CHAPTER 5

RELEASES OF RADIONUCLIDES TO SURFACE WATER

ABSTRACT

The five production reactors were the source of the majority of radionuclide releases to surface water from the Savannah River Site (SRS), primarily because most surface water releases came from the disassembly basins in the reactors areas. Releases of liquid effluents from the separations areas were discharged into storage tanks and seepage basins and not directly to the streams. Tritium and ^{137}Cs are the main radionuclides of concern from past releases to surface streams that eventually reached the Savannah River, and detailed source terms have been calculated for those radionuclides. Other radionuclides that are important depending upon the exposure pathways include ^{90}Sr, ^{131}I, the activation products, ^{60}Co, ^{32}P, and ^{65}Zn, and uranium releases from the M-Area. Surface water releases of radionuclides were highest in the early to middle 1960s and decreased into the 1980s. Our median estimate of release of tritium for all years (1952 through 1992) is 1.8 million Ci, with the 5th and 95th percentiles of the distribution of 1.3 million and 2.5 million, respectively. The median estimate of the total ^{137}Cs for all years is about 250 Ci with the 5th and 95th percentiles of the distribution of 100 and 600 Ci ^{137}Cs. The median estimate of the total ^{90}Sr for all years is about 100 Ci with the 5th and 95th percentiles of the distribution of 45 and 250 Ci ^{90}Sr. There is overall general agreement between our reconstructed release estimates to the Site streams based on original measurement data, supported by weekly and monthly reports from the 1960s and 1970s, and the annual total reported by SRS. There is more uncertainty associated with release estimates in the 1950s and 1960s than in later years because of improvements in monitoring capabilities and techniques and in methods to prevent the release of materials from the reactors and processing facilities.

INTRODUCTION

A general overview of the key operational areas at the SRS, and important liquid effluent release points may be helpful in understanding the sources of information and methods used for reconstructing the releases of key radionuclides released in liquid effluents from the SRS in the past. A detailed history of the SRS Site and processes can be found in a report produced during Phase I (Meyer et al. 1995), and a summary of that history in Chapter 2 of this current report. Some of the areas onsite that are important contributors to radionuclides in liquid effluents are reactor areas, separations areas, fuel fabrication area (M-Area), heavy water reprocessing (D-Area), and administration area (A-Area).

Reactor Areas

Five heavy water production reactors, called R, P, K, L, and C Reactors, operated at the Site. They were all constructed in the early 1950s and are located near the center of the Site in separate areas designated R-Area, P-Area, K-Area, L-Area, and C-Area and are separated by at least 2.5 mi. The operation periods for the reactors were: C Reactor 1954-1987, K Reactor 1954-1988, L Reactor 1954-1968 and 1985-1988, P Reactor 1954-1988, and R Reactor 1954-1964. A restart of

the K Reactor began in 1991. Plutonium and tritium—the primary products of the SRS reactors—are created by uranium and lithium absorption of neutrons. Tritium is also by ternary fission of transuranic elements in the reactor fuels and targets. The controlled fission process within the reactors produced enormous amounts of energy. These are heavy-water moderated reactors, which means that heavy water is circulated in a closed system through heat exchangers to moderate and cool the reactors. Water from the Savannah River and Par Pond provided the cooling water for the heat exchangers. The heavy water in the heat exchangers was cooled by ordinary water from the Savannah River. The secondary cooling water from the Savannah River was returned to the river through the Site streams. The river water did not come into direct contact with the reactor core but was used to cool the heavy water that served as a moderator and primary coolant. This secondary cooling water was stored in holding basins at each reactor area, passed through the heat exchangers as needed and then discharged as wastewater to the Site streams. In 1958, Par Pond, a 2,700-acre human-made lake, began providing cooling water from the P and R Reactors. The pond water was pumped to the P and R Reactors and returned to Par Pond, which allowed more river water to be pumped to L, K, and C Reactors. In 1985, L Lake was formed by damming Steel Creek to provide cooling water for the L Reactor to comply with the permit to restart the L Reactor. Before Par Pond and L-Lake, the water was discharged directly to Lower Three Runs Creek (from P and R Reactors), to Steel Creek (from L Reactor), to Pen Branch (from K Reactor), or to Four Mile Creek (from C Reactor).

The Reactor Support Facility for each reactor consisted of an Assembly Area, Disassembly Area and Purification Area (Figure 5-1) (Bauer 1986). In the Disassembly Area, depleted fuel and irradiated materials were stored for a time to allow short-lived fission products to decay and then were disassembled and loaded into casks. All disassembly operations were performed remotely under water. Materials were transferred sequentially through the four sections of the basin through narrow vertical gateways between sections. Each basin contained four main sections:

1. Vertical tube storage (VTS) assemblies suspended by an overhead monorail system for cooling.
2. Disassembly basin, where slugs from target assemblies containing plutonium were placed in buckets for shipment to F Canyon, and fuel assemblies were bundled for shipment to H Canyon.
3. Horizontal and bucket storage, where buckets and bundles were stored before transfer.
4. Transfer bay, where materials were loaded into shipping casks for transport to the separations areas.

The main source of radionuclide releases to surface water from the reactors was from the VTS and disassembly basins. Spent fuel and irradiated target elements were removed from the reactor and stored in these large water-filled basins adjacent to the reactor building. The water in the basins cooled the components and provided shielding for the radioactive components. During the years of peak operations (1950s and 1960s), materials remained in the disassembly basins for less than nine months, but the storage period was longer than nine months in later years. The 17 to 30 foot-deep basins had 3-ft thick concrete walls and held 3.5 million gal of water in C-Area, L-Area, and K-Area and about 4.8 million gal in the P-Area and R-Area.

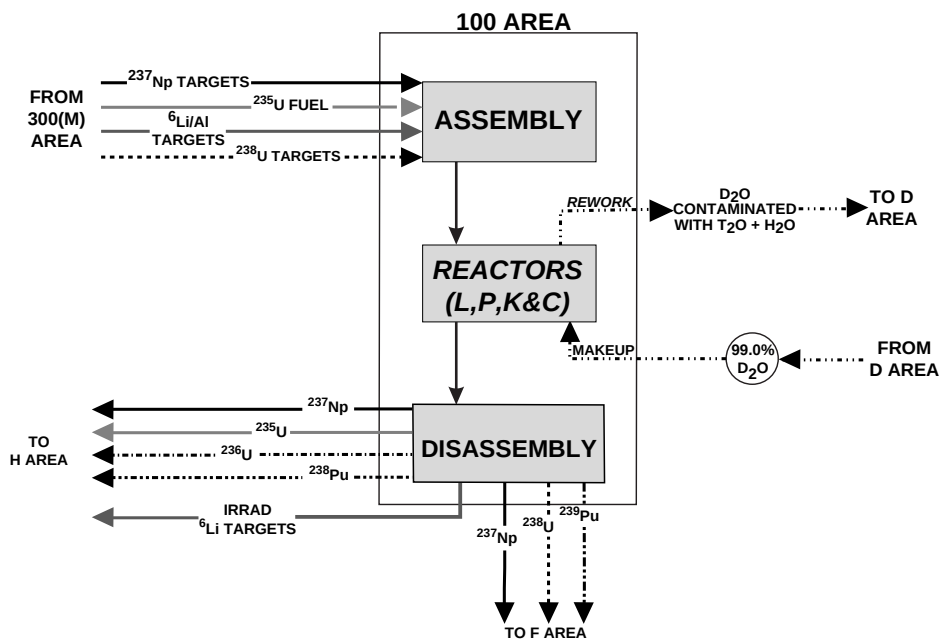

Figure 5-1. Materials flow diagram for the reactor (100) areas at the SRS. The area is comprised of the reactors as well as an assembly area, which receives fuel and target materials from the 300-M Area, and the disassembly area, which contains the vertical tube storage and disassembly basins that sent products, irradiated fuel, and irradiated targets to the F-Area and H-Area. The moderator for the reactor, deuterium oxide (D_2O), comes from the Heavy Water Plant in D-Area, and was returned to the D-Area for processing when the heavy water became degraded with tritium oxide (T_2O) and light or natural (H_2O) water. (Redrawn from Bauer 1986).

From startup to the mid-1960s, visual clarity was maintained in the basins by continuously purging them with fresh, filtered river water. The basin purge water was discharged to the Site streams along with the secondary cooling water. In the mid-1960s filters were added to the VTS basins. Sand filters were added to the disassembly basins in the early 1970s and continuous basin purging to the Site streams was discontinued (Murphy et al. 1991). However, periodic purges continued directly to the Site streams from 1970 to 1977 to reduce the tritium exposures to operating personnel because the sand filters did not remove tritium. Finally, in 1978, the purge water from the fuel storage basins was discharged to the seepage basins (Reinig et al. 1973, Towler 1980).

Separations Areas

Products produced in the reactors were separated chemically in the F-Area and H-Area separations area, located near the center of the SRS. Complex chemical and physical processes in the F-Canyon and H-Canyon Buildings, 200-F and 200-H, separated uranium, plutonium, and fission products. Major operational milestones for both the F-Area and H-Area were increases in the processing capacities throughout the 1950s and 1960s. Both the F-Area and H-Area were operated throughout the 1980s and 1990s. The separated plutonium and uranium were then

transferred to other facilities in the F-Area and H-Area and processed into solid forms. Fission products were stored in high-level waste tanks in the separations areas. Originally, the tritium received in the separations areas was a by-product of plutonium production. By 1955, greater production of tritium was needed, and a second tritium production line became operational in 1957. Cooling water for portions of the F-Area separations process line was pumped from deep wells and discharged to Four Mile Creek after use. At times, this water contained measurable amounts of radioactivity because of the occurrence of cooling coil leaks. Cooling coils are used in the head end dissolvers, which contain a mixture of many fission and activation products. Uranium is a predominant chemical constituent of this mixture (Evans et al. 1992). The F-Area liquid effluent consists of process cooling water, sanitary wastewater treatment effluents, and spill runoff, similar to the H-Area effluent.

The H and F facilities each had their own waste tanks, seepage basins and retention basins, but they shared a common burial ground (643-G). For processes in both 221-H and 221-F, liquid high activity wastes were evaporated to recover nitric acid and reduce volume, neutralized with sodium hydroxide and sent to the Building 241 tanks for storage. Radioactive wastes were transferred from both buildings in four stainless steel waste headers along the east side of the buildings in pipes that ran at a gradual slope within a hollow concrete encasement to the Building 241 waste tanks. If actinides were present, the evaporated high activity concentrate may have been run through the ion exchange primary recovery column and frames in the separations area to recover plutonium and neptunium before being sent to the waste tanks.

Most of the liquid low activity waste was concentrated by evaporation, neutralized with sodium hydroxide and stored in the Building 241 tanks. Equipment removed from the canyon was decontaminated by jetted detergent and chemicals before repair. Equipment to be discarded was often cleaned first, in a special cell called the swimming pool, to be classified as low activity waste. Solid waste from most of the facilities was trucked to the burial ground (Building 643-G) near the F-Area and H-Area (Figure 5-2). Solid waste was shipped from the hot canyon by rail.

Some of the tritium releases from the separations area originated as tritium-contaminated air. In 1973, a special study was conducted in 200-H during November and December to determine the source of the apparent tritium contamination in the 221-H cooling water systems. Results suggested that more than 90% of the tritium released to the streams from the 221-H cooling water systems could originate as tritium-contaminated air near 285-H, which was “cleaned” as it passed (or was drawn) through the cascading water in the cooling tower (Epting 1974). Sampling stations, set up near the 285-H cooling water tower, were designed to measure the influence of airborne tritium near the tower on a covered container of clean, deionized water. The station also collected tritium-contaminated rainwater. Airborne tritium concentrations in the vicinity of the cooling water tower were determined using the silica gel moisture collection method (see Chapter 4.1). Results showed that there was sufficient ambient airborne tritium to account for an average of 3 Ci mo^{-1} (and higher during periods of high stack releases from the tritium facilities) of releases to the stream via cooling water releases. There was an apparent direct relationship between tritium air activity near the 285-H tower and tritium concentrations in the tower cooling water. It was noted that less than 10% of the tritium releases via 221-H cooling water were thought to be due to canyon building processes (Epting 1974).

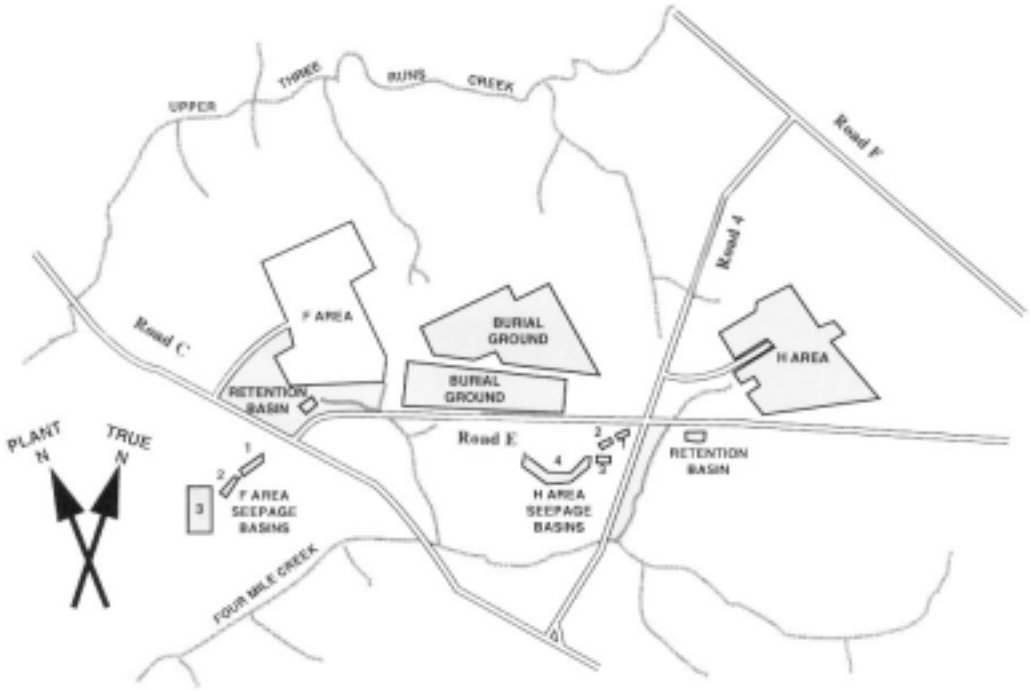

Figure 5-2. A diagram showing one location of waste areas at the SRS—the radioactive burial grounds, and seepage and retention basins in the separations area at the SRS. (Redrawn from Ice 1971).

Fuel Fabrication Area, M-Area

The fuel fabrication facilities produced fuel and target elements from aluminum and uranium that were then sent to the 100-Area production reactors. There was also a treatment facility for radioactive liquid wastes in the M-Area. See Meyer et al (1995) and Chapter 2 for a thorough description of the fuel fabrication area. Over time, changes in the fuel and targets were made, new and different fuels were tested, and more efficient and productive fuels were designed (Pelfrey 1987; Pickett 1997). The reactor assemblies became increasingly complex as different products were desired and more efficient cooling means were developed.

Of the radioactivity released from this area, uranium dominated other radionuclides in liquid effluents (Horton 1955b). The largest releases of uranium onsite occurred from the M-Area to Tim's Branch and to the seepage basins after the seepage basins were put into service in 1973. The main sources of uranium in liquid effluents from this area were from:

- Uranium in etching solutions—The uranium metal was electrolytically plated with nickel and then clad, or covered, to protect it from contact with the reactor coolant because uranium corrodes rapidly in water. The moderator and coolant used at the SRS was heavy water. The cladding also served as a barrier to keep fission products from contacting the water. For the aluminum cover or cladding to bond to the

uranium, the uranium target slugs had to be etched with acid. As a result, the etching solution contained up to 300 g L^{-1} of uranium (Evans et al. 1992).

- Ruptured aluminum seals on uranium slugs—After cladding, the aluminum seals on the uranium slugs were tested by heating the slugs under pressure in an autoclave. If there were leaks in the cladding, slug rupture occurred and those slugs could then be rejected for use. After rupture, the autoclave had to be washed out, and particulate uranium oxide was present in this wash-down water of the autoclave.
- Chemical removal of the aluminum and nickel cladding from the rejected slugs, using nitric acid—When a uranium target slug was rejected, the aluminum and nickel cladding was removed using nitric acid. Even though some of this dissolved material was precipitated and filtered to remove the suspended solids, much of it was carried along in the waste liquids (Evans et al. 1992).

Heavy Water Reprocessing, or D-Area

A heavy water production plant, in D-Area, began operation in 1953 to concentrate heavy water from Savannah River water to moderate and cool the Site's reactors. Heavy water was produced in the Heavy Water Plant, 400-D, from October 1952 to 1982. The Heavy Water Plant extracted deuterium oxide, D_2O from Savannah River water by dual temperature exchange using hydrogen sulfide (H_2S). Natural river water contains D_2O at a concentration of about 150 ppm, or 0.015%. The Dana Plant, a facility in Dana, Indiana, was a precursor to the SRS facility and produced heavy water by the GS process for the SRS (Bebbington 1990). The facility stopped production in 1981 because there was a sufficient supply of heavy water. A heavy water reprocessing facility, a coal-fired power plant, and a laboratory that analyzes process effluent samples are also located in the D-Area (See Meyer et al. 1995 and Chapter 2).

Administration Area, or A-Area

The A (Administration) Area contained organizations that provided direct support for SRS operations. Generally, the 700-numbered buildings were offices, laundries, fire stations, maintenance buildings, research laboratories, with a lower potential for offsite release of hazardous materials than the Reactor Areas or separations areas. The Savannah River Laboratory (SRL), or Savannah River Technology Center (SRTC) as it is now called, are located here as well as the University of Georgia's Savannah River Ecology Laboratory (SREL).

Although not located in the A-Area, the TNX and CMX Semiworks were considered to be a part of the SRL and were some of the first facilities to operate at the SRS. CMX and TNX were code designations and had no logical derivation according to Bebbington (1990). CMX investigated problems associated with using Savannah River water for cooling, and it housed the river water pumps (water treatment took place at the reactors), a pressure facility for testing of reactor elements, and a hydraulic test facility. CMX was shut down in 1984. The TNX facility, one of the first facilities to operate at the Site, provided technical support, pilot data, and personnel training. In later years, the facility was involved in waste processing research and development. By the end of the first 5 years, all of the basic production facilities were in operation and the products, plutonium metal and tritium gas, were being delivered.

GENERAL HYDROLOGY

The Savannah River is the principal river in the area with an average velocity of about 3 to 5 mi hr^{-1} (MPH) or 4.4–7.3 ft s^{-1} and average flow in 11,000 cubic feet per second (cfs) (Ice 1971). The water table at the Site varies from ground surface to slightly more than 100 ft deep. Groundwater in the area occurs as perched water tables, normal water tables and artesian aquifers (Ice 1971). The principal aquifer is the Tuscaloosa formation, with a potential water yield ranging from 30,000 to 400,000 gal per day per foot. In the chemical separations areas (F-Area and H-Area), the groundwater velocity ranges from a few hundredths of a foot per day to nearly 1 ft d^{-1} near flowing streams. Both solids and liquid wastes have been stored at or just beneath ground level during much of its operational history. Thus, the local hydrology and geology play an important role in assessing the releases via surface water.

Onsite Streams

Both chemicals and radionuclides in liquid effluents from the SRS facilities are released either directly into onsite streams or first into seepage basins and then into onsite streams. In the early years of operations, most liquid waste was discharged to streams or swampy areas. With time, seepage basins, settling ponds, tanks, and retention basins were used. Contaminants from these often entered groundwater then migrated to the nearest downgradient surface water.

Of the radionuclides that reach the water table, tritium is the most important because of its high mobility and abundance. Low concentrations of tritium are present in liquid waste streams and burial ground leachates as tritiated water. The tritiated water moves with the groundwater, some of which outcrops to surface streams. A study in 1983 (Christensen and Gordon 1983) reported that the waste sites that are the principal contributors of tritium from shallow groundwater to surface water at the SRS are

- K-Area containment basin—about 10,000 Ci y^{-1} outcrops to a tributary of Pen Branch
- H-Area seepage basins—about 7,000 Ci y^{-1} outcrops to Four Mile Creek and tributaries
- F-Area seepage basins—about 2,000 Ci y^{-1} outcrops to Four Mile Creek
- Radioactive burial grounds—an estimated 200 Ci y^{-1} outcrops to a tributary of Four Mile Creek.

Almost all of the SRS is drained by five main streams: Upper Three Runs Creek, Four Mile Creek, Pen Branch, Steel Creek and Lower Three Runs Creek. The onsite streams meander for 8–13 km (5–8 mi) onsite, through the Savannah River swamp system before eventually discharging offsite into the Savannah River (Westinghouse 1996). One smaller stream in the northeast area of the Site drains to the Salkehatchie River rather than the Savannah River.

The following section describes each of the major onsite streams and close-up maps for the five Site streams show the drainage patterns of various facilities to specific streams. These diagrams show that Upper Three Runs received runoff from the storm sewer from the north side of F-Area and seepage from the F-Area ash basin. At Road C, Tim's Branch discharges industrial, laboratory, water treatment, and sanitary wastewater treatment effluents from A-Area and M-

Area to Upper Three Runs. Steed's Pond, located on Tim's Branch just north of Road 2, has served as a mixing and settling pond for A-Area and M-Area wastes. The creek merges with the C-Reactor cooling water about 0.5 mi south of Road 3. The operating reactor cooling water flow is much greater than that in the receiving stream, and as a consequence, materials already in Four Mile Creek are greatly diluted. This may be advantageous from the standpoint of release concentration limits, but it makes analyses more difficult.

The onsite streams of particular importance for carrying reactor effluents are Steel Creek and Lower Three Runs Creek. Steel Creek received discharges from the L and P Reactor Areas in the early years before Par Pond was constructed and filled in late 1958. After that, the P Reactor cooling water was released to a canal for recirculation in Par Pond. The K Reactor effluent from Pen Branch was discharged to Steel Creek via the Savannah River swamp through one major and one or more minor streams about 0.25 mi above Steel Creek mouth. Lower Three Runs Creek received some discharge from the R Reactor area before 1958 when Par Pond opened. The overflow from Par Pond to Lower Three Runs carried runoff from the northeast portion of the plant, sewage treatment effluent, water treatment chemicals added to P Reactor cooling water, and any materials leaking to the cooling water.

Significant modifications to creek drainage patterns, channel depth, and surrounding vegetation configurations have occurred over the years as a result of SRS discharges. The streams most heavily affected were Steel Creek, Pen Branch, and Four Mile Creek where small deltas have been deposited at their creek mouths where they enter the Savannah River swamp system or flood plain (Ruby et al. 1981). These sedimentation deltas have resulted from erosion of stream banks because the stream channels have carried many times their natural flows. For example, the natural flow in Steel Creek of about 35 cfs increased roughly ten-fold (250-400 cfs) when effluents from the L and P Reactors were discharged into Steel Creek in the late 1950s and early 1960s.

The Savannah River swamp system borders the Savannah River for about 16 km (10 mi) and averages about 2.4 km (1.5 mi) wide. About 10,000 acres of the SRS forest lie on the Site from Upper Three Runs Creek to Steel Creek. The Savannah River flow records its highest levels in the winter and spring, and lowest levels in the summer and fall. The river historically overflows its channel and floods the swamps bordering the Site when its elevation rises higher than 27 m above mean sea level at the SRS Boat Dock. This level corresponds to flows equal to or greater than about 438 $m^3\ s^{-1}$. Studies have indicated that the swamp was flooded approximately 20% (74 days per year on the average) during the period 1958 to 1967 (Westinghouse 1996). Clearly, the onsite streams play an important role in receiving, transporting, removing and diluting materials before they are released offsite in the Savannah River.

Upper Three Runs Creek

This creek is the only SRS stream with headwaters arising offsite, north of the Savannah River Plant (Figure 5-3). This stream and its major tributaries, Tinker Creek and Tim's Branch, drain an area of about 545 km^2 (Evans et al. 1992). Upper Three Runs has the largest natural flow of any plant stream. The average flow of 257 $ft^3\ s^{-1}$ is slightly increased by a Site contribution of less than 10 $ft^3\ s^{-1}$. Before the point where the creeks converge, the streams received no plant process discharges prior to the construction of the Effluent Treatment Facility (ETF) in 1988 but

are subject to runoff of herbicides from activities at the U.S. Forest Service-Savannah River Environmental Laboratory facilities, and part of the H-Area storm sewer system.

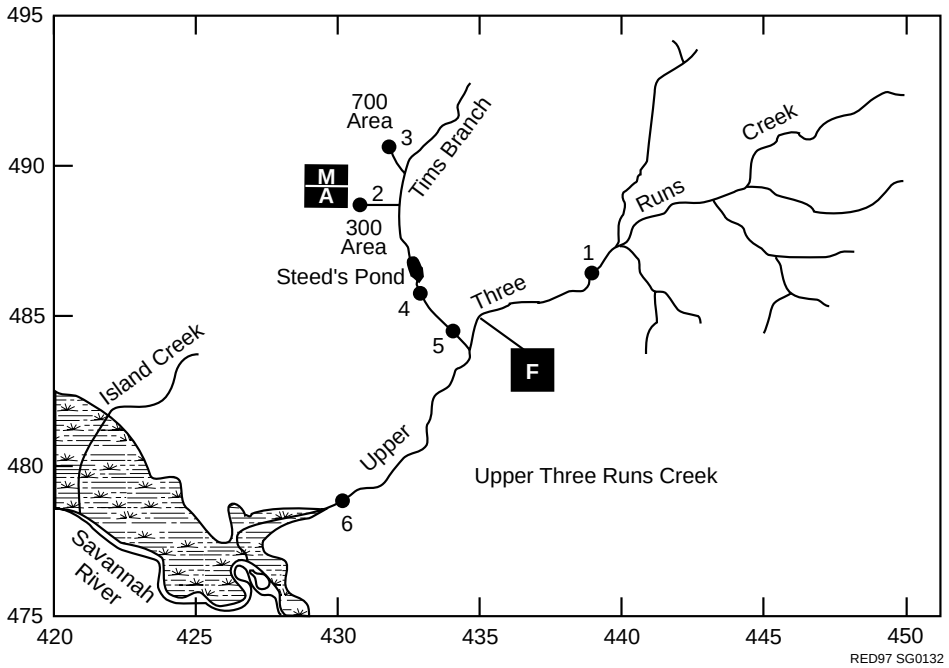

Figure 5-3. Map of the Upper Three Runs Creek and its tributaries at the SRS with water sampling locations in the early years. Sampling location 6 represents the point where Upper Three Runs Creek crosses Road A, the final sampling location onsite before the creek flowed into the Savannah River. The numbers on the axes are from an SRS coordinate system used in the 1950s. (Redrawn from Alexander and Horton 1956).

At Road C, Tim's Branch discharges industrial, laboratory, water treatment, and sanitary wastewater treatment effluents from A-Area and M-Area to Upper Three Runs. The M-Area fuel fabrication facility discharges liquid effluents to Tim's Branch, which contains large quantities of acids, bases, and salts with widely varying pH values. Starting in early 1973, these wastes were diverted to a settling basin. Other discharges to the A-Area and M-Area effluents include drainage from such activities as power facilities, automobile maintenance, and equipment overhaul and can contain contaminants such as oils, greases, detergents, corrosion inhibitors, boiler water chemicals and other trace chemicals, and pesticides. Steed's Pond, located on Tim's Branch just north of Road 2, has served as a mixing and settling pond for A-Area and M-Area wastes, and the Site routinely sampled the outfall from the pond to determine deposition or holdup in the pond.

From Road C to Road A, the only source of releases should be runoff from power line right-of-way herbicide treatment, sanitary wastewater treatment effluent, and, possibly, corrosion inhibitors. Between Road A and the Savannah River, the stream is again subject to drainage from offsite activities, particularly from the land area recently released to the U.S. Forest Service.

Four Mile Creek

This creek rises on SRS property, about 1.5 mi east of its first confluence with an SRS discharge, the effluent from H-Area separations at a point a short distance east of Road C (Figure 5-4). The H-Area discharge consists of process cooling water, which contains water treatment chemicals, possibly corrosion inhibitors, sanitary wastewater treatment effluent, and, potentially, runoff from spills. The effluent also contains sulfuric acid and caustic solutions from ion exchange resin regeneration in the water treatment plant. An ash pit is located just south of the area, below Road E. Leaching from this pit would reach Four Mile Creek by way of a small stream that joins the H-Area effluent. Between Road 4 and Road C, the creek receives water from the H-Area seepage basins that has permeated the soil to the water table and moved with the ground water to the outcrop location at the stream (Ashley 1964).

Discharges from the F-Area reached Four Mile Creek just before Road C. Below Road C, outcropping of water permeating from the F-Area seepage basins added to the further discharge of materials to the creek. Below this point, Four Mile Creek received sanitary wastewater effluent; traces of oil, chemicals, and solvents from the Central Shop Area; runoff from nonpoint sources; and possible leaching from the C-Area ash pit. The creek merges with the C Reactor cooling water about 0.5 mi south of Road 3. The operating reactor cooling water flow was much greater than that in the receiving stream; as a consequence, materials already in Four Mile Creek were greatly diluted. This was considered advantageous from the standpoint of release concentration limits, but it is a disadvantage in that analyses become more difficult.

From the junction with the C-Area effluent, Four Mile Creek flows for about 7 mi before reaching the swamp. This section of the stream received no point discharges, only runoff from highway and power line right-of-way. The water was dispersed in the swamp and part of the flow reached the river through Beaver Dam Creek, part at the mouth of Four Mile Creek, and part, after flow in the swamp parallel to the river, through Steel Creek. At the mouth of Four Mile Creek, the temperature of the discharge to the river was usually several degrees higher than the ambient river temperature. In fact, this discharge had the most significant thermal effect on the river of any SRS discharge.

Beaver Dam Creek

This creek receives the effluent from the D-Area facilities, including the heavy water production plant and a coal-burning powerhouse (Figure 5-4). Conventional water treatment chemicals to condition the water for various uses are present in the discharge. Water used for process and powerhouse condenser cooling is elevated in temperature when discharged. Leaching from the ash pits at the 400-D power house is source of releases of chemicals to this stream. The drainage systems from the 400-D power house consisted of a 300-m channel that connected the ash settling basin and the SRS swamp. The water flowed through the swamp to Beaver Dam Creek. Water from Beaver Dam Creek (1.3 km in distance from the confluence) entered the swamp and followed the edge of the swamp to the entrance of Four Mile Creek (Newman et al. 1985). The creek follows a meandering route through the swamp enroute to the river, including mixing with a portion of the Four Mile Creek flow.

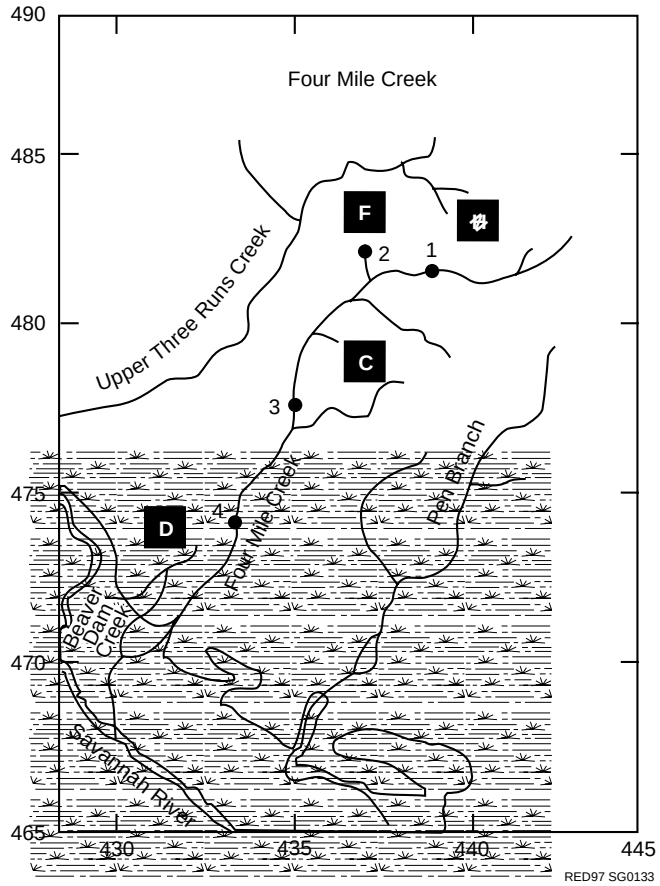

Figure 5-4. Map of Four Mile Creek and its tributaries at the SRS with water sampling locations in the early years. Sampling location 4 represents the point where Four Mile Creek crosses Road A, the final sampling location onsite before the creek flowed into the Savannah River. The numbers on the axes reflect an internal SRS coordinate system used in the 1950s. (Redrawn from Alexander and Horton 1956).

Pen Branch

Pen Branch, above its confluence with the K Reactor effluent, receives only drainage water from sources such as Central Shops, the L-Area ash pit, and herbicide applications (Figure 5-5). The K-Area cooling water discharged into Indian Grave Branch, a tributary of Pen Branch. The materials released in the cooling water were similar to those described for the C Area. Leachate from the K-Area ash pit drained into Indian Grave Branch above its confluence with Pen Branch about 0.5 mi north of Road A. Pen Branch discharges to the swamp about 4 to 5 mi below Road A and is subject only to runoff along this distance. Pen Branch water then flowed through the swamp, dissipating heat from K Reactor cooling water, and joined Steel Creek about 0.5 mi from its mouth.

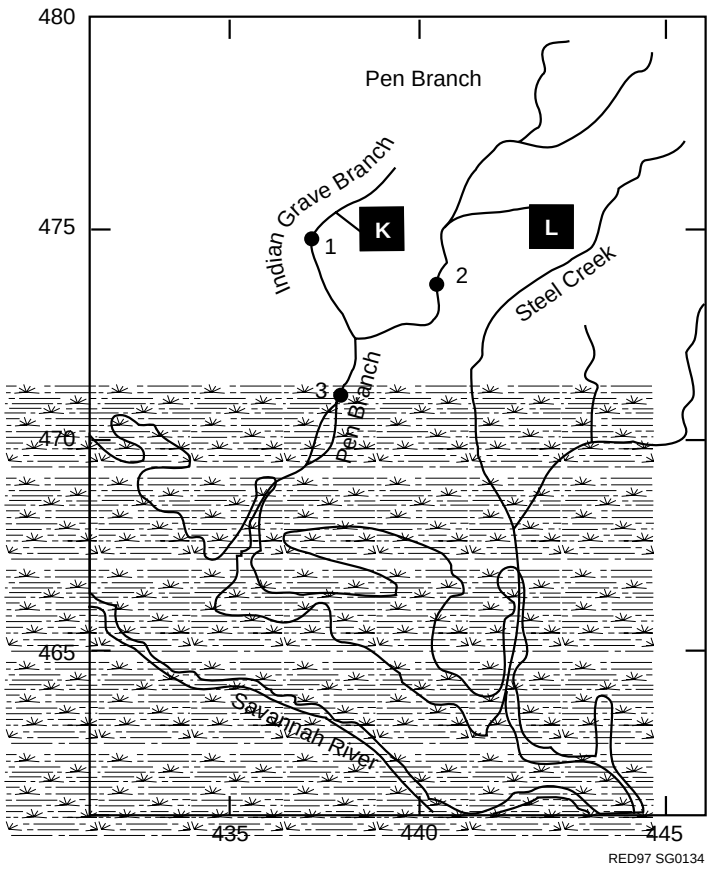

Figure 5-5. Map of Pen Branch and its tributaries at the SRS and water sampling locations in the early years. Sampling location 3 represents the point where Pen Branch crosses Road A, the final sampling location onsite before the creek flowed into the Savannah River. The numbers on the axes are reflect an SRS coordinate system used in the 1950s. (Redrawn from Alexander and Horton 1956).

Steel Creek

After SRS operations began, Steel Creek headwaters were diverted to receive miscellaneous discharges from the 105-P Reactor building (Figure 5-6). The P Reactor cooling water was not released directly to Steel Creek but to a canal system for recirculation in Par Pond after Par Pond was constructed in 1958. The P Reactor supply water came from the river pumphouses and, to a greater extent, from the Par Pond Pumphouse 681-6G. From P Reactor to its confluence with Meyers Branch, just north of Road A, Steel Creek was subject only to runoff, including possible drainage from waste disposal and storage facilities in the L Area. No point discharges were made to Steel Creek below Meyers Branch. The K Reactor effluent from Pen Branch was discharged to Steel Creek via the swamp through one major and one or more minor streams about 0.5 mi above Steel Creek mouth. The water in Pen Branch cooled as it moved through the swamp, and the thermal effects of Steel Creek discharges to the river were somewhat dissipated.

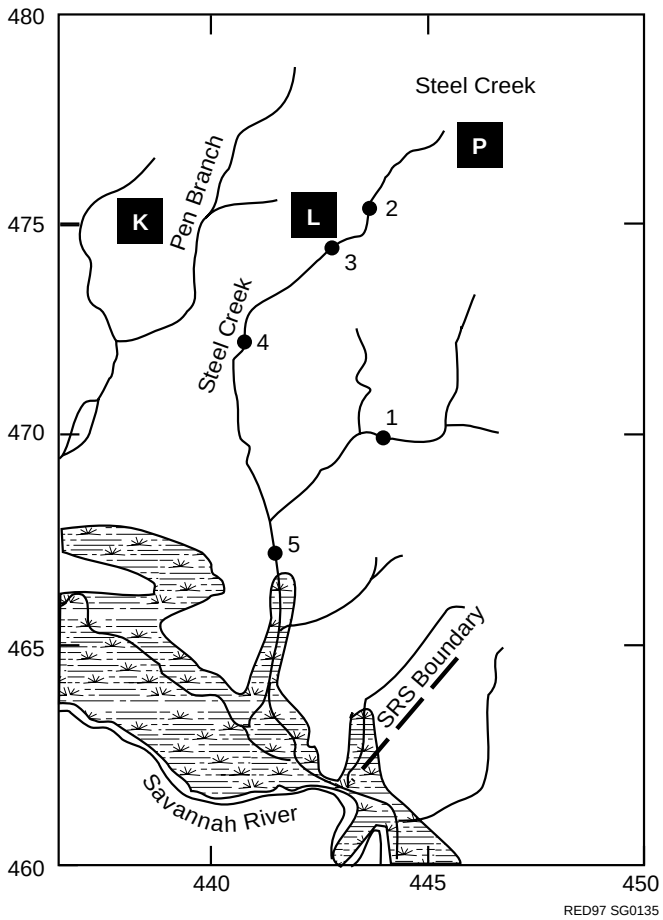

Figure 5-6. Map of Steel Creek and its tributaries at the SRS and water sampling locations in the early years. Sampling location 5 represents the point Steel Creek crosses Road A, the final sampling location onsite before the creek flowed into the Savannah River. The numbers on the axes reflect an internal SRS coordinate system used in the 1950s. (Redrawn from Alexander and Horton 1956).

Lower Three Runs Creek

Lower Three Runs (LTR) Creek has the second largest drainage area of streams on the SRS. Both P- and R-Reactors have discharge effluent to the LTR system. The overflow from Par Pond to Lower Three Runs Creek carried runoff from the northeast portion of the plant, sewage treatment effluent, water treatment chemicals added to P Reactor cooling water, and any materials leaking to the cooling water such as sodium borate from the emergency cooling water system (Figure 5-7). Monitoring at this point (where Par Pond overflowed to the Lower Three Runs Creek) provided the major source of discharge inventory data from SRS to this creek. It also proved necessary to monitor the P-Area cooling water discharge to the canal system. Just below the Par Pond dam overflow, the Barnwell Nuclear Fuel Plant effluent was discharged to Lower Three Runs, and still further downstream, the effluent from other offsite facilities, as well as SRS runoff entered this creek. Below Patterson's Mill Bridge, the stream was still within the SRS property boundary, but the corridor was so narrow that it afforded little protection for runoff from

surrounding agricultural activities. In addition, the J. P. Stevens Co. textile facility effluent was discharged to the stream between Tabernacle Church Road and S. C. Highway 125. The Site considered that monitoring at locations along the corridor and at the mouth of the Lower Three Runs provided data to contest alleged discharge violations (by comparison with Par Pond overflow data). Because of the presence of these other non-SRS discharge points, the Site felt that monitoring stations along the corridor and at the mouth of the Lower Three Runs Creek would not be useful to determine discharge inventories.

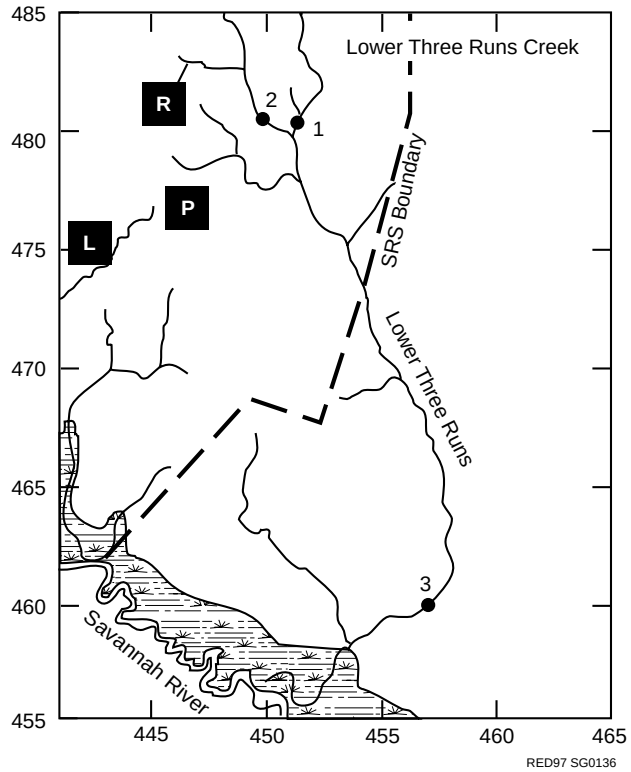

Figure 5-7. Map of Lower Three Runs Creek and its tributaries at the SRS with water sampling locations in the early years. The numbers on the axes reflect an internal SRS coordinate system used in the 1950s. (Redrawn from Alexander and Horton 1956).

Site Stream Flow

Site stream flow data can provide an indication of processing activities onsite and are important for calculating total levels of activity released when radionuclide concentration is measured. Data like these play an important role in reconstructing releases from the Site streams to the Savannah River because the flow rate directly affects the dilution factor.

The U.S. Geological Survey (USGS) measured stream flow on four of the five Site streams beginning in 1959. The flow for Four Mile Creek and Steel Creek were influenced to a large extent by SRS operations. Four Mile Creek received effluents from the separations areas and cooling water from the C Reactor. In addition, shallow groundwater outcrops to Four Mile Creek from the H-Area and F-Area seepage basins and from the radioactive burial grounds. Because the C Reactor cooling water contributed a large fraction of water to the Four Mile Creek flow, the

general flow in Four Mile Creek decreased dramatically when the C Reactor was not operational. Figure 5-8 shows the flow rate in Four Mile Creek and how greatly it was influenced by C Reactor operations (Ashley 1959). Figure 5-9 shows the flow rates of Steel Creek and Lower Three Runs Creek from 1959 through 1967.

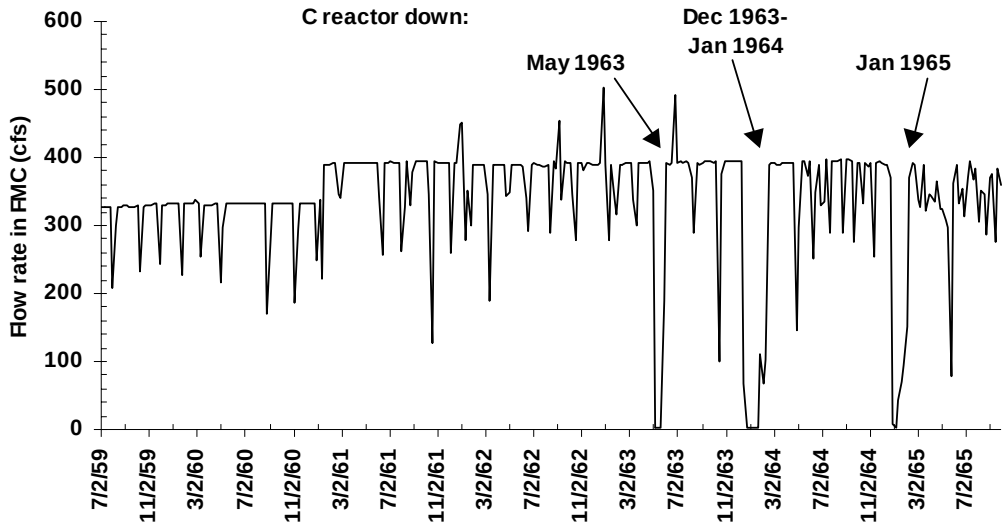

Figure 5-8. The flow rate in cfs for Four Mile Creek from July 1957 through June 1965. Four Mile Creek received cooling water and liquid effluent discharges from the C Reactor and the separations areas. The flow rate in Four Mile Creek more than doubled from that measured upstream when the C Reactor was operating. The dates indicate some of the times when the C Reactor was not operating and, therefore, was not discharging cooling water and other liquid effluents into Four Mile Creek.

Statistical characteristics of the flow rates in each of the streams discharging liquid wastes from SRS are given in Table 5-1. Steel Creek had the highest average flow rate of almost 500 cfs and Upper three Runs had the lowest. These data, along with concentration measurements made at the same locations, were used to calculate the total quantities of key radionuclides discharged from the Site to the Savannah River.

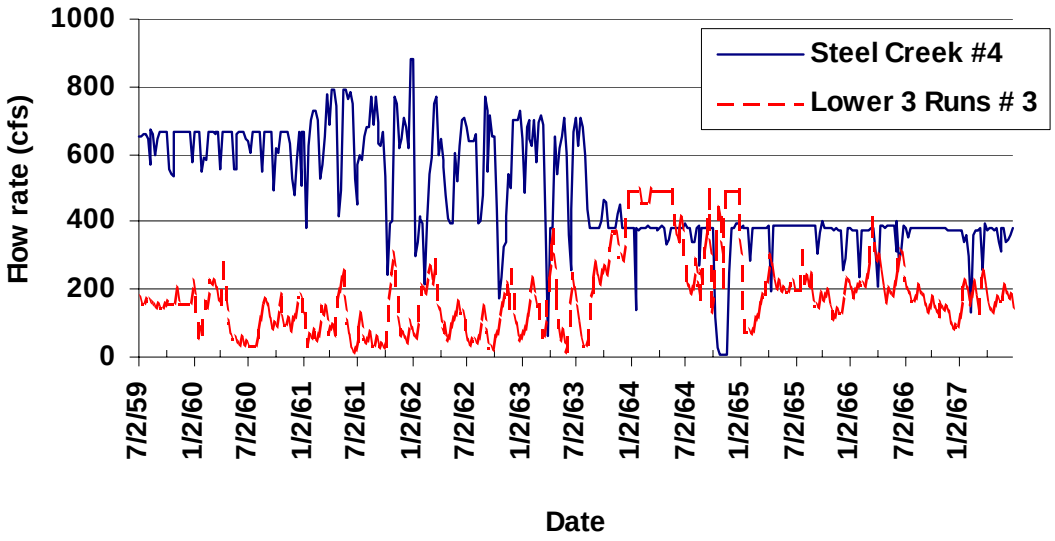

Figure 5-9. Flow rate in Steel Creek and Lower Three Runs Creeks from July 1959 through June 1967 measured at Road A on each stream by the USGS. The noticeable decrease in flow rate in Steel Creek and increase in Lower Three Runs in the fall of 1963 coincided with fuel failures and other operational problems in the R Reactor. The R Reactor was permanently shut down in 1964, at which time the flow of cooling water from the R Reactor to Steel Creek was stopped, and some cooling water from the P Reactor was diverted to Lower Three Runs. Cooling water flow from the reactors at SRS was a major contributor to stream water.

SRS WASTEWATER CONTROL

The SRS implemented various measures to control and limit the volume of liquid effluents that entered the Site streams directly and would eventually drain into the Savannah River. The liquid wastes from the Reactor Areas consisted primarily of disassembly basin water purged to maintain clarity in the basins. Each reactor area also had a 50 million gallon basin for emergency conditions to retain water that contained short lived radionuclides. In the separation areas, liquid effluents originated as cooling water from the process buildings in the F-Area and H-Area, from runoff to storm sewers, and from the seepage basins (Du Pont 1973).

The cooling water was classified as either circulated cooling water (circulated in the single-purpose coils of the canyon vessels) or segregated cooling water (circulated in the dual-purpose coils for heating and cooling). It was more likely to have radionuclide releases to the streams from the segregated system (Ice 1971). In both the circulated and segregated water system, the water was monitored and armed with alarms that would indicate high levels of either alpha or beta-gamma activity. If this occurred, the water was diverted to a retention basin or to the seepage basins. Storm sewer water from the separation areas was normally discharged to the Four Mile Creek. When spills or other sources of contamination threatened to drain into the storm sewers, emergency measures were implemented. These measures involved damning the streams into which the sewer discharged and then pumping the impounded water into the seepage basins.

Table 5-1. Flow Rate Characteristics of SRS Site Streams in the 1960s[a]

Flow rate parameter	Upper Three Runs[b]	Four Mile Creek #6	Pen Branch #3	Steel Creek #5	Lower Three Runs #2	Par Pond	Savannah River
	Flow Rate (cfs)						
Median		374	387	395	162	337	
Average	270	340	349	486	184	305	11,000[c]
Standard deviation		87	87	164	119	118	
Maximum	550	502	518	880	496	486	
Minimum	190	0	3	6	12	3	

[a] Based on reported weekly flow measurements made by the USGS from July 1959 through June 1967 (Ashley 1959). These values were converted into cfs for this table.

[b] Upper Three Runs has the largest natural flow rate (about 257 cfs) of the onsite streams at SRS; plant contributions raise the natural flow rate by only about 10 cfs. This stream and its major tributaries, Tinker Creek and Tim's Branch, drain an area of about 545 km^3 (Evans et al. 1992).

[c] This is equal to about 300 $m^3\ s^{-1}$; the average velocity of the Savannah River is 5–7 feet per second (3–5 mi per hour) (Zeigler et al. 1985)

Seepage Basins

Seepage basins were constructed in each of the two chemical separations areas before startup in 1955. They were originally designed and arranged so that liquid discharged to basin 1 overflowed to basin 2, and finally overflowed to basin 3 (Ice 1971). Tables 5-2 and 5-3 provide statistics on the history, size, and seepage characteristics of the basins. There is an underground seepline that roughly parallels Four Mile Creek that carries some contaminants from the seepage basins into FMC. The distance and flow time between the seepline and FMC varies depending on location (Table 5-3). Christensen and Gordon (1983) gave estimates of about 9 years travel time from the F-Area SB and from 1-4 years for contaminants from the H-Area. Thus, the location of the outcropping from groundwater to FMC is not a single point. The outcrop from F-Area occurs between sampling locations 2 and 3 (Figure 5-4). For H-Area, the outcrop to FMC occurs between sampling locations 1 and 2; thus, the distance is shorter but still not a single point location. This entire area is wet and swampy and the seep line actually comes quite close to FMC in several places.

Immobilization plus radioactive decay greatly reduce the amount of radioactive material eventually leaving control of the SRS (Marter 1969). Marter and others calculated the fraction of each radionuclide that would eventually move through the soil by a groundwater/soil transport equation that takes into account:

Table 5-2. Characteristics and General Information about the Seepage Basins in the Separations Areas

Location	Basin No.	Bldg. No.	Used	Area (acres)	Acre (ft^2)	Volume (gal)
200-F	-	904-49	1954–1955	1.3	56,550	na - NW of area
	1	904-41	1955–1988 [a]	0.34	14,800	1,000,000
	2	904-42	1955–1988 [a]	0.69	30,000	1,900,000
	3	904-43	1955–1988 [a]	4.44	193,000	14,000,000
Total F-Area seepage basins					238,000	
200-H	1	904-44	1955–1988[a]	0.29	12,615	1,000,000
	2	904-45	1955–1988 [a]	0.79	34,365	2,800,000
	3	904-46	1955–1962	3.3	143,550	2,300,000
	4	904-56	1962–1988 [a]	9.3	404,550	31,000,000 (crescent-shaped)
Total H-Area seepage basins					595,000	

a From Ice 1971; the F-Area and H-Area seepage basins were not used after 1988 when the Effluent Treatment Facility for the separations areas began operation.

Table 5-3. Seepage Characteristics of F-Area and H-Area Basins[a]

Parameter	F-Area	H-Area
Mean seepage rate (gal ft^{-2} d^{-1})	0.37	0.36
Distance to outcrop (ft)	1600	400–1400
Groundwater flow rate (ft d^{-1})	0.5	1.0
Travel time (y)	9	1–4
Basin evaporation (%)	25	27

[a] From Christensen and Gordon (1983).

The advantages of seepage basins over direct disposal to surface streams are that some ionic species are immobilized by the soil while others are greatly delayed in movement.

Using the known values for size and volume of the basins, it is possible to calculate an estimate of volume seeping from the basins over time:

Total area x mean seepage rate = volume releases per time
For F area:
238,000 ft^2 x 0.36 gal ft^{-2} d^{-1} = 88,060 gal per day from F-area seepage basins
= ~ 3.2 x 10^7 gallons per year from F-area seepage basins
For H-Area :
595,000 ft^2 x 0.36 gal ft^{-2} d^{-1} = 220,150 gal per day from H-area seepage basins
= ~ 8.0 x 10^7 gal year from the H-area seepage basins.

- Decay constant of the radionuclides
- Holdup time in the seepage basin
- Distance between the seepage basin and the nearest plant stream
- Adsorption distribution coefficient for the radionuclide in soil
- Weight of soil per unit volume of soil
- Volume of water per unit volume of soil
- Velocity of movement of groundwater (Evans et al. 1968).

Seepage basins were constructed at all reactor areas during the second half of 1957. The primary purpose was designed for disposing of waste discharged by underwater vacuum cleaning of the disassembly basins (Mealing et al. 1958). Interestingly, during the first year, at least, the radioactivity discharged to the basins was from sources other than vacuum cleaner discharges. The first R-Area seepage basin was placed in service on June 26, 1957 and received 9 Ci of nonvolatile beta activity from July through October. Then on November 8, 1957 a fuel failure in the R-Area lead to the discharge of over 1900 Ci of nonvolatile beta activity during the next 2 months. To handle the contaminated effluents, three additional seepage basins were placed in service in the R-Area on November 16, 21, and 29, respectively (Mealing et al. 1958). Special analyses of the seepage basin water collected on November 13 showed about 54% of the nonvolatile beta activity was due to radiostrontium, 14% to radiocesium and about 25% to rare earth elements. The maximum nonvolatile beta concentration observed in samples collected from these basins during this time was 8.5×10^8 pCi L^{-1}(Mealing et al 1958). The average nonvolatile beta activity measured in Lower Three Runs Creek ranged from about 200 to 300 pCi L^{-1}, and from 30 to 200 pCi L^{-1} in the Savannah River (Harvey et al. 1959). During the subsequent 6 months the concentration averaged about 25 pCi L^{-1} downstream of where Lower Three Runs enters the river (Harvey et al. 1959).

Sanitary Wastewater Treatment

When the plant was constructed in the early 1950s, primary treatment was provided for sanitary wastewater (sewage), which met South Carolina regulations. By the 1960s, regulations changed, and secondary wastewater treatment was required (Marter 1965). Du Pont Engineering Department Design developed secondary treatment facilities over time to comply with the state regulations.

Prefabricated extended aeration plants for the required secondary treatment were designed for 100-P, 200-F, 200-H, 700-A, and 400-D in the 1970s. Central Shops, with its variable work force, had a dual stabilization pond system. The remaining septic tank facilities were provided with individual tile fields, which were designed to retain liquid effluents in the soil. Table 5-4 lists the SRS wastewater treatment facilities in place by 1970.

Table 5-4. Sanitary Wastewater Facilities Present at the SRS by 1970[a]

Bldg. No.	Area	Flow (gpm)	Type of Plant in 1970	Discharge watershed
607-A	3/700	74	primary plant	Tim's Branch-Upper Three Runs
607-F	200-F	20	primary plant	Four Mile
607-H	200-H	20	septic tank	Four Mile
607-P	100-P	8	septic tank	Canal to Par Pond
607-D	400-D	8	septic tank	Beaver Dam Creek
607-K	100-K	8	septic tank	Pen Branch
607-C	100-C	10	septic tank	Four Mile
CMX-TNX		1	septic tank	Savannah River
607-U	TC-1	1	Imhoff Tank	Upper Three Runs
N. Lagoon	Central Shops	6	Lagoon	Four Mile
S. Lagoon	Central Shops	6	Lagoon	Pen Branch

[a] Adapted from Marter 1969.

High-Level Liquid Waste Storage

From the outset of operations at the SRS, special waste storage tanks were designed and installed to handle the huge quantities of liquid effluents that had activity levels too high for discharge to the seepage basins. Most high level radioactivity in liquid wastes was generated in the separations areas as a result of chemically processing the irradiated fuel from the reactor areas. The original storage tanks, eight in the F-Area and 4 in the H-Area, relied on gravity or steam-jet siphons for the transfer of radioactive liquids. Valves were not used so waste flow was directed to the appropriate tank by a change of pipe jumpers in diversion boxes located near the tanks. All piping was stainless steel and concrete trenches, which provided secondary containment, were equipped with leak collection and detection facilities. Some of the key highlights of the early waste handling history of the Site were described in the monthly and semi annual reports (Alexander and Horton 1956, Ashley 1967b, Du Pont 1958, Du Pont 1961-1962, Du Pont 1971). Some of the important events include:

- 1956 Four additional cooled, million-gallon waste storage tanks completed in the H-Area
- 1957 Four 1.3 million gal , uncooled tanks completed in F-Area
- 1958 Construction of four 1.3 million-gallon uncooled tanks started in H-Area; some leakage through steel tank wall in 3 H-Area tanks
- 1960 Operation of F-Area tank farm evaporator started
- 1962 Continuous inventory of fission products and alpha emitters in tanks begun
- 1963 Operation of H-Area tank farm evaporator started
- 1964 Total fission products in storage increased to 865 million Ci
- 1965 Total fission products in storage decreased to 745 million Ci; a fire in a tank ventilation filter due to an accumulation of vapor phase inhibitor; cesium removal ion exchange columns limits ^{137}Cs releases to seepage basins
- 1966 Total fission products in storage increased to 885 million Ci

- 1967 Total fission products in storage decreased to 851 million Ci; construction of four additional 1.3 million-gallon cooled waste tanks started in H-Area
- 1968 Total fission products in storage increased to 937 million Ci
- 1969 Total fission products in storage decreased to 798 million Ci; construction of 2 1.3 million-gallon cooled waste tanks started in F-Area
- 1970 Total fission products in storage decreased to 790 million Ci; several tiny explosions occurred at the H-Area tank farm evaporator; 7 of 36 cooling coils in one tank started leaking bringing he number of leaking coils to 17 and leaving 19 coils available for cooling.

Throughout this period, there was continual effort to reduce the volume of high-level effluent, usually by evaporation. From startup through about 1970, about 39 million gal of high and low level waste were sent to the tank farms in the F-Area and H–Area. Of that volume, 18 million gal remained in 1971, the other volume had been lost by evaporation and concentration of wastes (Ice 1971).

EFFLUENT AND ONSITE SAMPLING

Liquid effluents from the major release points at SRS were monitored from startup of operations at the SRS. This onsite effluent monitoring served to alert the Site personnel of problems that might be occurring and of the release of contaminants to the Site streams. A routine onsite monitoring program was established by 1952 and reports were issued routinely in the form of weekly, monthly and semiannual reports (For example, Du Pont 1953, Du Pont 1958).

Liquid Effluent Sampling Procedures

Water samples from the various reactor building effluent systems were monitored extensively to track levels of radioactivity leaving the facilities. Historically, the disassembly basins were the main source of radioactivity from the reactor areas to the Site streams, with some effluent contributed from the heat exchanger cooling water and the shield cooling water. To obtain gamma measurements from the disassembly basin flow to the Site streams in the early years, continuously operating ionization chambers were suspended in the liquid effluent stream of the weirs of the disassembly basin overflows and the gamma readings recorded. Water samples of the overflow from the disassembly basins to the streams were collected by six weir samplers located along the disassembly basins and canal during each shift at a designated time and recorded at approximately the same time as the gamma measurements were done (Du Pont circa1960).

In the 1950s, the heat exchanger cooling water was discharged by pipeline to an effluent ditch that drained to the Site streams. There was no continual monitoring of this discharged water; rather, periodic samples were taken through a hatch in the grating over the discharge tank in the 107 buildings at a designated time during each shift. The shield cooling water held in an underground tank and designated the 109 building, was located adjacent to the 105 building, the reactor building. The deionized water purged from the shield cooling systems was piped to the tank, The purged water contained corrosion products and fission and activation products from the reactors. The shield cooling water tank held up the water to permit radioactive decay of some

products and to allow settling of the heavier radioactive materials. Sampling was done during each shift through a hatch in the top of the overflow weir in the tank to allow for radioactive decay and to allow settling of materials.

In general, the Site stream and river water were analyzed for the same radionuclides as reactor effluent samples. Before the 1970s, suspended and dissolved solids were not separated for routine analyses. River flow was estimated from information provided by the USGS, while plant effluent stream flow measurements were supplied by the SRS Power Department. Table 5-5 provides a summary of the radionuclides in liquid effluents that were routinely monitored by the early 1960s.

In 1973, a major survey of the effluent and environmental monitoring program at the SRS found that overall improvements could be made and that the Site needed to make "substantial improvements in its capability to monitor nonradioactive releases because of new pollution control legislation" (Reinig et al 1973). Special attention was directed to ensure that representative samples were collected at the point of release. In general, effluents were continuously monitored at a major release point where releases may have been in excess of 10% of an operating guideline or have a potential for large unplanned releases. If the anticipated annual release from a specific emission point was less than 1% of the operating guideline then only grab sampling was the normal operating procedure. For some emission points, quantities released were calculated where emission were accurately established; for example the release of noble gases in irradiated fuels.

By the 1980s, all purges, or batch releases, from the disassembly basins were discharged to the reactor area seepage basins (from the P, L, and C Reactors) or to a containment basin (K Reactor). An in-line calibrated water meter measured the flow to the seepage basin during a purge. A sample was collected automatically every 3 minutes during a purge from the disassembly basin. The area survey health physicist removed the samples sent them to the Environmental Monitoring Laboratory for analyses at the end of each month. For process control, the Operating Department collected a sample every 4 hours during a purge and sent the sample to the 772-D Laboratory for gross alpha and beta, tritium and gamma pulse height analysis (Zeigler 1986). Meanwhile, continuous sampling of the effluent going to the process sewers was done. Every 2 minutes, a predetermined quantity, which was proportional to the expected average flow, was collected and a composite of the daily collection was analyzed. The Environmental Monitoring Laboratory analyzed the composite samples monthly for a number of radionuclides, including tritium, ^{137}Cs and ^{90}Sr.

During this same time, weekly grab samples were collected from the reactor heat exchanger cooling water and sent to the Environmental Monitoring Group for tritium analysis. The "high activity" cooling water gamma monitors had alarms in the central control rooms, but there were no alarms or monitors for tritium. The adequacy of weekly grab sampling was questioned although changes in tritium concentrations in such large volumes of water (approximately 180,000 gallons per minute) were said to occur very slowly (Zeigler 1986).

Table 5-5. Radionuclides Routinely Analyzed in Reactor and Separations Areas Effluents in 1962[a]

		Liquid Effluent	
Type of analyses	Radionuclide	Reactor to streams[b]	Separations to seepage basins[c]
Chemical separations	^{147}Pm	x	x
	^{91}Y	x	
	^{90}Sr	x	x
	$^{89,90}Sr$	x	x
	^{35}S	x	
	^{32}P	x	
	(Uranium and plutonium)		x
Gamma spectrometry	^{239}Np	x	
	^{233}Pa		x
	$^{141,144}Ce$	x	x
	$^{140}BaLa$	x	
	$^{134,137}Cs$	x	x
	^{131}I	x	x
	$^{124.125}Sb$		x
	$^{103.106}Ru$	x	x
	$^{95}ZrNb$	x	x
	^{65}Zn	x	
	$^{58,60}Co$	x	
	^{54}Mn	x	
	^{51}Cr	x	
Liquid Scintillation	^{3}H	x	x
Alpha Spectrometry	Alpha emitters[a]	x	x

[a] Adapted from Du Pont 1965; alpha releases from reactor areas were considered negligible; samples were counted for only gross alpha activity.
[b] For reactors, sampling frequency was varied according to fuel discharge and concentration of radionuclides in the disassembly basin. Flow estimates were made according to purge rates and the Ci release inventory based on flow and radionuclide concentration.
[c] In the separations facilities, liquid wastes were discharged to open seepage basins. Sampling volume was proportional to flow and was done continuously. Routine analyses were done on weekly samples.

Liquid effluent releases from the separations areas to streams were continuously sampled with a high-activity alarm in place in the 1980s. A paddlewheel sampler with a Brailsford battery operated pump was used at the H-Area and F-Area effluent discharge points. Samples were collected and analyzed weekly. The sample concentrations were combined with USGS flow data to determine release quantities. The volume in the H-Area effluent stream varied widely at times, causing sampler failure to occur. Releases from the M-Area to Tim's Branch were also continuously sampled with a flow-proportional (trebler) sampler in the effluent line and analyzed weekly. Only periodic calibrations of flow measurements were performed with the trebler and flow measurement calibration was noted as an area of uncertainty at that time (Ziegler 1986).

The Operations Department collected samples of the wastewaters that had been stored in tanks from the 420-D and 421-2D areas. The samples were analyzed for tritium and beta-gamma emitters. If the analyses were within the established limits, the water was released to Beaver Dam Creek (Figure 5-4). The quantity of water in each tank was determined from a sight glass reading before release to the creek. The area survey health physics team collected daily samples from the Brailsford pump sampler on effluents from 420-D and the process sewer downstream of all tritium rework facilities. There was no sampling point downstream from the Heavy Works facilities onsite so release estimates were based on measurements from the facilities. Because it was possible for radioactivity to be released to the effluent from routes other than the batch holding tanks, the uncertainties associated with tritium releases from the D-Area were much broader than those on Site streams where multiple sampling points were located (Marter 1969). On the other hand, the tritium releases from the D-Area were quite small when compared to releases from the reactor and separations facilities (See Figure 5-12).

Stream Sampling

Effluents from the processing and reactor areas onsite flowed into the five major Site streams, each described at the beginning of this chapter (Figures 5-3, 5-4, 5-5, 5-6, and 5-7). Water samples were collected at regular intervals from several locations on all Site streams, and at least one sample was taken above the location of the effluent discharge into each stream so increases in radioactivity could be detected (Horton 1954). The number of weekly water samples collected in this program increased from 384 samples during the first six months of 1953 (Albenesius 1954) to almost 1200 water samples by the end of 1954 (Horton 1955a). During the late 1950s about 950 water samples were collected in a typical six-month period from the onsite streams.

SRS initially took grab samples of streams and the Savannah River during the preoperational surveys and early monitoring programs. A 2 bottle siphon action apparatus was later used for continuous stream sampling. In 1958, a water wheel sampler was developed when line plugging and maintenance problems continued to occur with the siphon action apparatus (Johnson 1964). The water wheel continuously pumped a small volume through one system to a collection container. The second system was geared to pump at a rate of about 40–80 L wk^{-1} through the ion exchange column. The exact volume through the ion column was determined by multiplying the small volume collected in the first system by a known gear ratio of the pumps. Prefilters were placed before the ion column to prevent plugging with sediments.

The water sampling and monitoring methods for each stream depended upon the particular effluents they received. For example, paddlewheel samplers collected continuous samples of the effluents from the F-Area and H-Area that flowed into the Four Mile Creek. The anchored paddlewheel sampler floated in the stream, and collected samples in a small cup attached to the paddlewheel. As the stream flowed, the paddlewheel water was collected in the cup and emptied into a collection trough. The water flowed down the trough and into a collection bottle. The preferred paddlewheel samplers were also used on the streams at Road A, the final onsite sampling location for all streams before flowing into the Savannah River. On the other hand, effluents from the A and TNX areas were grab sampled and those from the M-Area were monitored with a trebler sampler. These latter areas did not contribute the levels of radioactivity that other facilities did (see Figures 5-12, and 5-13). Nevertheless, all Site streams were monitored routinely and fairly extensively from plant start up through the present day. Although

the actual sampling locations remained approximately the same over time, the designations for the sampling locations changed. Figure 5-10 shows the SRS site with major facilities, site streams, and roadways. While there were several sampling locations along each of the Site streams, the final sampling point onsite before the streams flowed into the Savannah River was at Road A. Table 5-6 summarizes the sampling location identification numbers from 1954 through 1980 on the five major Site streams (Upper Three Runs, Four Mile Creek, Pen Branch, Steel Creek, and Lower Three Runs), and their smaller tributaries. The table shows how the identification of sampling locations changed over the years, and, more interesting, is the fact that the number of sampling locations on Site streams at the SRS was greatest in the 1950s (24 locations), and decreased to less than half that number (10) by the mid-1970s.

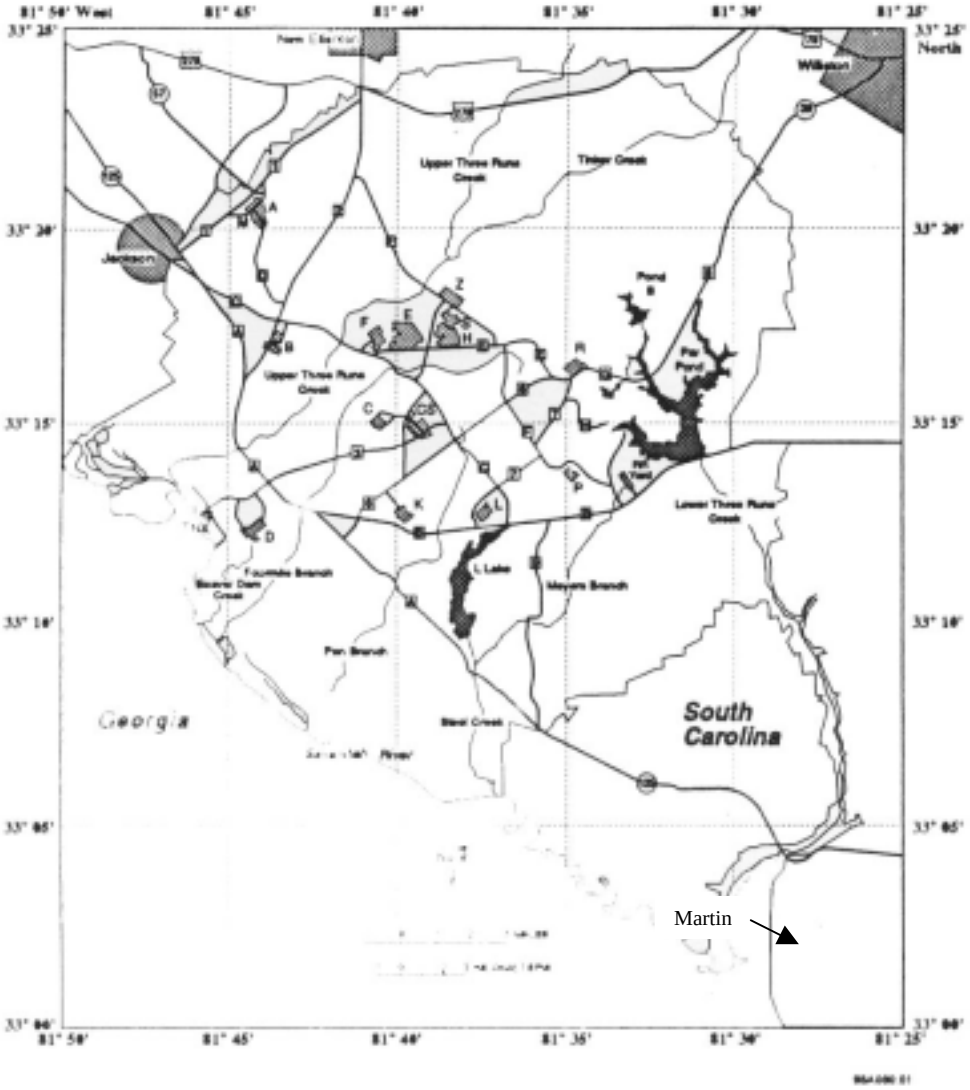

Figure 5-10. The SRS showing major facilities and roadways. The final sampling locations for Upper Three Runs, Four Mile Creek, Pen Branch and Steel Creek, before entering the Savannah River, were taken at Road A which becomes Highway 125 to the southeast.

Table 5-6. Identification Numbers for Stream Sampling Locations at the SRS[a]

		Sample location ID #							
Site stream	Physical Description	1954-1958	1959-1960	1961-1963	1964-1968	1969-1970	1971	1972	1973-1980
Upper Three Runs	**UTR at Road A**	**4**	**4**	**4**	**4**	**7**	**7**	**7**	**ns**
(UTR)	UTR at rail line	3	3	5	5	5	5	5	5
	Discharge stream -- F to UTR	2	2	2	2	2	2	2	2
	UTR at Tinker Crk	1	1	1	1	1	1	ns	ns
Tim's Branch	TB at UTR	5	5	ns	ns	Ns	ns	ns	ns
(TB)	Discharge stream - M/A to TB	3	3	4	4	4	4	4	ns
	N. offsite stream to TB	2	2	3	3	3	3	3	ns
Four Mile Creek	FMC at Fischer Pond Rd	7	ns	ns	ns	ns	ns	ns	ns
(FMC)	**FMC at Road A**	**6**	**6**	**10**	**11**	**12**	**12**	**12**	**6**
	FMC S. of C Reactor	5	5	ns	ns	ns	ns	ns	ns
	FMC NW of C Reactor	4	4	9	10	10	10	ns	ns
	FMC SW of F-Area	ns	ns	ns	ns	1	1	1	ns
	FMC at Road C (S of H-Area)	2	2	8	9	10	10	10	ns
	Discharge stream-F to FMC	3	3	7	8	8	8	8	ns
	Discharge stream-H to FMC	ns	ns	ns	ns	9	9	9	ns
	FMC at Tinker Crk	1	ns	ns	ns	ns	ns	ns	ns
Pen Branch	PB at SR swamp	3	3	11	12	13	13	ns	ns
(PB)	**PB at Road A**	**ns**	**ns**	**ns**	**ns**	**ns**	**ns**	**13**	**3**
	PB between K & L Reactors	7	ns	ns	ns	ns	ns	ns	ns
Indian Grave	Indian Grave Branch NW of K Reactor	2	2	ns	ns	ns	ns	ns	ns
Steel Creek	SC at PB confluence	ns	ns	ns	ns	ns	ns	ns	5
(SC)	**SC at Road A**	**4**	**4**	**12**	**13**	**14**	**14**	**14**	**4**
	Discharge stream-L to SC	3	3	ns	ns	ns	ns	ns	ns
	SC between L and P	2	2	ns	ns	ns	ns	ns	ns
Meyer's Branch	Meyer's Branch	1	ns	ns	ns	ns	ns	ns	ns
Lower Three Runs	LTR at Martin	ns	6	17	18	18	18	18	3
(LTR)	LTR at Stinson Bridge	3	ns	ns	ns	ns	ns	ns	ns
	LTR at Patterson Mill Bridge	ns	5	16	17	17	17	17	2
	LTR at upper E. branch	1	ns	ns	ns	ns	ns	ns	ns
	LTR at upper W. branch	2	ns	ns	ns	ns	ns	ns	ns
	Par Pond into LTR	na	4	ns	ns	ns	ns	ns	ns
	Par Pond into LTR-P branch	na	3	14	15	15	15	15	2
	Par Pond into LTR-R branch	na	2	15	16	16	16	16	3
Total Locations		24	22	16	16	18	18	16	10

[a] Adapted from Du Pont 1965 and Du Pont 1973.
ns = not sampled
na = not applicable, Par Pond was constructed later

DOCUMENTATION OF RELEASES

For surface water source term development, release estimates are calculated for the key radionuclides that are released offsite. For surface water, offsite means the point that the effluents drain into the Savannah River. Unlike atmospheric source term development where releases are estimated from the point of release from a particular facility into air, the liquid effluents are released from facilities to seepage basins or to onsite streams.

To reconstruct the releases to surface water, we have used the most original and basic information available. The most basic data for surface water releases are the original weekly results handwritten from the laboratory ledger books and retained at the SRS on aperture cards (which are similar to microfiche). *Radiological Assessments Corporation (RAC)* copied the measurement data for air, water, and environmental monitoring that were needed to complete this study. These references are called aperture card printouts throughout this report. Another useful information source for surface water source term development was the experimental notebooks of Clarice Ashley (Ashley 1959), in which she compiled analytical results of the surface water releases. Ashley tracked the results of the weekly stream flow and concentration measurements for nonvolatile beta, tritium, ^{131}I, strontium, and cesium at key sampling locations on Site streams. We corroborated the experimental notebooks of C. Ashley with the handwritten entries in the aperture card printouts.

These handwritten entries provided radionuclide concentration, flow rate data and some quality control information for the major onsite streams. The Site used these data to calculate and report release estimates as total activity in curies (Ci) in the appropriate monthly report. In the early years, the radioactivity in surface streams was usually reported as nonvolatile beta, and tritium. Beginning in the late 1950s specific radionuclides, like ^{137}Cs and ^{90}Sr, were measured at the facility discharge points, in the Site streams and in the Savannah River. For our work, we calculated monthly averages from these weekly data and compared the results to the values reported in monthly reports to check the validity and credibility of monthly or other summary types of reports. The monitoring data of radionuclides in the Site streams for the key radionuclides have been compiled in the Water Releases Excel workbook. In the electronic version of this document, double-clicking on the following hyperlink provides access to this workbook: Water Releases.xls.

Figure 5-11, which shows this type of comparison for 1962, supports quite good agreement between the two approaches. The calculated monthly totals from the weekly values reported in the ledger sheets give a better view of when the releases actually occurred. With the routine monthly reports, the releases were reported in the month after they occurred (e.g. February and May 1962) (Du Pont 1962). This most likely happened because the monthly reporting period was from the middle of one month to the middle of the month in which the report was issued. For the annual totals however, the values are very close. In some cases, we used these summary reports when more detailed daily or weekly data were not located.

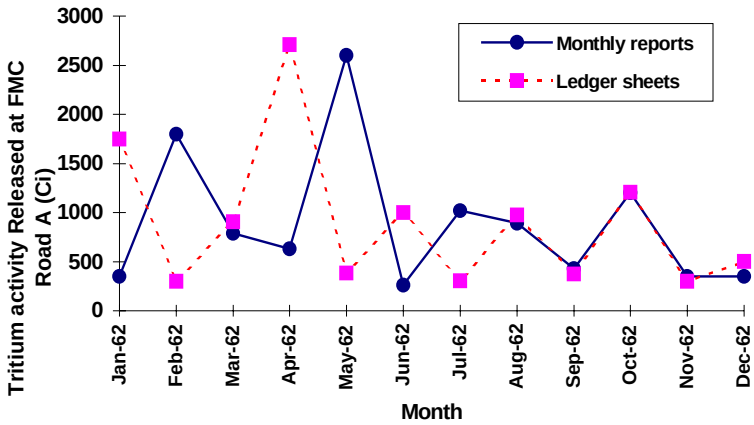

Figure 5-11. Comparison of monthly totals of tritium measured in Four Mile Creek at Road A in 1962. The solid line represents Site totals reported in monthly reports, and the dotted line shows the monthly totals calculated from weekly values in the handwritten ledger sheets. This sampling location at Road A was the final monitoring point on the Four Mile Creek before it flowed into the Savannah River.

Several complete series of monthly reports, as well as some semi-annual and annual reports for this period provided additional information for our source term reconstruction work. The monthly reports of most value were the: *Environmental Monitoring Monthly Report. Radiological Control and Methods* series (For example, Du Pont 1962, Du Pont 1964a, Du Pont 1964b, Du Pont 1967). For 1962 to 1964, these reports were each 25 to 30 pages in length, and provided quite thorough summaries of effluent and environmental monitoring for both airborne and waterborne releases. Related to surface water releases, these summaries report the liquid waste releases to separations area seepage basins for the nonvolatile beta emitters: ^{103,106}Ru, ^{89,90}Sr, ^{95}Zr-Nb, ^{137}Cs and ^{141,144}Ce; gross alpha, ^{3}H, and radioiodine. Furthermore, the average levels of activity of nonvolatile beta, radiostrontium, radiocesium, radioiodine, and tritium were reported for the onsite streams: Four Mile Creek, Pen Branch, Steel Creek, Lower Three Runs, and Par Pond. Radioactivity levels in these streams reflected discharge operations in the separations areas and reactor areas. Periodically, measurements of ^{35}S released to Four Mile Creek were reported. These documents also provided estimates of radioactivity measured in the Savannah River upstream and downstream of the discharge points from the Site. Estimates of total levels of nonvolatile beta, tritium ^{90}Sr, and ^{35}S were routinely reported.

RANKING THE RADIONUCLIDES RELEASED TO SURFACE WATER

Chapter 3 of this report describes the rationale and process used to determine the radionuclides released to water and air from the SRS that historically were the most important contributors to offsite dose. Some radionuclides were relatively more important to offsite residents than others; therefore it was important to focus our efforts on more important radionuclides so resources of the study were used most effectively. The relative importance of releases of these radionuclides to the environment depends upon the quantities released, differences in the potential for nuclide concentration in the environment, and the relative toxicity

of the radionuclides (as measured by their dose conversion factors). An updated methodology developed by the National Council on Radiation Protection and Measurements (NCRP) (NCRP 1996) was used to assess the relative importance of the identified radionuclides as potential contributors to offsite radiation dose.

For surface water pathways, the radionuclides that contribute 0.1% of the total screening radiation dose (our screening criterion) are ^{137}Cs, ^{60}Co, ^{3}H, ^{129}I, ^{131}I, ^{32}P, ^{238}Pu, ^{239,240}Pu, ^{89}Sr, ^{90}Sr, ^{35}S, ^{99}Tc, uranium, ^{91}Y, ^{65}Zn, and ^{95}Zr,Nb. The results of the NCRP screening for radionuclides in surface water showed that ^{137}Cs was the major contributor to offsite dose (~75% of the dose), with ^{60}Co, tritium, ^{131}I, ^{32}P, and ^{90}Sr jointly contributing about another 20%). Sulfur-35 and ^{65}Zn together contributed less than 4% to the screening dose via the surface water pathway, and all other radionuclides meeting our screening criteria (^{129}I, ^{239,240}Pu, ^{99}Tc, uranium, and ^{95}Zr,Nb) together contribute less than 2% to the screening dose. Refer to Chapter 3 for full details of the screening analysis.

In addition to ranking individual radionuclides for their relative importance to dose, we also evaluated the relative importance of the facilities at the SRS for their contribution to radioactivity in the Site streams. The monitoring data clearly indicate the facilities that historically contributed the highest levels of radioactivity in liquid effluents and to Site streams.

To focus our efforts on the key radionuclides and facilities, we compared the relative contributions of the major facilities to the total levels of tritium, total alpha and beta activity discharged to Site streams. Figure 5-12 summarizes results of measured tritium and beta activity discharged to Site streams from 1965 to 1969 and indicates clearly that the reactors and reactor areas are the largest contributors to total tritium and beta activity released to Site streams (Marter 1970). Beta activity represents a number of radionuclides that can be categorized as fission products and activation products. Important fission products from reactor operations at the SRS are ^{137}Cs, ^{90}Sr, and ^{131}I. Roughly 40% to 60% of the tritium and beta activity in liquid waste came from the reactor effluents. Another 30 to 40% originated from the seepage basins in the reprocessing and reactor areas. The Heavy Water Facility in D-Area contributed about 10% of the tritium activity in the liquid effluent waste stream. Figure 5-13 shows the results for alpha activity discharged in liquid effluents (Du Pont 1966, Du Pont 1967, Du Pont 1968, Marter 1970).

SOURCES OF UNCERTAINTY FOR RELEASES OF RADIONUCLIDES TO THE SAVANNAH RIVER

A number of factors and processes can affect the quantity of radionuclides that are released in liquid effluents from the reactor, separations and miscellaneous areas and eventually are released offsite into the Savannah River. As a result, a number of factors must be considered in developing the surface water source term with uncertainty estimates. Some of the factors include:

- Sampling and analytical procedures
- Properties of the radionuclide being evaluated
- Volume of liquid effluent released from the facility
- Holdup time in seepage basin, if applicable
- Distance between the seepage basin and the closest stream
- Velocity of groundwater movement where groundwater outcrops to surface streams
- Distance between Road A (the final sampling point onsite)and the river

- Adsorption distribution coefficient for radionuclides in soil (for seepage basins and the SRS swamp)
- Hold-up time in the Savannah River swamp.

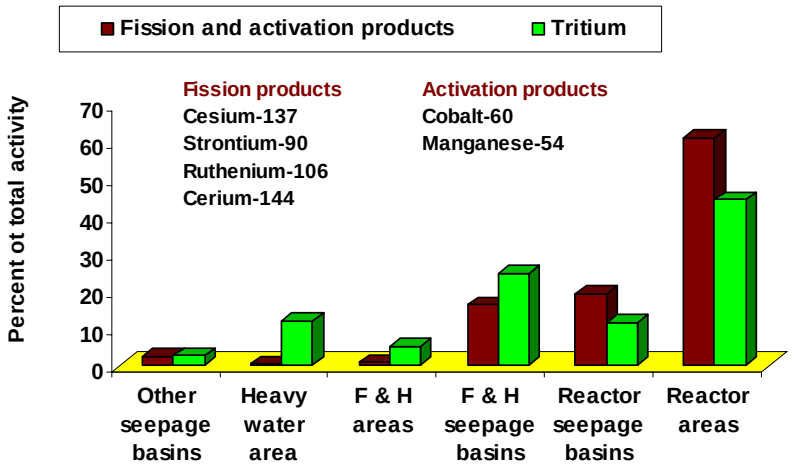

Figure 5-12. Distribution of tritium and beta activity (fission and activation products) in liquid wastes from six sources onsite. These percentages were based on operations in 1965–1969 (Du Pont 1966, Du Pont 1967, Du Pont 1968, Marter 1970).

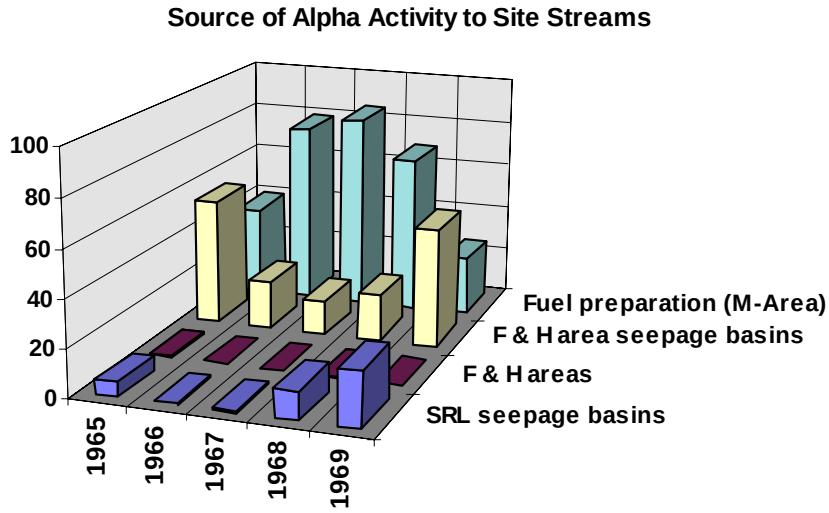

Figure 5-13. Sources of gross alpha activity in liquid wastes from six areas onsite. These percentages were based on operations in 1965–1969; however, in general, they reflect the pattern at other times as well. Roughly 60% of the alpha activity in liquid waste came from the M-Area. Another 30% originated from the F-Area and H-Area seepage basins and less than 10% came directly from the F-Area and H-Area and from the Savannah River Laboratory seepage basins.

For surface water source terms, we provided release estimates for the key radionuclides that were released offsite, which means the point that the effluents drain into the river. Unlike atmospheric source term development where releases are estimated from the point of release from a particular facility into air, the liquid effluents were released from facilities to seepage basins or to onsite streams, and still well within the site boundaries. Using effluent data measured directly at the point of release from the facilities is not appropriate if the goal is to provide estimates of release with uncertainty to the offsite environment where the public would have access to the surface water. This point of pubic access is the Savannah River and along the Lower Three Runs Creek to the southeast. Using measurements of radionuclides at points of release from the facilities is not suitable because the onsite streams flowed several miles before reaching the river.

An example of an ideal situation for surface water source term development might be having liquid effluents discharged from the facilities through a pipe directly into the Savannah River. In that case, measurements taken at the effluent release point to the river would be well suited for estimating releases from the site before any dilution occurred in the river. In the actual situation at the SRS, radionuclides were routinely measured onsite at several locations along each stream and then at several locations in the river. For the source term work at SRS, we reasoned that the last measurement point onsite would be the best data to use as the starting point. This location was at Road A where it intersected each stream. At these locations, a large quantity of data was available historically for all site streams. Measurements at several locations along Lower Three Runs had also been reported over the years. We used the data obtained at the Road A locations (final sampling point before discharge from the Site) on the five Site streams as the basis for our source term development. However, it was clear that some of the factors that might affect the release estimates that are listed above had not been evaluated quantitatively. The hold-up time in the swamp, and sampling and analytical procedures were the major components of the uncertainty analysis. The Water Releases workbook contains the details of the uncertainty calculations.

Effect of the SRS Swamp

The fact that the final measurement points of radionuclides in surface streams were located east of the Savannah River Swamp has important implications for the computation of the surface water source terms. Several studies help to understand the effect the swamp has on the discharge of certain radionuclides to the river. Beyond Road A, the onsite streams can meander for 4 to 5 mi or more onsite before discharging into the Savannah River. Most importantly, it is necessary to evaluate the impact of the Savannah River Swamp on retaining certain radionuclides, at least for a period of time, and thereby, changing the rate they move offsite into the Savannah River. On the other hand, periodic flooding of the swamp also affects the levels of radionuclides that are flushed from the swamp into the river.

Some of the key elements we considered and incorporated into the uncertainty estimates for releases to surface water are:

- The SRS swamp borders the Savannah River for about 16 km (10 mi) and averages about 2.4 km (~1.4 mi) wide
- The SRS swamp flooded about 20% of the time
- Elements are absorbed or retained by soils differently; a measure of this retention in soils is called the K_d value (See Table 5-7).

Clearly, the onsite streams and swamp play an important role in receiving, transporting, removing and diluting materials before they are released offsite in the Savannah River. Immobilization in soils plus radioactive decay reduce the amount of some contaminants entering the river. The Site conducted various studies on the manner in which the SRS swamp might act to remove and/or hold certain radionuclides for a period of time, or to prevent them from entering the Savannah River soon after being released from processing areas onsite. For example, the absorption distribution coefficient, or K_d value, is a measure of how materials are retained in soils. In one study at the SRS, K_d values varied from 50 mL g^{-1} for ^{90}Sr to 1000 mL g^{-1} for ^{137}Cs to 100,000 mL g^{-1} for ^{239}Pu (Marter 1969). Table 5-7 summarizes the key elements of that study. For soluble radionuclides, like tritium, the SRS swamp does not have a great impact on the levels released offsite; however, for more insoluble materials, like ^{137}Cs, the swamp influences the calculations of release estimates a great deal.

Table 5-7. Fraction of Radionuclides Released to Fuel Reprocessing Area Seepage Basins Reaching Four Mile Creek[a]

Isotope	Half life (y)	K_d at pH 1[b] mL g^{-1}	Fraction reaching creek from F-Area	Fraction reaching creek from H-Area
^{3}H	12.3	0	0.25[c]	0.4[c]
^{60}Co	5.2	100	0.1	0.5
^{90}Sr	27.7	50	0.7	0.9
^{106}Ru	1	800	1×10^{-16}	1×10^{-9}
^{134}Cs	2.1	1000	1×10^{-5}	0.01
^{137}Cs	26.6[e]	1000	0.2	0.7
^{147}Pm	2.5	100	0.01	0.3
^{239}Pu	24,000	10,000	0.9	0.9

[a] Selected from Marter (1969); partial listing of most significant radionuclides.
[b] Adsorption distribution coefficients reported in Marter (1969).
[c] Assuming 50% loss by evaporation.
[d] Through 1969, the Site reported that the only radionuclides reaching surface streams from the Fuel Reprocessing Area Seepage Basins were ^{3}H and ^{90}Sr. All other radionuclides shown, with the exception of ^{239}Pu, have been detected in ground water in the vicinity of the basins.
[e] The half-life of ^{137}Cs is 30.0 years (Schleien 1992).

Researchers at the Site carried out a number of studies to estimate the impact of the swamp, sediment retention and decay on the quantities of radionuclides that actually reached the river. They inferred that the advantages of seepage basin disposal over direct disposal to surface streams were that some ionic species were immobilized by the soil while others were greatly delayed in movement. They indicated that immobilization plus radioactive decay greatly reduced the amount of radioactive material eventually leaving control of the SRS (Evans et al 1968).

Retention in the streams and swamp

Radionuclide monitoring was quite extensive in the Site streams above the Road A location, but there were sparse measurements made between the Road A location and the Savannah River. Furthermore, the flow of each stream through the swamp is unique and not well understood. The onsite streams meander for 3–12 km (2-8 mi.) onsite before reaching the Savannah River Swamp System, which borders the Savannah River for about 16 km (10 mi) and averages about 2.4 km (1.5 mi) wide. Effluents from the Site flow through the swamp before discharging offsite into the Savannah River and are affected by seasonal variations water levels (Westinghouse 1996). Flows in the Savannah River are highest in the winter and spring, and lowest in the summer and fall. For these reasons, a simpler model was a more appropriate alternative than a complex model with more unknown parameters.

We chose a simple model to calculate the concentration of dissolved radionuclides and radionuclides attached to suspended and bottom sediments in the swamp (Jirka et al. 1983) because of the limited measurements of radionuclide in the surface water and sediments of the SRS swamp available historically. The model uses an unsteady, one-dimensional, liquid pathway submodel to calculate temporal and longitudinal distribution of dissolved radionuclide concentration as well as the concentration of radionuclides attached to suspended and bottom sediments of various sizes. The dissolved radionuclide concentration at a given location is found by applying the mass conservation equation with radioactive decay:

$$C_{x,t} = \frac{1}{Q_{x,t}} Q_{(x-\Delta x,\, t-\Delta t)} C_{(x-\Delta x,\, t-\Delta t)} e^{-\lambda \Delta t} \qquad (5\text{-}1)$$

where

$C_{x,t}$	=	dissolved radionuclide concentration at location *x* and time *t*,
$Q_{x,t}$	=	flow rate at location x and time t,
λ	=	radionuclide decay
$\Delta_{x,t}$	=	travel time in stream (s)
Δx	=	change in location (m)
x	=	distance (m)
t	=	time (s)

The travel time in the stream ($\Delta_{x,t}$) is the time it takes the radionuclide to move from its last point of measurement (e.g., Road A) to its entry into the Savannah River. For example, for Four Mile Creek this distance is about 12 km or 12,000 m. The travel time depends upon the flow rate and velocity of the particular stream. The final activity of the radionuclide entering the Savannah River is affected by the radioactive decay that might occur during this travel time.

The concentration of radionuclides attached to sediment (C_p) is calculated from the known dissolved radionuclide concentration and the distribution coefficient (K_d). The distribution coefficient, K_d is a measure of the amount of radionuclide sorbed on sediment.

$$K_d = \frac{\text{amount of radionuclide sorbed on sediment}}{\text{amount of radionuclide left in solution}} \qquad (5\text{-}2)$$

The K_d value for each radionuclide depends on various parameters, including sediment type and concentration, the flow characteristics, and water quality of the surface water and the contact time (Whicker and Schultz 1982). Table 5-8 gives the range and median values that have been reported in the literature for the elements of interest for releases from the SRS. In our source term calculations we assumed a lognormal distribution of K_d values for each element with the given median estimate and the range as the 5th and 95th percentiles of that distribution. We applied these equations to our source term calculations for releases of cesium and strontium in surface water releases from the SRS.

Table 5-8. K_d Values for Elements in Freshwater[a]

Element	Range (mL g^{-1})	Median (mL g^{-1})
Cesium	50–80,000	10,000[b]
Cobalt	1,000–71,000	5000
Iodine	0–75	10
Plutonium	100–10,000,000	100,000
Strontium	8–4000	100
Tritium	0	0

[a] From Jirka et al. 1983.
[b] Studies with cesium show an increase in K_d values with contact time; we assume a median value of 10,000 because of the longer contact period in the SRS swamp than in a flowing stream (Whicker and Schultz 1982).

In the following section, we illustrate the method using an example from 1961 for ^{137}Cs releases in four Mile Creek. This example uses the following assumptions and measurement data:

- Average concentration of ^{137}Cs at Road A in Four Mile Creek was about 10 pCi L^{-1} (see Table 5-11).
- The distance from the Road A location on Four Mile Creek to the Savannah River is about 12 km (12,000 m).
- At the Road A location, the average flow rates ranged from 270-490 cfs, with an average of 325 cfs or 9.2 m^3 s^{-1}. The SRS facilities contributed significant flow to the Site streams, as noted previously. For Four Mile Creek, the average flow rate upstream of the SRS effluent discharge point is approximately 0.35 m^3 s^{-1}. During the periods of intense operations at the Site, the flow rate in Four Mile Creek was increased to 10 m^3 s^{-1} ± 3 m^3 s^{-1} (340 ±90 cfs) at the Road A location. By about 1987 the flow rate in Four Mile Creek at Road A had returned to its historic flow rate of 0.5 m^3 s^{-1}.
- The average velocities varied from 0.03 to 0.08 m s^{-1} from the late 1950s through the mid 1960s (see Hydrology section). The average velocity in Four Mile Creek in the late 1950s through the mid 1960s was about 0.06 m s^{-1}.
- The half-life of ^{137}Cs is 30.0 y, so the decay rate (λ) is 7.3 x 10^{-10} per second (equal to (ln 2)/ half-life).
- For cesium, we assume a median K_d value of 10,000 mL per g^{-1} (Table 5-8) (Jirka et al. 1983).

Thus, in the early operational years, the travel time in Four Mile Creek,

$$\Delta t = x/t$$
$$= 12{,}000 \text{ m}/0.06 \text{ m s}^{-1} = 200{,}000 \text{ s (or ~55 hr)} \quad (5\text{-}3)$$

So the dissolved ^{137}Cs concentration 12 km downstream from Road A is calculated using equation (5-4):

$$C_{12\ km,\ FMC} = \frac{1}{(9 \text{ m}^3 \text{ s}^{-1}) + (0.35 \text{ m}^3 \text{ s}^{-1})} (9 \text{ m}^3 \text{ s}^{-1})\ (10 \text{ pCi L}^{-1}) e^{-7.3 \times 10^{-10} \text{ s}^{-1}\ 200{,}000 \text{ s}} \quad (5\text{-}4)$$

$$C_{12\ km,\ FMC\ water} = 9.6 \text{ pCi L}^{-1}$$

The results of this calculation show that the radioactive decay of ^{137}Cs as it travels the 12 km downstream plays a minor role in removing ^{137}Cs from the source term. The concentration of radionuclides attached to sediment (C_p) is then calculated from the known dissolved radionuclide concentration and the distribution coefficient (K_d) by:

$$C_{px,t} = K_d \times C_{x,t}. \quad (5\text{-}5)$$

$$C_{p\ 12\ km\ FMC} = 10{,}000 \text{ mL g}^{-1} \times 9.6 \text{ pCi L}^{-1} = 96 \text{ pCi g}^{-1}$$

The sediment transport rate, S_T, is found by knowing the flow rate and using constants for the particular sediment (Jirka et al. 1983):

$$S_T = aQ^b \quad (5\text{-}6)$$

The parameters, *a* and *b*, are constants that are estimated for each sediment size range. We assume that a = 0.0004 and b = 3, based on studies at the SRS (Ruby et al. 1981). Then the sediment transport rate becomes:

$$S_T = 0.0004\ (9 \text{ m}^3 \text{ s}^{-1})^3 \quad (5\text{-}7)$$
$$S_T = 0.29 \text{ g s}^{-1}$$

If we assume that the sediment carries all the particulate ^{137}Cs, then the sediment carries:

$$S_T \times C_{px} = 0.29 \text{ g s}^{-1} \times 96 \text{ pCi g}^{-1} = 28 \text{ pCi s}^{-1} \quad (5\text{-}8)$$

And the rate of dissolved ^{137}Cs carried downstream is:

$$C \times Q = 9.6 \text{ pCi L}^{-1} \times 9 \text{ m}^3 \text{ s}^{-1} \times 1000 \text{ L m}^{-3} = 86{,}400 \text{ pCi s}^{-1} \quad (5\text{-}9)$$

Finally, the total amount of ^{137}Cs being transported in FMC is the sum of dissolved and particulate ^{137}Cs so that

$$28 \text{ pCi s}^{-1} + 86{,}400 \text{ pCi s}^{-1} = 86{,}428 \text{ pCi } ^{137}\text{Cs s}^{-1} \quad (5\text{-}10)$$

With the release rate of 86,400 pCi s^{-1} for ^{137}Cs in Four Mile Creek water to the Savannah River, then over the period of a year, the total release to the river would be 2.7 Ci. The ^{137}Cs activity in sediment during that same year would be 880 mCi or 0.00088 Ci. This example does not incorporate the uncertainty distributions, which our source term estimates do. This model was applied to the estimation of annual release estimates for ^{137}Cs and ^{90}Sr. As described in the following sections, other sources of uncertainty were also considered.

Periodic flooding of the swamp

The SRS Swamp flooded about 20% of the time (74 days per year on the average) during 1958 to 1967. Historically the river overflowed its channel and flooded the swamps bordering the Site when its elevation rose higher than 27 meters above mean sea level at the SRS Boat Dock. This level corresponds to flows equal to or greater than about 438 cubic meters per second (or, about15,500 cfs). We have assumed that additional releases to the Savannah River occurred from the swamp when flooding occurred. We treat this uncertainty as a source of bias and increase releases for radionuclides that were retained in the swamp, like cesium and strontium, by 20% (with a range of 10%-30%) for most years. Because detailed records exist for rainfall amounts, this percentage varies. For years with very high rainfall amounts like 1964 and 1971, we assume a value of 40% (with a range of 25%- 60%. For years with low rainfall, we assume the swamp flooded only about 10% of the time (with a range of 5%-15%).

Sampling and Analytical Procedures

Sampling and analytical uncertainties are important to consider for all key radionuclides, but they are especially important for radionuclides, like tritium, that are not impacted heavily by flow through the SRS swamp. In this case, the sampling and analytical uncertainties are the major sources of uncertainty in the release estimates for tritium in surface waters. Nevertheless, the uncertainty associated with the release estimates must be considered for any radionuclides in surface water from the analytical errors in measurement of flow, and in sampling and determination of the radionuclide concentrations in the water. Appendix A describes the analytical and sampling procedures in detail.

Generally, there were differences of 10% or less in the volume going into the Site streams from the reactor basins. It appeared that the effluent volume to the Site streams was monitored reasonably well by both the Site and the USGS (Ashley 1959, 1964, 1967). Estimates of error for the routine concentration measurements varied with the radionuclide, the sample preparation and with the counting procedure. Nevertheless, SRS provided extensive documentation of its health physics sampling and analytical procedures over the years, both in standard procedures manuals

(Du Pont 1953, 1959, 1989) and also in frequent memoranda and interim reports (Albenesius and Meyer 1962, Ashley 1967a).

Table 5-9 summarizes the various factors that were considered in calculating revised release estimates of releases of tritium, ^{137}Cs, and ^{90}Sr in liquid effluents to Site streams that eventually entered the Savannah River. Several time periods are considered because of changes in the sampling procedures and analytical methods for the radionuclides. In addition, the retention within the swamp varied among the radionuclides and those factors were considered, as described above.

Table 5-9 Factors Considered in the Uncertainty Estimates for Surface Water Source Terms for Key Radionuclides

Radionuclide	Time period	Measured effluent release	Measured in stream at Road A	Estimated measurement uncertainty	Effect of swamp Kd values[a]	Effect of swamp Flooding
Tritium	1954-1957	yes	no	50%	none	none
	1958-1959	yes	yes	40%	none	none
	1960-1973	yes	yes	25%	none	none
	1974-1992	yes	yes	15%	none	none
^{137}Cs	1954-1958	no	no	60%	10,000	1.1 to 1.4[b]
	1959-1973	yes	yes	50%	10,000	1.1 to 1.4[b]
	1974-1992	yes	yes	25%	10,000	1.1 to 1.4[b]
^{90}Sr	1954-1960	some	no	60%	100	1.1 to 1.4[b]
	1961-1973	yes	yes	50%	100	1.1 to 1.4[b]
	1974-1992	yes	yes	25%	100	1.1 to 1.4[b]

[a] The median estimate is listed; the range of K_d values is: cesium (50–80,000) and strontium (8–4000).

[b] Based on rainfall amounts for each year; in general the swamp flooding about 20% of the time each year when the average rainfall was about 47 inches. 1964 had very heavy rainfall (73 in) and extensive flooding occurred, while 1954 was quite dry (28 in) with little swamp flooding.

TABULATION OF RELEASES

Releases of Beta-Gamma Emitters

Chapter 3 of this report describes our methods of selecting and ranking the key chemicals and radionuclides for in-depth source term development. Section 3.2 describes the results of our screening and ranking of radionuclides using the NCRP screening methodology (NCRP 1996). The results of that process showed that the for releases to surface water, the radionuclides, ^{137}Cs, tritium, ^{90}Sr, ^{60}Co, ^{32}P, and ^{131}I accounted for about 95% of the screening value to people living near the SRS. All of these radionuclides decay by beta emission. Furthermore, ^{137}Cs and tritium dominate the dose contributions for most major pathways. For that reason, we have focused most effort in the following sections on reconstructing releases of the beta emitters, tritium, ^{137}Cs, and ^{90}Sr. Release estimates are also described for ^{60}Co, ^{32}P, ^{131}I and uranium, which decays by alpha emission.

Tritium Release Estimates

The key processes potentially leading to tritium releases at the SRS are the:

- Reactor operations at the reactor facilities
- Recovery of transuranic elements in the separations facilities
- Recovery of tritium in the tritium facilities
- Laboratory research area
- Heavy water rework facility.

Tritium as a site product for SRS was created in the reactors by neutron capture in lithium-6 in the lithium/aluminum targets and control rods. This capture produced tritium in a concentrated form, which was then extracted and packaged for use for the weapons program. Most releases of this tritium product occurred as releases to the air during the extraction and packaging operations in the Tritium Facilities located in H-Area and constituted the majority of atmospheric releases from the SRS, ranging from 200,000 Ci to 520,000 Ci per year (see Chapter 4.1)

Tritium was also produced in SRS reactor moderator by activation of deuterium. The concentration of tritium in the heavy water was a function of the neutron flux in the reactor and the length of the irradiation, with the tritium concentration in the moderator building up slowly over the years. Some of this tritium was lost to air and to liquid effluents by evaporation of moderator leaks and by the carry over of tritium oxide on fuel and target elements during reactor discharge. Releases of tritium to air from each reactor were as high as 60,000 Ci per year depending on the tritium concentration in the moderator, roughly 10% to 25% of tritium releases to air from the Separations Area.. Direct releases of tritium to air from routine 400-D operations were about up to 2500 Ci per year (compared to about 200,000 Ci to 520,000 Ci of tritium from the Separations Area).

Direct releases of tritium to plant streams and the Savannah River from all SRS reactor areas ranged from about 5000 Ci in the 1980s up to 100,000 Ci in the 1960s. Tritium releases directly from the 400-D facilities to Beaver Creek Dam from the 400-D operation have ranged from about 2500 Ci in the 1980s to about 6000 Ci in the 1960s. Our approach to determining these overall release estimates for tritium is the topic of this section.

Tritium was released to onsite streams from numerous activities, but the largest quantities were primarily from reactor operations and associated activities (Figure 5-12). Tritiated water was released in liquid effluents directly to onsite streams and to the seepage basins, and from outcropping into streams from burial ground releases and shallow groundwater aquifers. Continuous proportional samples were taken of water leaving the separations areas to the seepage basins and to Four Mile Creek. A fraction of the composite sample was analyzed weekly with Beckman Model liquid scintillation counters having a tritium sensitivity of 300 pCi L^{-1} (± 10%) and calibrated with National Bureau of Standards samples (Jacober et al. 1973). Cooling water discharged to seepage basins and to the streams from the tritium facilities was also monitored but results showed very low concentration of tritium compared to other facilities onsite.

Releases of tritium from the reactor areas to onsite streams were monitored daily since the mid 1950s, and reported in Site documents such as aperture card printouts of weekly volume and concentration measurements of tritium. Table 5-10 summarizes the onsite tritium monitoring of releases to onsite streams and seepage basins through the 1970s. The Site made progress on

increasing the sensitivity of the tritium analysis, especially for large volume samples. Appendix A of this report contains a thorough description of these studies.

The basic tritium analytical procedure used the "tri-carb" liquid scintillation counter, which was placed in operation by June 1958 (Du Pont 1958). (See Appendix A). By September 1962, the analytical procedure used for tritium analysis was fairly well established at the SRS. For water samples, an initial detection limit of 5000 pCi L^{-1} with a counting time of 30 minutes was established. By the 1980s, liquid scintillation counting was still used for tritium determination in water but the recoveries were near 100% and the sensitivity or limits of detection were from 300 to 1000 pCi L^{-1} (Du Pont 1989).

Using these methods, tritium concentrations in water samples were determined from approximately 20 locations in onsite streams. There was extensive monitoring of tritium from facilities and at numerous locations in the Site streams. Tritium concentrations in the Site streams and streams flow rates were measured on a weekly basis beginning in 1959. These values were combined to provide estimates of tritium activity (in Ci) that were reported weekly or monthly.

Table 5-10. Tritium Monitoring of Liquid Effluent Releases at the SRS through the Mid-1970s[a]

Area	Emission point	Type and frequency of monitoring
Reactor areas	Disassembly basin	Once per day
	Thermal shield	Grab sample
	Heat exchanger	Once per week or more often
	Moderator spills	As they occurred
	Distillation cooling water	Daily
Separations areas		
To seepage basins	234-H cooling water	Grab sample weekly
	232-G lab drain	Grab sample weekly
	232-H floor drain	Grab sample when releases occurring
To streams	232-H cooling water	Grab sample weekly
	232-H floor drains	Grab sample weekly
	238-H cooling water	Grab sample weekly
	238-H floor drains	Grab sample weekly
	234-H cooling water	Grab sample weekly
	236-H floor drains	Grab sample weekly
Savannah River Laboratory	776-A low level system hold tank	Monthly composite sample analyzed
Heavy Water Plant	772-D	Continuous
	420-D waste water	Each drain sampled and analyzed
	421-2D	Hold tank grab sample

[a] Adapted from Towler (1980).

Correlation of Radionuclide Releases with Reactor Power Levels. The radionuclides released to air and water from the SRS resulted primarily from operations of the five reactors and separations areas. For times when radionuclides were not routinely measured in the effluent, it is reasonable to suppose that releases of some radionuclides could be correlated with power levels

or production operations at these facilities. For example, because radionuclides released to surface water originated primarily from the reactors, it seemed reasonable to correlate releases of radionuclides that result from reactor operation (such as fission and activation products) with the duration and power levels of each reactor. This correlation between power levels and measurement values could form the basis of a method to calculate releases of particular radionuclides that were not measured during certain time periods, especially in the 1950s. [The power levels for the five operating reactors at the SRS were compiled for all years of operation. Refer to Chapter 2, *SRS Facilities: History of Processes and Operat*ions, for details.]

Figure 5-14 shows the variation with time of the reactor power levels and the total tritium activity measured at Road A in the Site streams. The reactor power levels decline from about 250 to 300 gigawatt days (GWd) in the early 1960s to about 100 GWd in by 1970. Tritium losses to the Site streams averaged about 500 to 7000 Ci mo^{-1}; however, they varied widely with releases exceeding 20,000 Ci in February 1963 and in May, June and August 1964. This comparison shows that the total tritium activity measured at Road A in the Site streams does not correlate positively with reactor power levels. The Site noted that releases of radioactivity to the reactor area seepage basins and the Site streams were not easily correlated to operating time periods because many releases of liquid effluents occurred from the storage basins when the reactors were not operating (Evans et al. 1992).

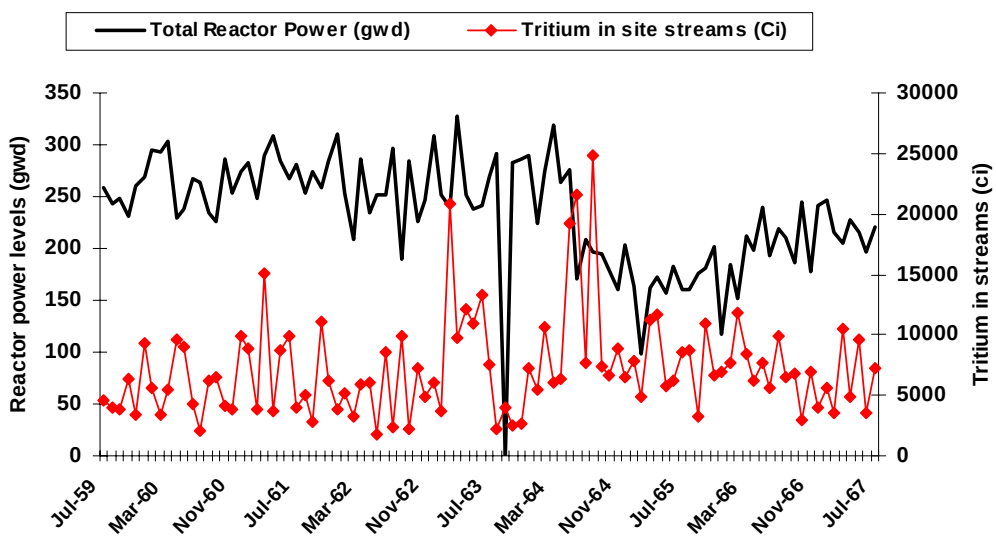

Figure 5-14. Variation with time of the reactor power levels and the total tritium activity measured at Road A in the Site streams. (See Chapter 2 for data source).

Tritium Release Data. We calculated the total tritium discharged each week to the Site streams multiplying the weekly tritium concentration (picocuries per liter) and the total flow measured that week. The datasets are available in the electronic Water Release Excel file. The tritium release data are presented in the following order:

1. data from the various streams will be compared to provide perspective on which Site streams had the highest levels of tritium.
2. the monthly time trends of tritium in Site streams reflect production trends and unplanned events or spills that occurred onsite.

3. we combined the monthly totals of activity measured at Road A for all Site streams with uncertainties to provide our annual release estimates.

Figure 5-15 shows the variation in weekly tritium concentrations measured in Four Mile Creek at the Road A sampling location. The figure shows tritium concentrations measured from July 1959 through December 1963 ranging from about 10,000 to 100,000 pCi L^{-1}, with peaks up to 1,000,000 pCi L^{-1} (Ashley 1959). For comparison, the broken lines indicate the general range of average background tritium levels measured in surface water throughout the U. S. during this period. Tritium levels were higher during that period throughout the U.S. because of fallout from weapons testing (see Chapter 6). Figure 5-16 covers a shorter time period, from 1962 through 1964, and compares tritium concentrations in Steel Creek with those measured in Savannah River during the same time. Tritium concentrations ranged from about 20,000 to 100,000 pCi L^{-1}, while those in the Savannah River were lower, 3000 to 30,000 pCi L^{-1}. This figure shows another point of reference, the current U.S. Environmental Protection Agency drinking water standard for tritium of 20,000 pCi L^{-1} (or, 20 pCi mL^{-1}) (Zeigler et al. 1985). This is an example of the type of analytical data that the SRS collected and maintained. We collected and used these types of measurement data along with flow rate measurements in these streams for reconstructing historic releases of tritium from the SRS.

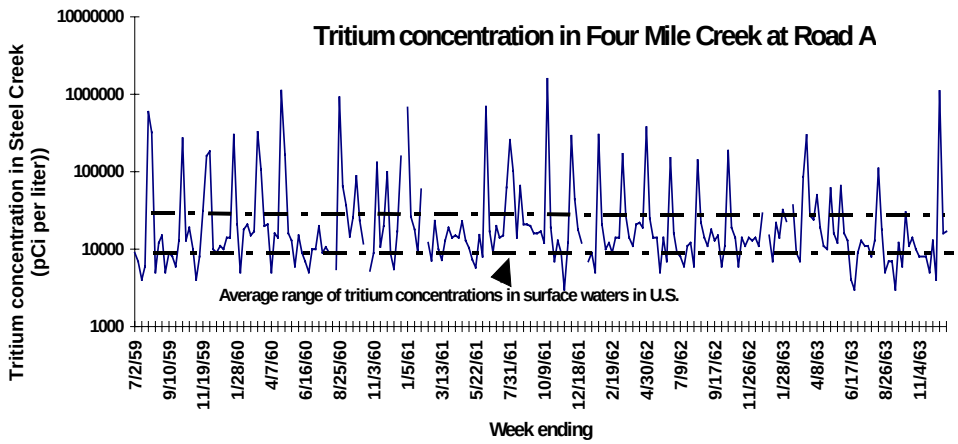

Figure 5-15. Weekly tritium concentrations measured in Four Mile Creek (FMC) at Road A (sampler location #6) from July 1959 through December 1963. This location was the final monitoring point on the FMC before it flowed into the Savannah River. The dashed lines represent the range of average tritium concentrations measured in surface waters around the U. S. during this time (Ashley 1959, Murphy and Carlton 1991).

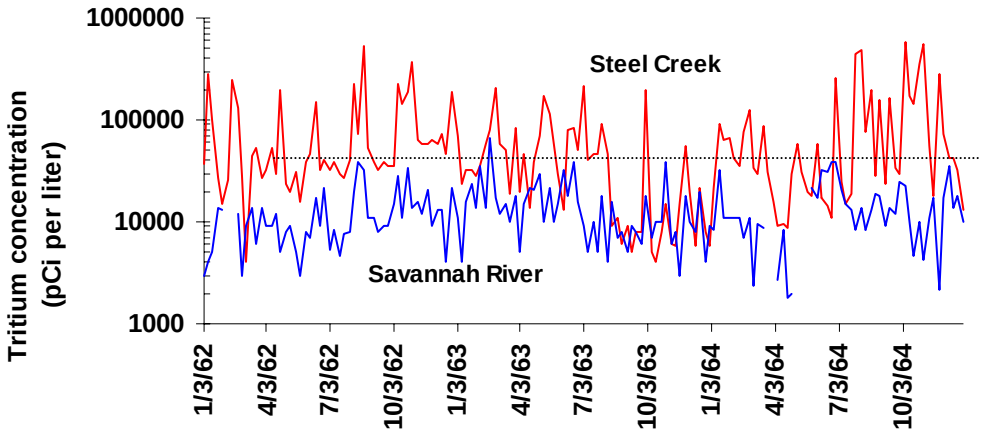

Figure 5-16. Weekly tritium concentrations measured in Steel Creek at Road A and in the Savannah River downstream of the SRS at U.S. Highway 301 from January 1962 through December 1964. The dashed line represents the current U.S. Environmental Protection Agency drinking water standard, and is added for reference purposes (Zeigler et al. 1985).

When the tritium concentrations are combined with the water volume data, then total activity can be compared among the Site streams. In Figure 5-17, the weekly tritium activities, plotted for the five major Site streams, show that the highest tritium activities were generally present in Four Mile Creek, Pen Branch, and Steel Creek. Historically, the Upper Three Runs and Lower Three Runs Creeks had lower levels of activity. These values are based on measurements taken at Road A, the final sampling location before the streams flowed into the Savannah River. Total tritium levels to streams ranged from about 5,000 to 12,000 Ci per month during this time. Par Pond received liquid effluents from the R and P Reactor areas beginning in 1958 and thus prevented their direct release to the surface streams.

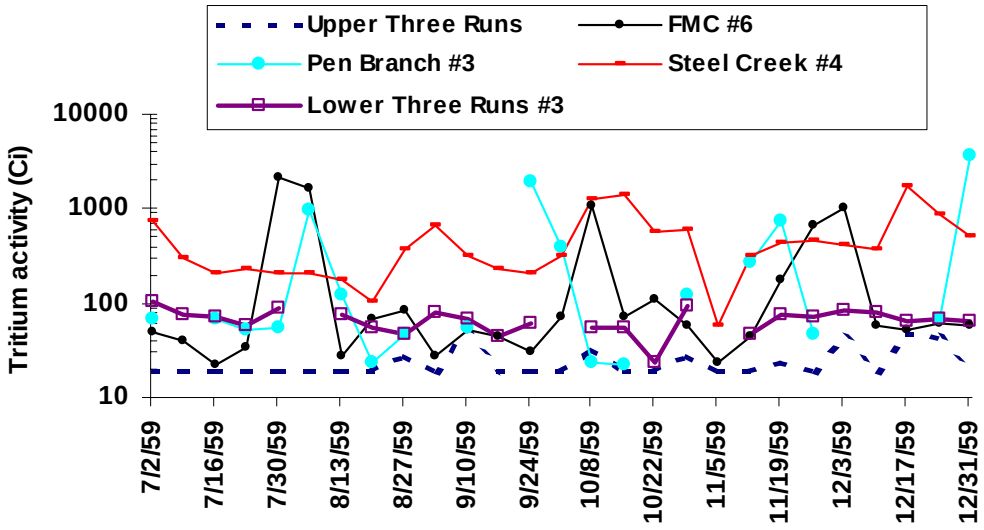

Figure 5-17. Tritium activity in the five Site streams at the SRS measured weekly beginning in the late 1950s. The pattern of higher tritium levels in Four Mile Creek, Pen Branch, and Steel Creek was seen throughout most of the operational history of the Site. In general, radioactivity levels in the Upper Three Runs and Lower Three Runs Creeks were lower. The figure establishes that Steel Creek, which received effluents from the reactor areas, and Four Mile Creek, which received effluents form the separations area, C Reactor area, and some from the D-Area, historically had the highest tritium activities.

Figure 5-18 shows this pattern over a longer period of time and in a different way. The pie chart shows that over 35% of the total tritium activity measured in transport in Site streams was found in Steel Creek, which received effluents from the L and P Reactors. Pen Branch and Four Mile Creek each carried roughly 25% to 30% of the activity. The Upper and Lower Three Runs carried much lower levels. Upper Three Runs historically transported lower levels of tritium because it received discharges only from the north F-Area (Du Pont 1966, Du Pont 1967, Du Pont 1968, Marter 1970).

The SRS began accounting for the amount of tritium released when it began operations in 1953. Actual measurements of the amount of tritium in transport in SRS streams and the Savannah River began in the latter half of 1959. There are 2 broad types of tritium releases from the SRS: (1) direct releases from the SRS facilities themselves and (2) migration of tritium mainly from the seepage basins in the F-area and H-area, the burial ground, and the K-Area containment basin. In the early years almost 100% of the tritium releases to streams was from direct releases. Figure 5-19 shows that releases due to tritium migration from the F-Area and H-Area seepage basins became more important in terms of accounting for releases to the Site streams and eventually to the Savannah River. Since the mid 1970s migration and outcropping to streams have accounted for most of the SRS tritium releases to surface water (Zeigler et al. 1985, Murphy et al 1991, Murphy and Carlton 1991). The Site has reported fairly good agreement in the inventory of tritium measured at the point of release, in the plant streams before entry into the river and in the Savannah River.

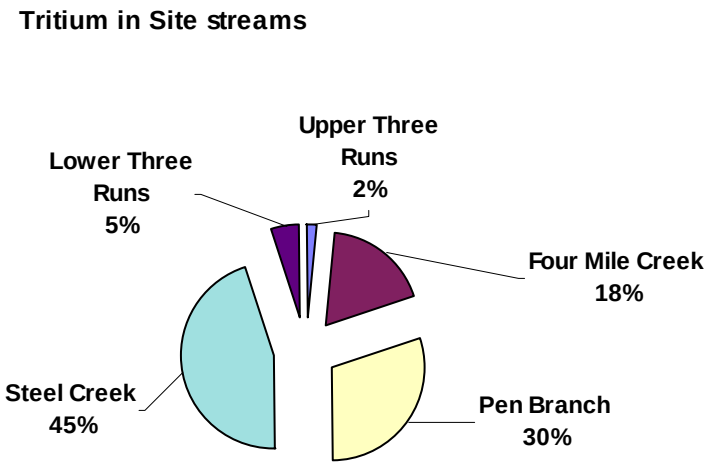

Figure 5-18. The relative amount of total tritium activity in the major Site streams, based on weekly measured values from 1959-1967 in the streams at the last onsite location before the streams emptied into the Savannah River. During this period, Steel Creek, Pen Branch, and Four Mile Creek carried similar levels of tritium in the water, while Upper Three Runs and Lower Three Runs (LTR) water had much lower tritium activity. One reason for lower levels in Lower three Runs was that liquid releases from the P and R Reactors were discharged to Par Pond instead of directly to LTR after 1958 Area (Du Pont 1966, Du Pont 1967, Du Pont 1968, Marter 1970).

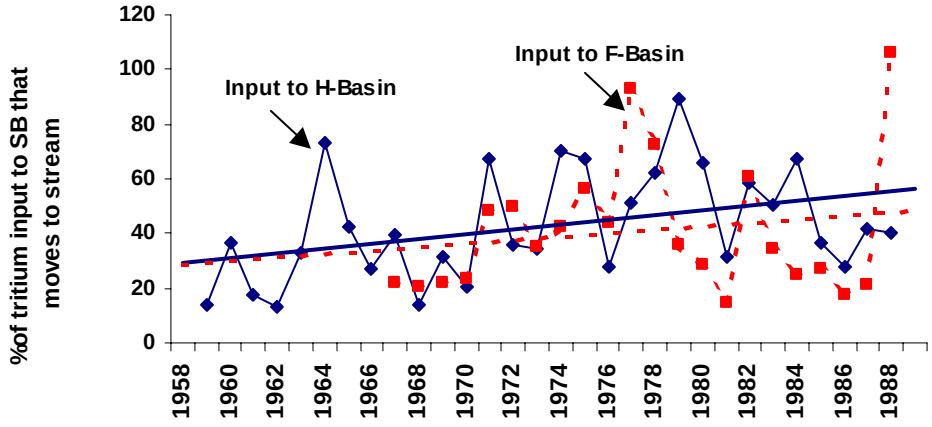

Figure 5-19. Gradual increase in tritium activity seeping from the F-Area and H-Area seepage basins to the Site streams as a percentage of the original input. Migration from the seepage basins and outcropping to Site streams has been the major source of tritium releases in more recent years after the reactor operations stopped.

We reconstructed releases from the SRS using the measured weekly values of concentration and flow rate. We considered several sources of uncertainty, with the analytical errors of sampling and analytical errors being the most important for tritium releases, as previously described in the Uncertainty section of this chapter. We used CrystalBall™ to account for those uncertainties in our calculations (Decisioneering 1995). All Excel spreadsheets with these calculations accompany this report. Figure 5-20 displays the results of our source term reconstruction for tritium, with associated uncertainty estimates. Annual median release estimates were 35,000 Ci or less before 1959 and after 1979. Over 80,000 Ci of tritium was released in 1961, 1963, 1964, 1965, 1966, 1967 and 1968. The highest median release of 121,000 Ci was in 1964 and the 5th and 95th percentiles of the distribution are 84,000 and 177,000 Ci. The median estimate of the total for all years is about 1,800,000 Ci with the 5th and 95th percentiles of the distribution of 1,300,000 and 2,500,000 Ci tritium. There is overall general agreement between our reconstructed tritium release estimates to the Site streams based on the original measurement data and supported by weekly and monthly reports from the 1960s and 1970s and the annual total reported by the SRS.

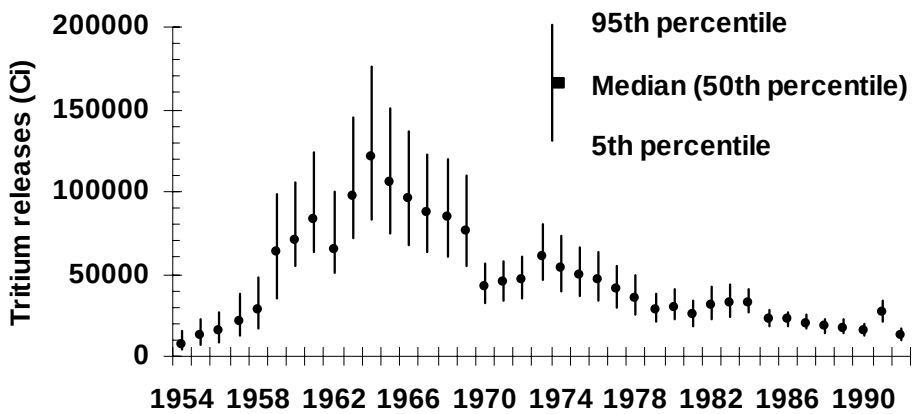

Figure 5-20. Estimates of tritium releases to surface water from the SRS with uncertainty estimates. Each year is represented by a vertical line that represents the 95th (top) and 5th (bottom) percentiles of the distribution of releases. The median or 50th percentile is shown as the filled shape in the center.

The time-dependent release pattern of tritium to surface water reflects the operations and events that were occurring onsite as shown in Figure 5-21. Tritium releases to surface water peaked in 1964 when all reactors were operating. Quite a large number of fuel failures and other unplanned events like spills occurred in the late 1950s through 1964, when the R Reactor was permanently shut down. In addition, the Site processed short-cooled fuel on several occasions during this time, which led to higher levels of radioactivity releases to the streams when the disassembly basins were purged (Murphy et al. 1991). This same pattern is seen with the releases of ^{137}Cs and ^{90}Sr. (see subsequent sections for details). In addition, rainfall levels affected releases of radionuclides in liquid effluents (Figure 5-22). Some of the largest rainfall amounts

occurred in 1964, which led to increased runoff and SRS swamp flooding. Whereas monthly average rainfall for the area is between 3 and 6 in, over twice that level fell in January (8 in), July (10 in) and August (12 in) 1964. Another factor that must be considered in putting Site releases into perspective is weapons fallout and the effect it may have had on contributing to radioactivity levels in the area. (See Chapter 6 of this report).

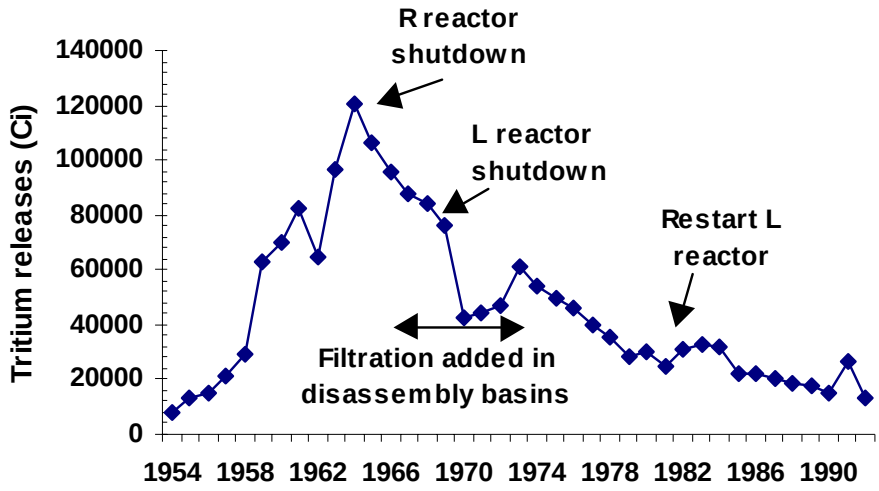

Figure 5-21. Comparison of annual tritium release estimates with some key operational events that affected releases of radioactivity to liquid effluents at the SRS (Du Pont 1958, Du Pont 1962, Du Pont 1966.

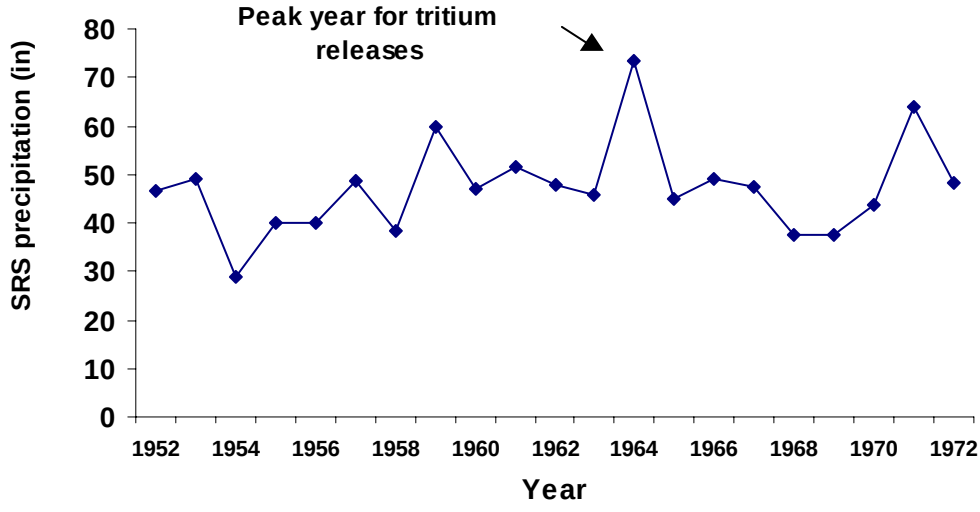

Figure 5-22 Annual rainfall levels in the SRS area from 1952 through 1972. The annual totals ranged from about 29 in. in 1954 to almost 75 in. in 1964, the year with the highest releases of tritium to Site streams.

Cesium Release Estimates

Most of the radiocesium at the SRS was formed as a byproduct of the nuclear fuel and targets during operation of the five production reactors. Many isotopes of cesium were produced in SRS reactors as activation products (e.g., ^{131}Cs, ^{132}Cs, and ^{134}Cs) and fission products (e.g., ^{136}Cs, ^{137}Cs, ^{138}Cs). Cesium-134 and ^{137}Cs are the most important for the purposes of offsite radiation exposure because of their longer half-lives of 2.1 years and 30 years, respectively. Of these, ^{137}Cs has had the greatest impact on the SRS environment. In the past, ^{137}Cs concentrations were usually measured in the:

- liquid effluent outfalls just downstream of the discharge point but before the discharged liquids drained into the Site stream
- streams at a number of locations, and
- Savannah River.

For source term reconstruction, we focused on measurements made in the Site streams and when available, in the effluent outfall.

Over the years, there were various steps taken in the reactor areas to minimize releases of radioactivity. In 1963, basin water was recirculated through the heat exchangers to be cooled and deionized to remove radionuclides, which reduced the discharge volumes considerably. Before 1963, basin water was continuously purged to Site streams. Later permanent sandfilters were installed to main water clarity. (see the reactor areas section of this chapter for details). Most of the radiocesium released in the early years originated in failed fuel elements. The largest ^{137}Cs releases appear to have occurred in 1957 in association with several fuel failures in the R Reactor. The Site monitored aqueous releases of cesium in several ways (Carlton et al. 1992a):

- Cooling water was monitored for beta-gamma activity beginning in 1955, and later, specific radiochemical analyses for radiocesium were done.
- The purge or wastewater from the disassembly basins was routinely analyzed for ^{137}Cs beginning in the mid 1950s
- Gamma spectrometry was introduced in the early 1960s; this technique along with the use of the ion exchange column enhanced the ability to detect low-levels of cesium in stream water (see Table 5-5).

The bases for our ^{137}Cs release estimates are the reported Site measurements made in the Site streams and in the effluent discharges from the facilities to the streams. As with the tritium measurements, weekly concentrations and flow rates were carefully tabulated for the source term reconstruction (see Tritium Release section in this chapter). The Site made weekly measurements of cesium activity at several locations in the Site streams beginning in the late 1950s. The reported concentration measurements and flow data from the aperture card printouts and from weekly records that Ashley (1959) compiled were the basis of our release estimates for ^{137}Cs. As shown in Figure 5-23 over 50% of the total ^{137}Cs activity measured in transport in the SRS streams was measured in Steel Creek, which received discharges from the L, P and R Reactors. Overall, each of the other streams contributed about 15% of the total quantity of ^{137}Cs.

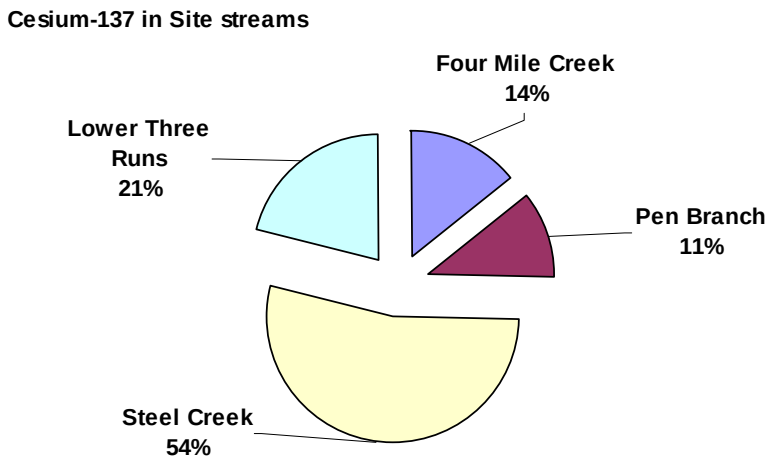

Figure 5-23. The relative amount of total cesium activity in the major Site streams measured weekly from 1959-1967 in onsite streams at Road A, the last onsite location before the streams emptied into the Savannah River. Over half of the ^{137}Cs activity released during this period was present in Steel Creek.

Radiocesium-specific measurements were made in the Site streams beginning in 1960. Before that time, only nonvolatile beta activity was measured in the preoperational survey and from the beginning of plant operations. To estimate ^{137}Cs releases for times when ^{137}Cs specific measurements were not made, we calculated a ratio of ^{137}Cs to nonvolatile beta activity when both measurements were made at the same time and same locations. We used this ratio, along with the nonvolatile beta activity measurements, to estimate levels of ^{137}Cs activity in the Site streams at Road A when ^{137}Cs -specific measurements were not made. Table 5-11 presents the data used to calculate the ratios, and the resulting average ratio and standard deviation for each stream that was used in the uncertainty calculations for cesium releases from the Site. For the annual release estimates for ^{137}Cs, there is more uncertainty associated with our release estimates prior to 1958 because the estimates are based on our calculated ratios and measured nonvolatile beta activity rather than on direct measurements of ^{137}Cs.

Figure 5-24 tracks the change in ^{137}Cs concentrations in the Site streams over time. From the time of the pre startup background survey (Reinig et al. 1953) through 1958, ^{137}Cs levels were higher in Lower Three Runs than in other Site streams that received discharges from the reactor areas. Prior to 1958 and the opening of Par Pond, water in the reactor disassembly basins was purged directly to the Site streams. This logarithmic scale shows a similar pattern of cesium concentrations in the Site streams from about 1958 when Par Pond was operational through the end of 1964. Then in late 1963 levels of ^{137}Cs in Steel Creek increased dramatically from about 25 pCi L^{-1} to a peak of almost 450 pCi L^{-1} in 1968 due to major spills and releases in the area. All reactor cooling water discharges to Steel Creek ceased when L Area reactor was shut down in February 1968.

Table 5-11. Ratio of ^{137}Cs to Nonvolatile Beta Activity in Site Streams from July 1958 through December 1962[a]

	Four Mile Creek (pCi L^{-1})			Pen Branch (pCi L^{-1})			Steel Creek (pCi L^{-1})			Lower Three Runs (pCi L^{-1})		
Date	NVB[b]	^{137}Cs	^{137}Cs/ NVB	NVB	^{137}Cs	^{137}Cs/ NVB	NVB	^{137}Cs	^{137}Cs/ NVB	NVB	^{137}Cs	^{137}Cs/ NVB
Jul-Dec 1958	110	11	0.10	470	7	0.01	120	12	0.10	130	38	0.29
Jan-Jun 1959	4300	14	0.00	640	6	0.01	300	16	0.05	74	19	0.26
Jul-Dec 1959	350	6	0.02	250	6	0.02	120	11	0.09	49	16	0.33
Jan-Jun 1960	400	7	0.02	360	8	0.02	530	8	0.02	20	8	0.40
Jul-Dec 1960	260	7	0.03	260	*7*	0.03	200	*9*	0.05	40	12	0.30
Jan-Jun 1961	260	14	0.05	190	6	0.03	220	14	0.06	24	9	0.38
Jul-Dec 1961	230	*9*	0.04	120	*13*	0.11	160	*10*	0.06	27	7	0.26
Jan-Jun 1962	120	12	0.10	540	10	0.02	230	26	0.11	50	11	0.22
Jul-Dec 1962	78	38	0.49	110	12	0.11	270	31	0.11	30.00	10.0	0.33
Average			0.11			0.04			0.07			0.31
Standard deviation			0.15			0.04			0.03			0.06

[a] The measurement results were reported monthly and semiannually in the Health Physics Regional Monitoring Semiannual Reports for those time periods; the ratio was used to estimate ^{137}Cs releases for early operational periods at SRS when only nonvolatile beta activity was measured.
[b] NVB = nonvolatile beta activity.

After that time, only L-Area and P-Area disassembly basin purges, undiluted by cooling water, were discharged to the creek. This action resulted in higher concentrations of radioactive materials (primarily ^{134,137}Cs from a few failed experimental fuel elements) in the stream water, even though the quantity of radioactive materials released to the creek began decreasing after 1968 (Marter 1970).

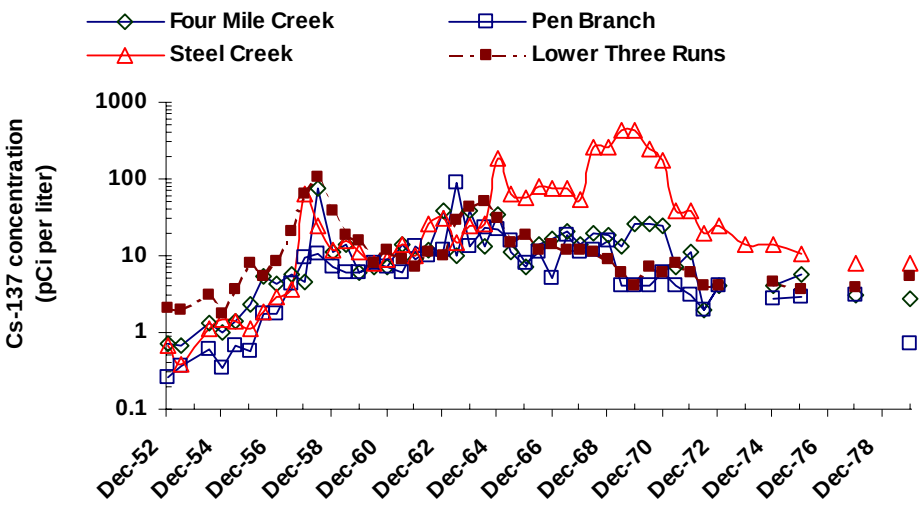

Figure 5-24. Semiannual average concentrations of ^{137}Cs in Four Mile Creek, Pen Branch, Steel Creek and Lower Three Runs measured at Road A. The figure shows a similar pattern of cesium concentrations in the Site streams from 1958 through 1964. From 1964 through 1975, the concentrations were much higher in Steel Creek than in the other Site streams.

These concentration measurements are then combined with weekly flow data to calculate the weekly reported releases of ^{137}Cs at Road A. Figure 5-25 shows the weekly releases of ^{137}Cs in the four main Site streams from April 1960, when measurements of ^{137}Cs began, through June 1967. Several peak releases are marked with the date and the stream where the high levels were measured. In June 1964, almost 50 Ci of ^{137}Cs were measured in Lower Three Runs Creek when a major spill occurred. These data are the basis of our source term calculations for ^{137}Cs releases.

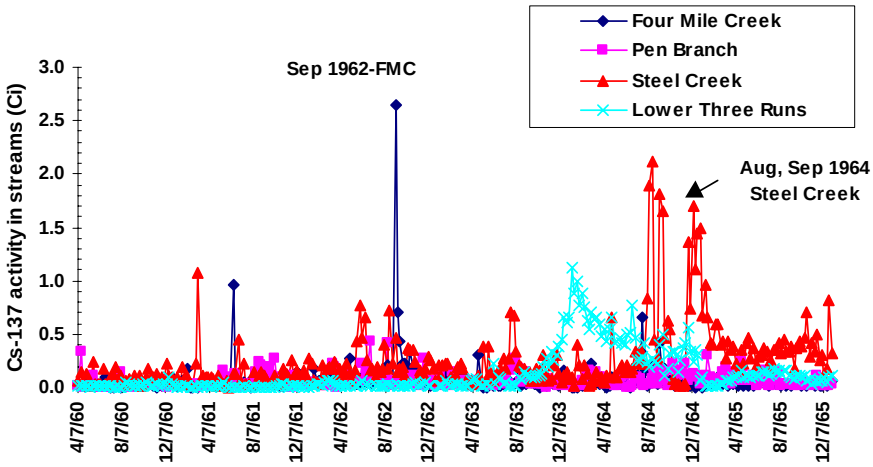

Figure 5-25. Comparison of ^{137}Cs activity in Site streams from 1960–1965. Several peak releases, marked with the date, correspond to unplanned but documented releases.

Studies of ^{137}Cs in the SRS streams and the swamp area have provided information to estimate uncertainties for our revised ^{137}Cs source terms. For example, the Site measured ^{137}Cs in Four Mile Creek (FMC) at Road 4 (just below where H-Area effluents entered), Road C (just below where F-Area effluents entered), and Road A. We used these measurements to calculate a fraction of Cs remaining at each location downstream from the facility discharge point to estimate a range of ^{137}Cs activity that may have reached the Savannah River (Du Pont 1967, 1968a). Roughly 70% (with a range of about 25% to 90%) of the ^{137}Cs measured at either Road 4 or Road C reached Road A (roughly 5 mi downstream in FMC). In this case, Four Mile Creek meanders at least another 7 mi., some through the SRS swamp before reaching the Savannah River. Some of that distance is similar to the conditions from the point of releases to the Road A location on Four Mile Creek and we can assume that under similar conditions that 70% of the activity measured at Road A might reach the river (or about 50% of the original activity measured at Road C).

Site studies support this general range of cesium retention in Site streams. A 1974 study and discussion of ^{137}Cs in effluent streams (Du Pont 1973) concludes that an average of 75% of annual Site releases of ^{137}Cs in liquid effluents are measured in transport in Site streams at Road A and 19% downstream in the river at Highway 301. These percentages were fairly consistent with a similar study in 1963 and 1964 where 80% of the ^{137}Cs released was measured in stream water and 30% in river water at Highway 301 (Du Pont 1964a, 1964b). We have used these estimates of retention in the Site stream beds in our estimates of cesium releases with associated uncertainty distributions.

The Site made a comprehensive radiological survey of the Lower Three Runs Creek corridor from May to August 1971 (Du Pont 1971). Researchers surveyed 11 stations beginning immediately below Par Pond dam and extending to just above the mouth of the main channel of the creek. The survey determined the flow of the Lower Three Runs Creek near its mouth, the distribution of radioactivity levels in sediments, soils, and terrestrial vegetation at various distances from the stream, and the concentrations of specific radionuclides in a few resident aquatic organisms in Lower Three Runs Creek. Measurable concentrations of ^{137}Cs were detected in streambed sediments and core samples. Usually the upper segments had higher concentrations of cesium. Maximum concentration of ^{137}Cs in sediment cores were less than maximum concentrations in soil cores collected at various distances from the stream in the flood plain bordering Lower Three Runs. This was attributed to the flooding conditions that occurred before R area reactor shutdown in 1964. Background levels of ^{137}Cs in soil cores were seen only in samples taken several hundred feet from the stream.

Figure 5-26 displays the results of our source term reconstruction for ^{137}Cs with associated uncertainty estimates. Annual median release estimates were approximately 0.1 Ci or less before 1955 and after 1976. Over 10 Ci of ^{137}Cs were released annually from 1964–1967, with the highest median release estimate in 1964 of about 20 Ci with the 5th and 95th percentiles of the distribution of 50 and 120 Ci. The median estimate of the total ^{137}Cs for all years is about 250 Ci with the 5th and 95th percentiles of the distribution of 100 and 600 Ci ^{137}Cs. There is overall general agreement between our reconstructed ^{137}Cs release estimates to the Site streams based on the original measurement data and supported by weekly and monthly reports from the 1960s and 1970s and the annual total reported by SRS. The small peak in the late 1980s corresponds to the restart of the L Reactor.

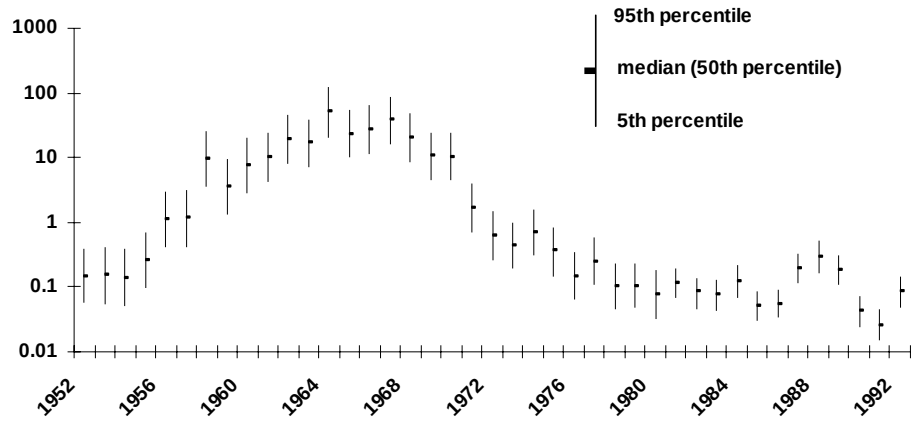

Figure 5-26. Estimates of ^{137}Cs releases to surface water from the SRS with uncertainty estimates, shown on a logrithmic scale. Each year is represented by a vertical line that represents the 95th (top) and 5th (bottom) percentiles of the distribution of releases with the median or 50th percentile shown as the filled shape in the center.

The time-dependent release pattern of ^{137}Cs to surface water reflects the operations and events that occurred onsite as shown in Figure 5-27. Releases of ^{137}Cs to surface water peaked in 1964 when all reactors were operating. Quite a large number of fuel failures and other unplanned events like spills occurred in the late 1950s through 1964, when the R Reactor was permanently shut down (Du Pont 1958, Du Pont 1962, Du Pont 1966). In addition, the Site processed short-cooled fuel on several occasions during this time, which led to higher levels of radioactivity releases to the streams when the disassembly basins were purged (Carlton et al. 1992a).

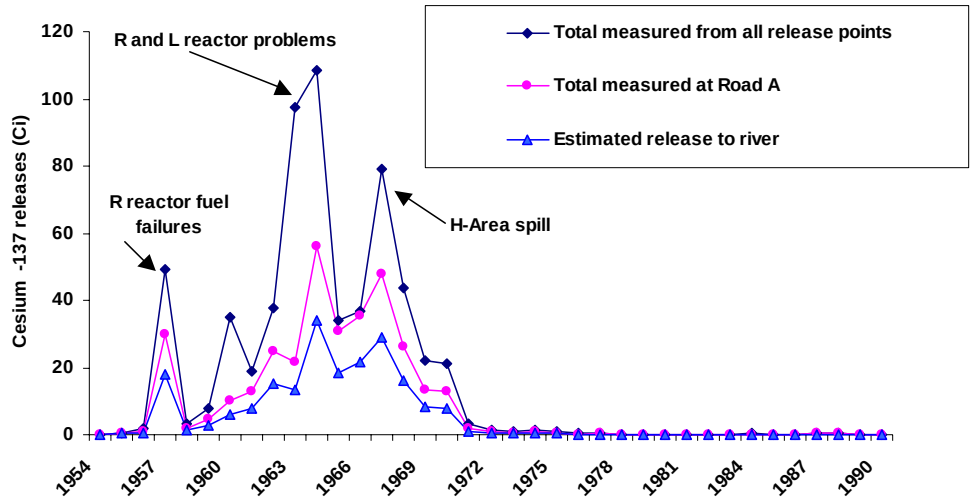

Figure 5-27. Comparison of ^{137}Cs activity from 1954 through 1990 released from the facilities, measured in onsite streams and estimated to have been released to the Savannah River.

Health Physics Regional Monitoring reports for 1957 and 1958 describe a number of fuel failures at the L, P and R Reactors that contributed to increased levels of nonvolatile beta activity in the Site streams. Further isotopic analyses were done in some instances and provided a rough estimate of levels of radiocesium and radiostrontium in the Site streams before routine monitoring for these specific radionuclides were done beginning in mid 1958. For example, the increased discharge of nonvolatile beta in disassembly basin water from L-Area and P-Area peaked in August and September 1957 due to failed fuel elements in those reactors. In addition, Site personnel had discontinued cooling water flow from L-Area in preparation for a process improvement shutdown, and concentrations of nonvolatile beta activity remained fairly high until mid-November 1957 when cooling water flow was resumed (Mealing et al. 1958).

Strontium Release Estimates

Natural mechanism for radiostrontium production is fission of naturally occurring uranium and thorium. Such fission occurs either by spontaneous fission decay or by neutrons in nature inducing fission (Carlton et al. 1992b). Two human-made sources have contributed to the radiostrontium in the global environment. These events are:

- Above-ground nuclear weapons test introduced about 22,000,000 Ci ^{90}Sr to the atmosphere, mostly through the mid-1960s
- Nuclear reactor operation (Satellite Cosmos 954 accident in 1978 released about 84 Ci ^{90}Sr at re-entry into earth's atmosphere and Chernobyl nuclear reactor accident in 1986 released 34,000 Ci ^{90}Sr.

Most radiostrontium at the SRS originated as fission products in the nuclear fuel and targets during the operation of the five production reactors. There are over 12 isotopes of radiostrontium produced but five of these have half-lives less than a second and, except for ^{89}Sr and ^{90}Sr, the others have half-lives of less than 10 hours. During normal reactor operations, radiostrontium is contained within the cladding of fuel and target elements during irradiation and cooling. Cooling is the interval between the end of irradiation and the beginning of chemical separations. The irradiated materials were stored underwater in large basins of water called disassembly and cooling basins. Beginning in the 1970s, the Site established a standard cooling for most irradiated materials of at least 200 days. This length of time limited the emissions of radioiodine significantly. About 200 days reduced the inventory of ^{89}Sr by a factor of about 16 but had little effect on the ^{90}Sr inventory.

Figures 5-28 and 5-29 show the general release pattern for radiostrontium to the Site streams, a pattern that is similar to that seen for tritium and ^{137}Cs releases. The pie chart indicates that over 60% of the total ^{89,90}Sr activity measured in transport in Site streams was found in Steel Creek, which received effluents from the L and P Reactors. Pen Branch carried roughly 15% of the activity, while Four Mile Creek and Lower Three Runs carried approximately 10% of the activity. Upper Three Runs transported very small amounts of ^{89,90}Sr, if any, because it received discharges only from the northside of the F-Area. The reactors were clearly the largest source of ^{90}Sr to the streams (Figure-29).

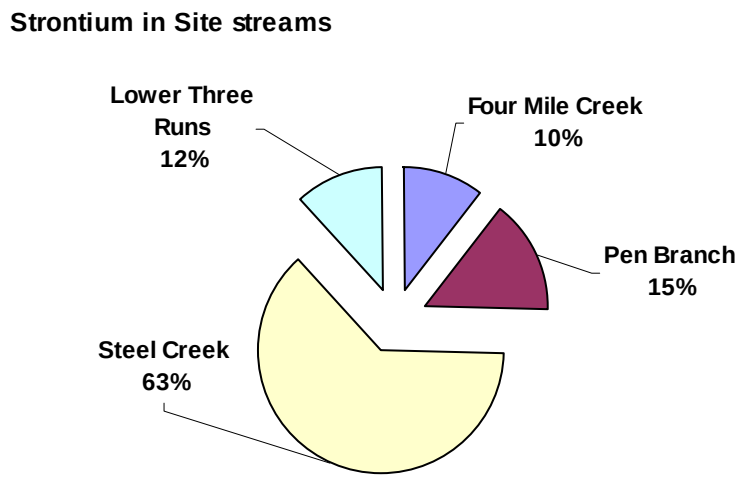

Figure 5-28. The relative amount of total strontium activity in the major Site streams measured weekly from 1960-1967 in onsite streams at Road A, the last onsite location before the streams emptied into the Savannah River. Over half of the ^{89,90}Sr activity released during this period was present in Steel Creek.

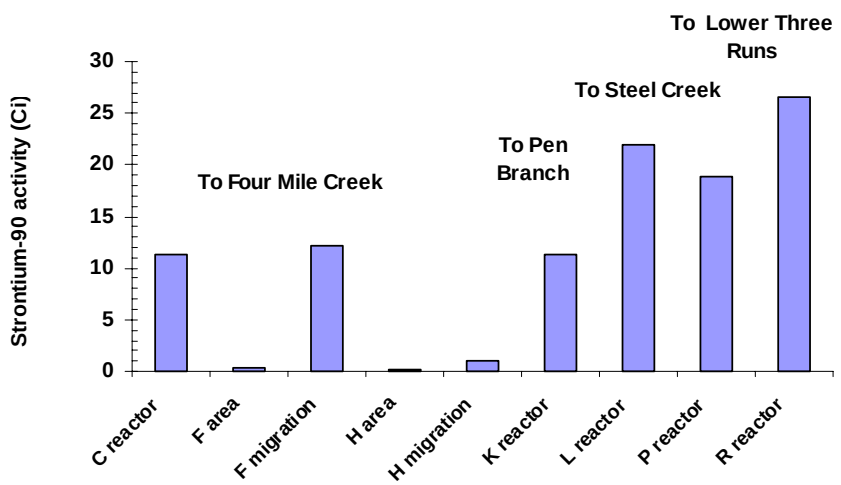

Figure 5-29. Releases of ^{90}Sr from various sources at the SRS over all years of operations. The reactors were the primary source of releases of ^{90}Sr (and other fission and activation products) at the SRS. Releases to Steel Creek were the highest because the reactors in the L-Area, P-Area, and R-Area discharged to that creek at different times over the years. However, liquid effluent releases from the P Reactor also went to the Lower Three Runs system.

Radiostrontium was measured in the reactor basin purge water beginning in the mid 1950s and-in the Site streams beginning in 1960. Before that time, only nonvolatile beta activity was measured in the preoperational survey and from the beginning of plant operations. To estimate ^{90}Sr releases for times when ^{90}Sr-specific measurements were not made, we calculated a ratio of ^{90}Sr to nonvolatile beta activity when both measurements were made at the same time and same locations. We used this ratio, along with the nonvolatile beta activity measurements to estimate levels of ^{90}Sr activity in the streams at Road A when ^{90}Sr-specific measurements were not made. This procedure is the same as we followed in estimating ^{137}Cs releases when specific measurements were not available (see Cesium release section in this chapter).

Our revised release estimates for ^{90}Sr were based on reported concentrations and flow measurements made in the streams, combined with uncertainties that have been described previously. Figure 5-30 shows the reported six-month totals of ^{90}Sr release to the Site streams. Figure 5-31 displays the results of our source term reconstruction for ^{90}Sr with associated uncertainty estimates. Annual median release estimates were approximately 1 Ci or less before 1957 and after 1976. We estimate that over 15 Ci of ^{90}Sr was released to the river in 1960 and over 5 Ci in 1962–1967 annually from 1964–1968. The median estimate of the total ^{90}Sr for all years is about 100 Ci with the 5th and 95th percentiles of the distribution of 45 and 250 Ci ^{90}Sr.

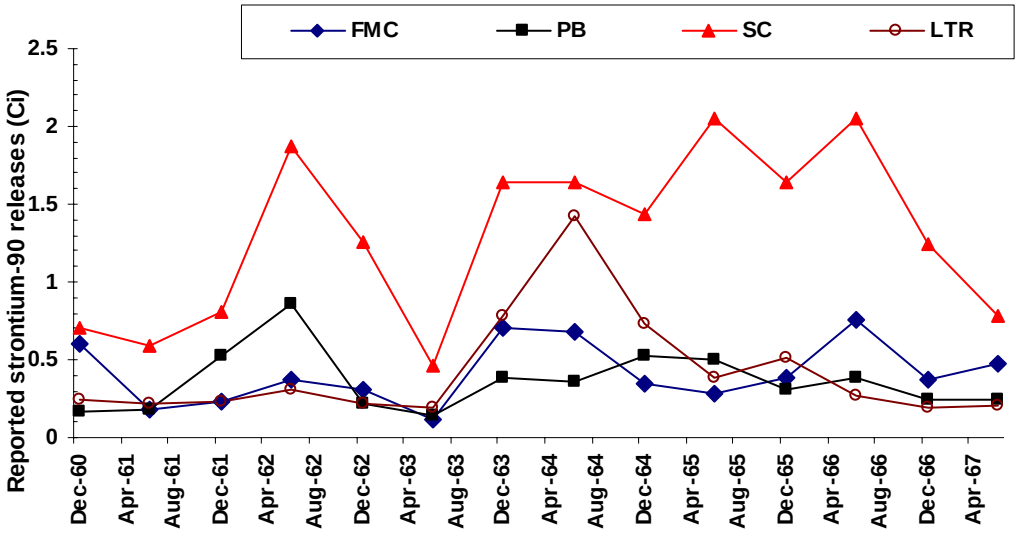

Figure 5-30. Semi-annual activity levels of ^{90}Sr in the streams based on the reported concentration measurements and flow rate in the Site streams at Road A.

The rate of migration from the seepage basins in the F-Area and H-Area have gradually declined since about 1970 when ^{90}Sr releases were estimated and measured, but the releases to Site streams from groundwater outcrops has been the biggest source of ^{90}Sr during that time.

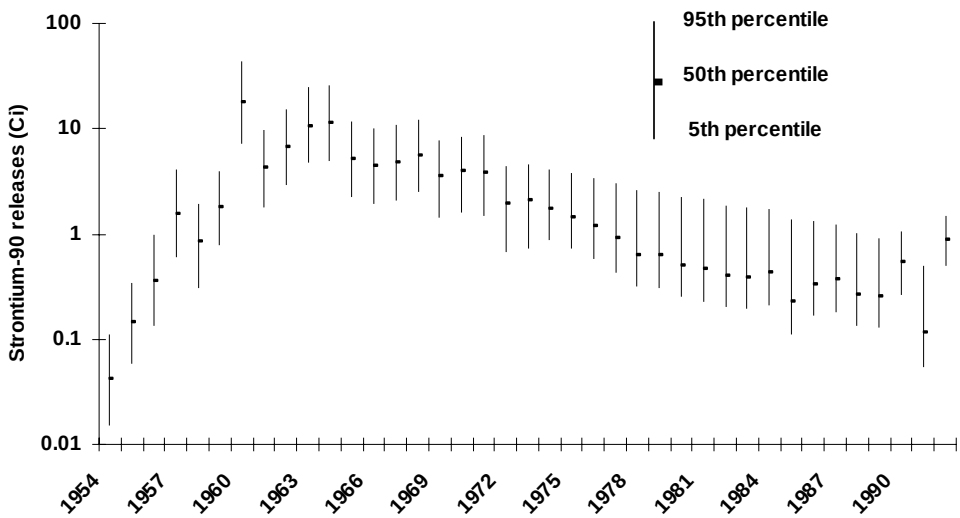

Figure 5-31. Estimates of ^{90}Sr releases to surface water from the SRS with uncertainty estimates, shown on a logrithmic scale. Each year is represented by a vertical line that represents the 95th (top) and 5th (bottom) percentiles of the distribution of releases with the median or 50th percentile shown as the filled shape in the center.

Releases of Iodine-131

As with the other fission and activation products important in SRS releases, the major sources of ^{131}I at the SRS were the chemical separations areas and the reactor areas. Figure 5-32 presents the releases of ^{131}I in liquid effluents to the streams and seepage basins from the SRS.

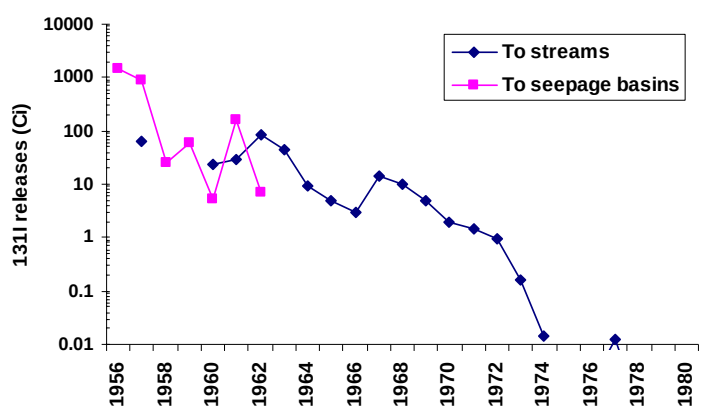

Figure 5-32. Reported releases of ^{131}I to Site streams and to seepage basins from the SRS. The large increase in releases to the seepage basins in 1961 resulted from the dissolution of short-cooled fuel elements in the separations area.

In the early years, a chloroform extraction technique was used to separate ^{131}I from stream and river water samples. This procedure had a sensitivity of about 6 pCi L^{-1}. In January 1963, a new technique was introduced for the analyses of ^{131}I in water that used an ion-exchange cation and anion resins to concentrate the radionuclides. The sensitivity of this method for ^{131}I in 40 L of sample was 1 pCi L^{-1} (Boni 1963).

Releases of Activation Products

The SRS released activation products in liquid effluents from facilities and operations that were similar to those responsible for releases of fission products to surface water. Chapter 4.3 in this report provides some background on the releases of activation products to air and surface waters from the SRS. Three activation products, ^{60}Co, ^{35}S and ^{32}P, may be of some importance for offsite residents if certain exposure pathways are considered (see Chapter 3 of this report). Our evaluation of reported releases of these radionuclides indicates that the historic record does include memos, and reports documenting interest in and monitoring of these materials. We have compiled measurement data for these radionuclides that can be used for later exposure assessment if such an assessment if warranted. The following sections provide summaries of information of these radionuclides.

Cobalt is chemically similar to iron and like iron, can exist in Co(II) and Co(III) oxidation states. However, Co(III) is only dominant at conditions beyond the oxidation limits of water at 25° C and 1 atmosphere, and thus in water Co(II) is dominant form. In the acidic conditions typical of SRS waters, cobalt is relatively mobile. Sorption to solids in the water can slow down cobalt migration. Factors such as pH, mineral content and grain size can strongly influence sorption. This is reflected in the range of K_d values (ratio of amount of constituent sorbed to solid phase over the concentration in water at equilibrium) of 0.2 to 3800 ml per gram (see Table 5-8).

The largest quantities of ^{60}Co were released from the SRS to the seepage basins in the F-Area and H-Area. Seepage from the basins released some activation products to the Site streams, particularly to Four Mile Creek. Figure 5-33 shows that more ^{60}Co was released to the Site streams than to the seepage basins in the early years. In the mid 1970s this trend generally changed when releases to the streams were more tightly controlled. This pattern is similar for the release of other activation products, too.

A second activation product released to surface water is ^{35}S. Prior to 1961, ^{35}S was not included in the reactor area release summaries. In the last quarter of 1961, specific analysis for ^{35}S was done and was detected in disassembly basin water from the reactor areas. The reason for doing ^{35}S analysis was because there was an imbalance between the nonvolatile beta activity in reactor effluents and the total estimated disassembly basins releases based on stream data during October 1961. The stream activity exceeded releases from the seepage basins.

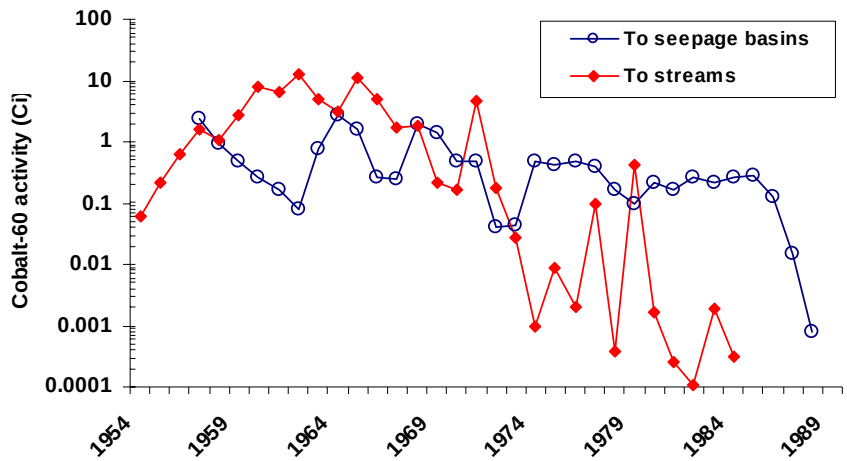

Figure 5-33. Reported releases of ^{60}Co to Site streams and seepage basins from the SRS. The releases of ^{60}Co to the streams were higher than releases to the seepage basins in the early years. The pattern is reversed in later years.

By January 1962, Site personnel were investigating the source of the ^{35}S. One thought was that the amount of ^{35}S found in the C-Area process water (0.015 uCi per ml) could have originated from chlorine and/or sulfur impurities introduced into the system in concentrations less than 1 ppm. The ^{35}Cl (n,p) ^{35}S reaction appeared to be the most likely source of ^{35}S, requiring levels of chloride impurities as low as 100 ppb (Du Pont 1961). In April 1962, it was demonstrated that the ^{35}S releases coincided with reactor outages and originated from fuel decontamination test tank that contained oxide layers from fuel assemblies. This was also a source for ^{32}P discharges.

The chemistry section at the SRS continued to standardize and optimize the chemistry methods for ^{35}S. Because the concentrations were generally very close to the lower detection limit (Du Pont 1961), the Site analysts continued to improve the method. Figure 5-34 compares the reported total releases of ^{60}Co, ^{32}P, and ^{65}Zn to streams from facilities at the SRS. This figure demonstrates that, just as with tritium and fisson product releases, the reactors were the main source of activation products released to Site streams. Measured releases of radionuclides in the various streams, combined with the known reactor operational times, indicate the while some reactors worked relatively well with minor complications over the years, others, like the R Reactor, were quite error and problem-prone. For example, the R Reactor operated only through 1964 but contributed the largest releases of some radionuclides, like ^{65}Zn to the Site streams.

In summary, the releases of key beta-gamma emitters (tritium, ^{137}Cs, ^{90}Sr) were greatest from the reactor areas. Although most releases from these areas to streams came from the disassembly basins and not directly from operations of the reactors, the overall pattern of releases over time are related to reactor operations. Figure 5-35 clearly demonstrates that the reactor power levels were highest in the early to mid-1960s because all five reactors were in full operation. Fuel failures and other problems in the R and the L Reactor areas accounted for some of the higher releases of radionuclides to water during the early 1960s. Finally, the quantities of activation

products released to air and water were quite small compared to the quantities of fission products, like ^{137}Cs, and ^{90}Sr that were released.

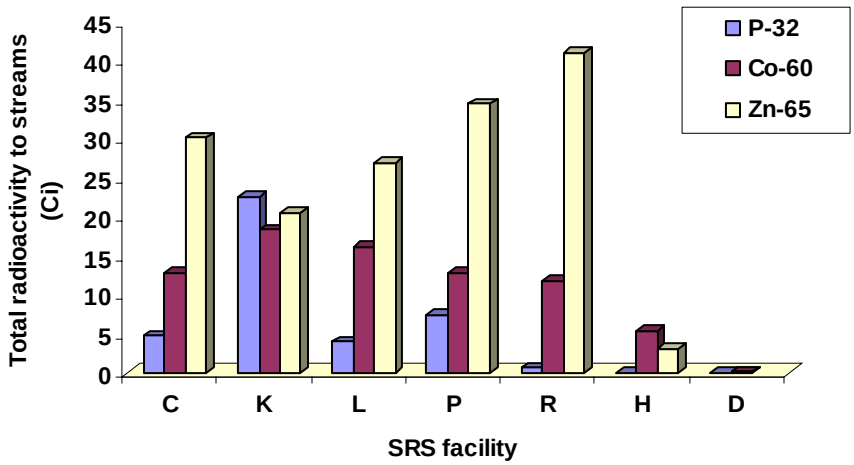

Figure 5-34. Comparison of total releases for all years from the SRS of ^{32}P, ^{60}Co, and ^{65}Zn. C, K, L, P, and R refer to the five operating reactors at the SRS. In this figure, H refers to the H-Area and D refers to the Heavy Water or D-Area.

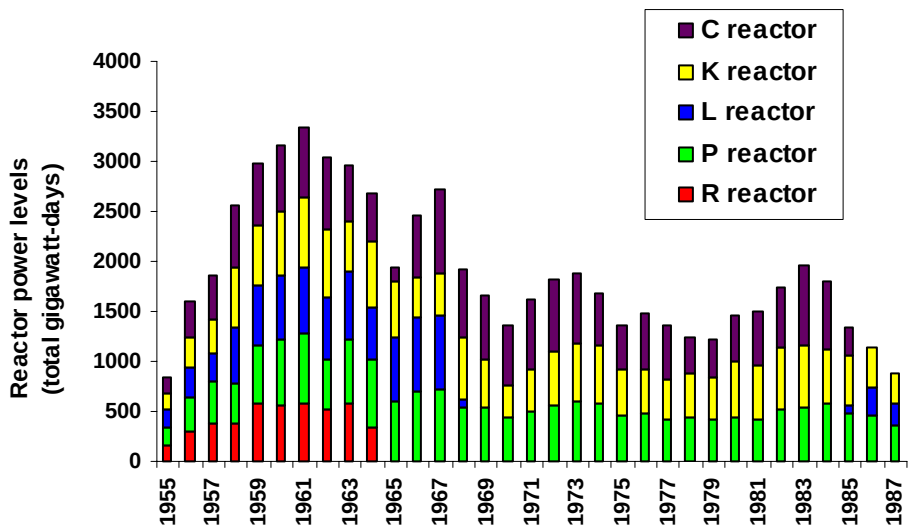

Figure 5-35. Reactor power levels for the operation reactors from 1952 through 1988. The operation periods for the reactors were: C Reactor 1954-1987, K Reactor 1954-1988, L Reactor 1954-1968 and 1985-1988, P Reactor 1954-1988 and the R Reactor 1954-1964. A restart of the K Reactor began in 1991. From 1954 through 1964, all reactors were operating.

Releases of Alpha Emitters: Uranium

Our radionuclide screening did not indicate that uranium would be a major contributor to offsite radiation doses; however, there were fairly large quantities of uranium released to Site streams, especially from the reactor materials area or M-Area. Furthermore, future exposure pathway analysis may indicate that although uranium may not be important radiologically, it may be important for its potential chemical toxicity (see Chapter 18 in this report). For these reasons, we summarize some of the release information that we have compiled for uranium.

In the 1950s, the SRS used an extraction method to separate uranium and plutonium from environmental samples like vegetation, soil, and water. This newer TBP extraction was based on a method commonly used at that time using nitric acid and tri-n-butyl phosphate in n-tetradecane diluent, and replaced the ether extraction method, which was more hazardous in the laboratory. From spiked sample studies, uranium and plutonium were nearly equally recovered. The average recoveries were about 76% for vegetation, 76% for soil and 82 ± 15% for water (Geiger circa1958). The procedure was used as part of the health physics regional monitoring program at the SRS. In the 1960s, uranium and plutonium were extracted by liquid ion exchange using tri-isooctulamine (TIOA) and then alpha counted (Johnson 1968a). One liter water samples were prepared for both extraction procedures with a final 10-15 ml volume transferred to a counting planchet for drying and alpha scintillation counting. The TIOA extraction method continued to be used for determining the concentration of uranium and plutonium in water to recent times (Du Pont 1989).

Uranium was produced and discharged from several areas at the SRS. The largest releases of uranium onsite occurred from the M-Area to Tim's Branch and to the seepage basins after they were put into service in 1973. Liquid overflow from several points in this process was discharged to the sewer system and directly to the surface streams, especially in the early years. The direct release of drain water from the uranium target production facility in M-Area was stopped in 1973 when the process sewer discharges were diverted to the newly opened M-Area seepage basin (Evans et al. 1992). In 1979, the primary uranium release streams were diverted to the settling basins and, by May 1982, all releases of untreated process wastewater were diverted to the settling basin.

Within the M-Area, the 300-Area was one of the first operations to begin at the Site. It was also one of the first sources of Site contamination to be of concern (Albenesius 1954). From startup in October 1952 through February 1953, an estimated 100 lb of uranium was discharged in the 300-Area effluent system to Tim's Branch. After that time, releases of uranium from the M-Area were routinely monitored and reported (Alexander and Horton 1956). Figure 5-36 shows that during the first 8 years, the 6-month uranium release totals varied from about 16 lb to over 4000 lb in late 1963, when a new etching process was introduced in September 1963. The 1960s began an upward trend in uranium releases to Tim's Branch that peaked in 1969 when over 25,000 lb of uranium was released. During this time, there was concern about the amount of uranium being released (Johnson 1968b).

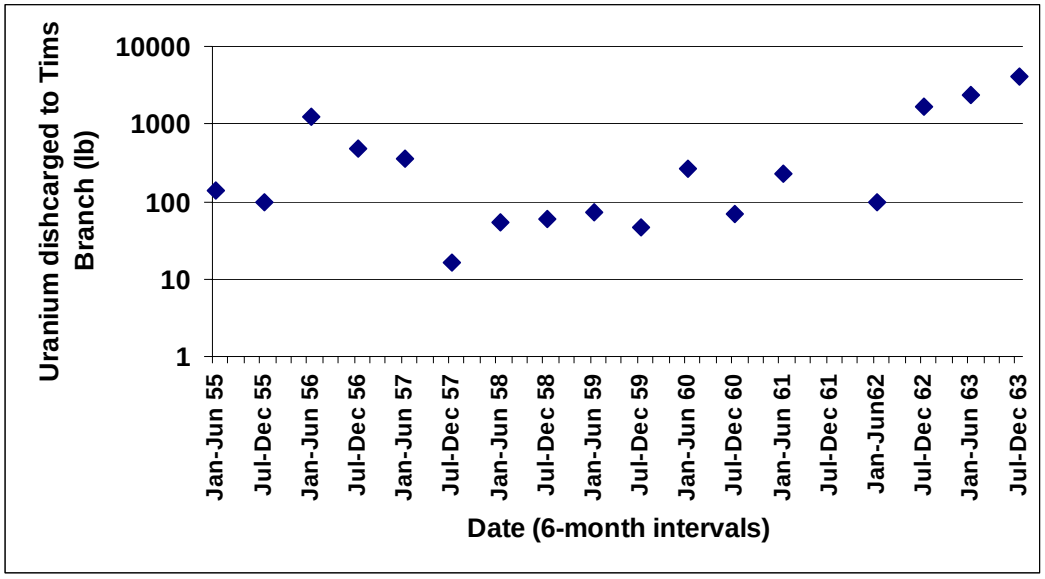

Figure 5-36. Measured releases of uranium to Tim's Branch from the M-Area from January 1955 through December 1963, reported in the health physics semi-annual progress reports.

Figure 5-37 illustrates that the largest releases of uranium to Tim's Branch occurred from 1964 to 1970. The high releases in the mid to late 1960s were due to the loss of uranium etching solution and the overflow of normal autoclave waste from the holding tank.

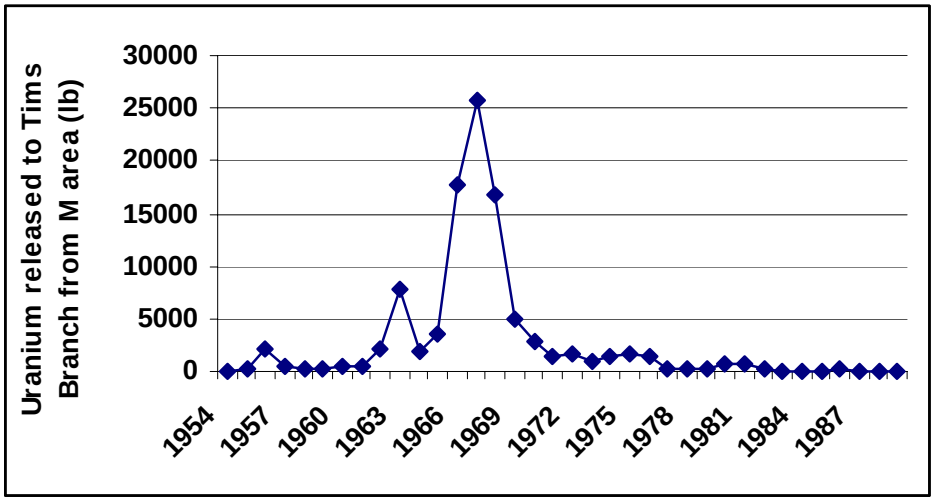

Figure 5-37. Reported annual uranium releases to Tim's Branch from M-Area. One curie uranium is equivalent to 6190 lb. uranium; therefore, 25000 lb uranium = ~4 Ci uranium. The total quantity of uranium release over this period was approximately 153,000 lb. (~25 Ci).

There were small releases of uranium from the separations area. Cooling water for portions of the F-Area and H-Area separations process line was pumped from deep wells and discharged to Four Mile Creek (FMC) after use. The water contained measurable amounts of radioactivity because of cooling coil leaks. Uranium was a predominant chemical constituent of this mixture (Evans et al. 1992). Cooling water discharged to Four Mile Creek was not analyzed specifically for uranium, and the activity was reported as "unidentified alpha." There were high releases of unidentified alpha reported in 1955 (0.045 C) and 1956 (0.037 Ci) to Four Mile Creek that were associated with evaporator coil leaks. Alpha spectrometry measurements on a few samples in 1955 attributed most activity to ^{238}U. If we assume all activity was due to ^{238}U then these activity levels correspond to about 300 lb. in 1955 and 240 lb in 1956. These conversions are based on a specific activity for ^{238}U of 1.24 Bq g^{-1}, or 6580 Ci lb^{-1}. (If we assume the uranium was natural uranium, then the activity levels correspond to about 140 lb. in 1955 and 120 lb. in 1956. The specific activity for "natural" uranium [99.27% ^{238}U, 72% ^{235}U and 0.0055% ^{234}U] is 692 nanocuries g^{-1}, or 3190 lb Ci^{-1}). High releases in 1984 (0.025 Ci unidentified alpha) were probably depleted uranium in runoff from the uranium recovery facilities (A-Line) to the storm sewer system. This activity corresponds to about 80 lb. of natural uranium.

For the reactor areas, the Site began measuring gross alpha in liquid effluents from the reactor areas in 1972, and there were no specific analyses for uranium. Figure 5-38 shows the pattern of alpha activity measured in wastewater from the reactors to the Site steams and to the seepage basins after 1972. These values are not corrected for the natural uranium content of the incoming river water, which is the source of the purge water for the reactor area

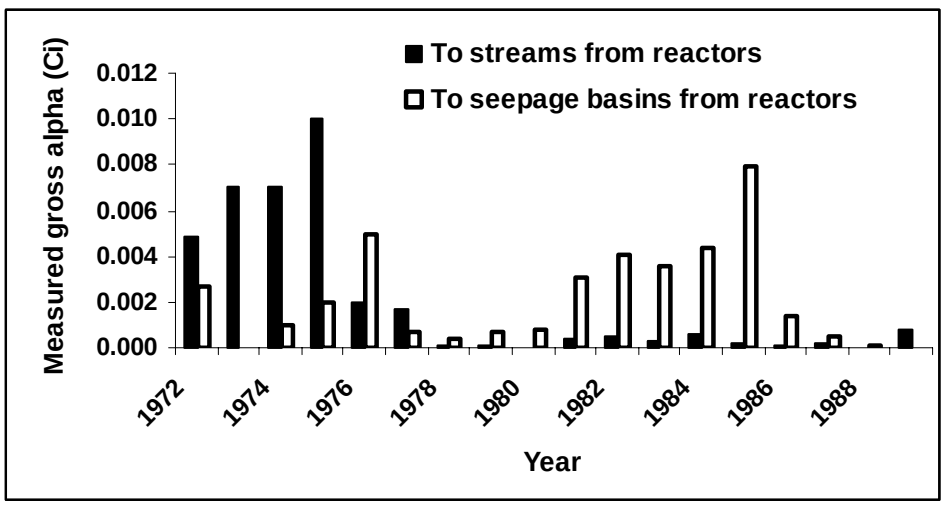

Figure 5-38. Measured releases of gross alpha from reactor areas to Site streams and the seepage basins from 1972–1989. These values are not corrected for the natural uranium content of the incoming river water, which was the source of the purge water. No measurements were made prior to 1972 and there were no specific analyses for uranium. The figure demonstrates a trend seen for other radionuclides discharged in liquid effluents, that is, that in later years, more effluents were discharged to the seepage basins rather than directly to the Site streams.

Looking at the concentration of uranium in stream water should provide insight into the quantity of uranium that may have reached the Savannah River. If we assume that all alpha activity is due to natural uranium, then this leads to release estimates of about 0.005 to 0.01 Ci (~15-30 lb of uranium) per year from 1972-1975. Much lower levels of alpha activity in wastewater from the reactor areas were reported in other years when measurements were made. These levels are for the wastewater entering Site streams. The uranium activity that reached offsite locations in the Savannah River would be much less. In the early years, only gross alpha was measured in Tim's Branch but this provides a good indication of the movement of uranium and other alpha emitters in Tim's Branch. Figure 5-39 shows that alpha activity increased dramatically in Tim's Branch after receiving the 700-Area effluent. Then a significant drop off in activity occurred as the water moved downstream. Near the confluence of Tim's Branch with Upper Three Runs and then at a point in Upper Three Runs 2 mi before Upper Three Runs discharges into the Savannah River, the alpha activity was near the levels measured in Tim's Branch upstream of the M-Area discharge points. While Savannah River water was not monitored specifically for uranium, alpha activity was measured routinely since plant startup. There were no differences in alpha activity levels upstream and downstream of the plant, even in the late 1960s, when the largest releases of uranium occurred from the M-Area.

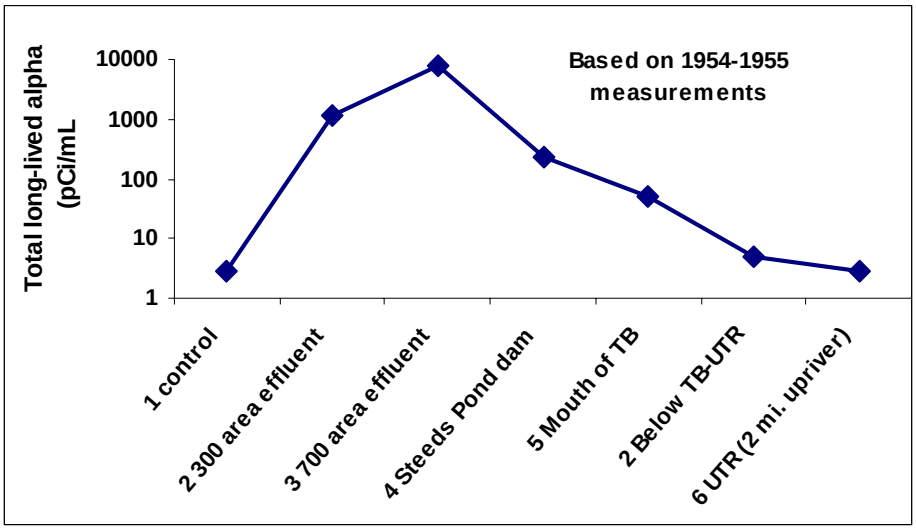

Figure 5-39. Alpha activity measured at various locations in Tim's Branch in 1954 and 1955. Alpha activity increased dramatically in Tim's Branch after receiving the 700 area effluent, then showed a significant drop off as the water moved downstream.

Studies have shown that uranium has deposited in the sediments of the streambed, thus accounting for the dramatic decrease in water with distance downstream. As early as June 1954, a study was underway to determine the distribution of uranium, released from the 300 and 700 areas, which had been removed from the water of Tim's Branch (Horton 1955a). The study included the determination of the distribution of uranium in the streambed, the uranium absorption by vegetation growing in and surrounding the stream, and the uranium content of the surrounding groundwater. There were 11 sampling stations at various locations. Horton 1955a contains complete soil sample data

Analysis of the collected data revealed no significant difference between the uranium content of the streambed at any of the sampling depths from 1 in. up to and including 6 to 9 in. There is a significant decrease in the uranium content of the samples collected below a depth of 9 in. in the streambed or at the water table at a distance of 6 ft from the edge of the stream. The maximum concentration of uranium in the streambed was at a location about 5 yd below the 300-Area discharge. The uranium content of the groundwater varied with sample location. At one point, uranium was detected at a distance of 45 ft from the edge of the stream. The uranium content of vegetation growing in the stream water was significantly higher than the uranium content of the vegetation growing near the stream. No uranium in vegetation was detected as the sampling locations became more removed from the effluent discharges.

SUMMARY

The Site had a fairly broad effluent monitoring program for releases of key radionuclides from the main facilities onsite to the streams and seepage basins. Measurements of alpha, beta, and some tritium were made from the beginning of operations and a preoperational survey helped to establish background levels of alpha and beta activity in some of the Site streams and in the Savannah River. We located many early records of measured concentrations and effluent flow to the streams for use in reconstructing past releases. The documentation, in the form of weekly and monthly reports, was quite extensive and there has been a great quantity of information to review.

To develop the surface water source terms, the process of screening using the NCRP methodology has focused our efforts and resources on those radionuclides that potentially are the largest contributors to radiation dose to those living offsite during the operational years at the SRS. Tritium, ^{137}Cs and ^{90}Sr are the main radionuclides of concern from past releases to surface streams that eventually reached the Savannah River, and we estimated detailed source terms for those radionuclides. Other radionuclides that also may be important depending upon the exposure pathways include ^{131}I, the activation products, ^{60}Co, ^{32}P, and ^{65}Zn, and uranium releases from the M-Area.

Surface water releases of radionuclides were highest in the early to middle 1960s and decreased into the 1980s. Our median estimate of release of tritium for all years (1952 through 1992) is 1.8 million Ci, with the 5th and 95th percentiles of the distribution of 1.3 million and 2.5 million, respectively. The median estimate of the total ^{137}Cs for all years is about 250 Ci with the 5th and 95th percentiles of the distribution of 100 and 600 Ci ^{137}Cs. There is overall general agreement between our reconstructed ^{137}Cs release estimates to the Site streams based on the original measurement data and supported by weekly and monthly reports from the 1960s and 1970s and the annual total reported by SRS. The median estimate of the total ^{90}Sr for all years is about 100 Ci with the 5th and 95th percentiles of the distribution of 45 and 250 Ci ^{90}Sr.

As would be expected, there is more uncertainty associated with release estimates in the 1950s and 1960s than in later years because of improvements in monitoring capabilities and techniques and in methods to prevent the release of materials from the reactors and processing facilities. The reactors were the source of the majority of radionuclide releases to surface water, primarily because most surface water releases came from the disassembly basins in the reactors areas, while releases of liquid effluents from the separations areas were discharged into storage tanks and seepage basins from the start of operations and not directly to the streams.

Nature, in the form of precipitation and the SRS swamp, played an important role in surface water releases. The year of highest releases to streams for tritium and ^{137}Cs (1964) was also the year with exceptionally high rainfall that increased runoff and led to heavy flooding of the SRS swamp. While the swamp was flooded cyclically about 20% of the time, the swamp also affected the mobility of some radionuclides flowing in stream water to the Savannah River by physical and chemical processes.

Finally, it is difficult to understand the full meaning of these release estimates before completing subsequent studies, where radiation doses and risks associated with these releases can be evaluated.

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