Radionuclide and Pathway Analysis for the Savannah River Site*, SRNL-STI-2011-0503, Savannah River National Laboratory, Aiken, SC (2011).
2. M. EISENBUD, *Environmental Radioactivity*, Academic Press, pp272 and 280, (1987).
3. CDC/NCI 2002: *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, August (2001).
4. K.F. GAINES AND 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 (August 2011).
5. M.H. PALLER, P.D. FLEDDERMAN, AND G.T. JANNIK, *Changes in Cs-137 Concentrations in Soil and Vegetation on the Floodplain of the Savannah River over a 30 Year Period*. *Journal of Environmental Radioactivity*, doi:10.1016/j.envrad.2008.04.001 (2008).

## ACKNOWLEDGEMENTS

This manuscript has been co-authored by Savannah River Nuclear Solutions, LLC under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting this article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this work, or allow others to do so, for United States Government purposes.