CHAPTER 4.1

RELEASES OF TRITIUM TO THE ATMOSPHERE

ABSTRACT

Tritium (^{3}H or T) is one of the principle nuclear materials produced at the Savannah River Site (SRS). The key processes that have lead to atmospheric tritium releases are reactor operations (100 Areas), recovery of transuranic elements in the separations facilities (200 Areas), recovery of tritium in the tritium processing facilities (200 Areas), laboratory research area, and heavy water rework facility. The majority of the tritium releases came from the reactor facilities and the tritium processing facilities. This chapter presents brief histories and descriptions of operations at these facilities and the SRS source term.

The releases of tritium from the SRS are generally very well documented in published reports. Records of releases have been kept since almost the beginning of operations. The main sources of information were SRS weekly and monthly reports. In the cases where daily or shift data were found in logbooks, we used them to check the monthly published data. In general, there is quite good agreement between the different release values reported in the different sources. Because in most cases the summary values were within 10% of the published values, it is likely that most "unchecked" monthly values are also valid.

The research on the quality of the atmospheric tritium release data reported by SRS has given us no indication that there were serious or consistent errors made in carrying out the tritium release monitoring. However, it is important that the best estimates presented in this report are used in conjunction with the uncertainty associated with each of the values.

INTRODUCTION

Tritium is one of the principle nuclear materials produced at the SRS. It is the heaviest and only radioactive isotope of hydrogen with a physical half-life of 12.5 years. Tritium is used to multiply the firepower of plutonium in nuclear weapons. Tritium processing operations have been performed at the SRS facility since October 1955, beginning in Building 232-F. In July 1957, the process was moved to Building 232-H and operations doubled by 1958. This section discusses the atmospheric releases of tritium to the environment, called the atmospheric source term. Chapter 5 presents the liquid releases.

Tritium is produced naturally by cosmic ray interactions and as a fission product in nuclear reactors. The nuclear reactions that produce tritium are

- Fission of lithium by neutron irradiation of targets
- Reaction of neutrons with the heavy water moderator used in reactors
- Ternary fission of transuranic elements in the reactor fuels and targets.

At SRS, tritium was originally produced as a by-product of producing plutonium in the reactors. Lithium-aluminum control rods were used in R-Reactor and other reactors from the first day of operation to produce tritium and control reactor power. When additional tritium was needed beginning in 1955, enriched uranium was used in the reactors, which allowed for increased loading of lithium-aluminum targets for tritium production.

Gaseous T_2 at room temperature tends to form elemental tritium (HT) by reaction with gaseous hydrogen. Tritium in this form does not constitute a significant radiation health hazard because the beta particle given off during its decay is of low energy and has low penetrating power. Tritium closely follows the reactions of ordinary hydrogen when it exists as tritiated water or vapor (T_2O; tritium oxide [HTO]) and is taken into the body quite easily, resulting in a higher factor of risk. Tritium is released from SRS in both elemental and oxide forms.

The key processes that have lead to tritium releases at the SRS are

- Reactor operations (100 Areas)
- Recovery of transuranic elements in the separations facilities (200 Areas)
- Recovery of tritium in the tritium processing facilities (200 Areas)
- Laboratory research area
- Heavy water rework facility.

The majority of the tritium releases came from the reactor facilities and the tritium processing facilities. This chapter presents brief histories and descriptions of operations at these facilities and the reconstructed SRS atmospheric tritium source term. Appendix E presents a detailed description of the monitoring equipment used to monitor atmospheric tritium releases and an uncertainty analysis to be applied to reported releases from the different monitors.

REACTOR FACILITIES

History

Heavy water moderated and cooled reactors using natural uranium fuel were used at the Savannah River Plant (SRP). The modified reactors were called pi-Pile and were from Argonne National Laboratory (Stetson et al. 1963). The first reactor came online in December 1953, and by March 28, 1955, all five reactors were in operation. Reactor locations were designated as the 100 Areas (e.g., 100-R was the name of the R-Reactor area).

The R-Reactor went into operation on December 28, 1953. Full power was added stepwise until the spring of 1955 when all reactors were at their design limit of about 500 MW. During the second period (from July 1955 to November 1956), there was an extension of power ascension to about twice the original design limit. During the third period (from December 1956 to October 1958), there were major increases in the capacity of the primary and secondary cooling systems. During this period, all reactors were shut down for 2–3 months each to replace D_2O pumps and piping. The pumps were replaced from December 1957 to July 1958. Cooling water supply from the Savannah River was augmented by the construction of Par Pond (Stetson et al. 1963).

From October 1958 to January 2, 1962, there were no dramatic increases in reactor power levels (only about 2 to 10%). From January 2, 1962, until the end of 1963, there were extended shut downs to install containment facilities, which were built in phases during that period (Stetson et al. 1963).

The reactors were devoted primarily to the production of plutonium and tritium. On September 5, 1956, L-Reactor changed to 100% tritium production and operated with enriched uranium fuel most of the time. C-Reactor converted to enriched uranium fuel March 3, 1957, and K-Reactor converted on August 5, 1957. After that time, the operating reactors alternated between plutonium and tritium production. Tritium production peaked in 1958, when three

reactors operated on enriched fuel. By January 1959, K-Reactor and C- Reactor began to alternate between enriched and natural uranium fuel (Stetson et al. 1963).

During Fiscal Year (FY) 1959 and FY 1960 (during this period of time, the government FY ran from July 1 to June 30), about 40% of reactor operation was devoted to tritium production and about 60% to plutonium production. From July 1960 to late 1963, about 25% of reactor operations was devoted to tritium production (Stetson et al. 1963). The R-Reactor was placed on standby status in 1964 and was never restarted. The L-Reactor was placed on standby in 1968 and later restarted. The other three reactors operated in the 1970s, producing both tritium and plutonium.

In 1982, it was planned that at startup L-Reactor would devote 3% of its capacity to ^{240}Pu production, which would involve five sets of targets to be discharged during each fuel cycle. These conditions were expected to increase the tritium evaporation from the disassembly basin and seepage basin (Dukes and Benjamin 1982). Table 4.1-1 summaries reactor operations.

Production reactors processes at SRP included reactor operation, an analytical laboratory, and facilities for reworking the degraded heavy water moderator (D_2O). The major reworking facilities consisted of vacuum fractional distillation equipment, minor rework operations, and a facility for cleaning the drums in which the heavy water was stored and transported (Longtin et al. 1973; Jacober et al. 1973).

Table 4.1-1. Summary of Reactor Operations

Reactor	Dates of operation	Comments
C	1955–1987	Placed in cold standby[a] in 1987; now shutdown
K	1954–1988	1991 put in cold standby; now shutdown
L	1954–1968	Now shutdown
	1985–1988	
P	1954–1988	Now shutdown
R	1953–1964	Now shutdown

[a] Cold standby: available to restart after refueling and upgrading.

The major part of the work to restore isotopic purity to the D_2O was done by vacuum fractional distillation in the 400 Area. When not in use, the heavy water was stored in drums. The drum wash facility was used to remove the small amounts of tritiated D_2O that remained when the drums were emptied (Longtin et al. 1973; Jacober et al. 1973).

Releases of tritium from the production reactors were due almost entirely to releases of moderator, either as liquid or vapor from the reactor systems and from the heavy water rework facility. Small amounts could have been released from the reactor blanket gas system and from irradiated lithium-bearing control rods and targets stored underwater in the disassembly basins (Longtin et al. 1973; Jacober et al. 1973).

Reactor Stacks Tritium Monitoring

Early years (pre 1970s)

During the early years, before 1970, the two main types of monitoring systems employed to measure airborne tritium releases from SRS reactors were the silica gel and dehumidifier monitors. These methods were used to monitor the total releases of tritiated water from the reactor stacks. Initially, direct measure of the loss of moderator from the reactor was used to determine tritium losses, but this measured both atmospheric and direct liquid releases, so tritiated water releases in the stack were monitored.

Silica gel was used to measure tritium vapor release from the reactors and to determine the amount of moderator that was lost from the reactor stacks (Miller 1956a). This method was difficult and inaccurate, so there was a shift to dehumidifiers (Miller 1956a; Kiger 1955).

A bit later, Kanne ionization chambers were also employed for online monitoring (Caldwell 1958). Kanne chambers cannot differentiate between tritium and other radioactive gases, so they were used as real-time monitors to alarm at preset activity levels and not to determine the quantitative atmospheric tritium releases (Longtin et al. 1973).

In the late 1950s, the two methods of estimating total tritium releases in ventilation air discharged from the reactor buildings (e.g., 105-R) were cited in Caldwell (1958) as follows:

1. Inventory losses derived from observed reductions in moderator inventory. This was considered accurate within a factor of 2 because of the difficulty in differentiating stack release from other moderator losses through sump collection, losses to the disassembly basin water during reactor discharge, or other mechanisms.
2. Computing stack releases based on the percent of moderator (D_2O) contained in the water samples condensed from the stack stream. A dehumidifier was operated three times a week for a total of 24 hours during normal operations, but it was not used during reactor outages. This method provided an estimate of the amount of moderator that was lost and, therefore, the total tritium released during the measurement period, but it did not provide rate or time of releases. For this reason, it was considered a good method of monitoring only during continuous routine operations.

Kanne ionization chambers were considered the most practical systems available. They were used in reactors for online alarm systems and at the Tritium Facilities for personnel monitoring and for stacks and central air ducts monitoring (Conway 1956). A central system connecting sampling lines to numerous points, controlled by valving, was considered ideal. By 1965, Tyson (1965) noted the need for the new and improved method, the stack tritium monitor (STM).

1970s

For each reactor process, releases were monitored in the stack at +148 ft and in three exhaust ducts. A 1-in. diameter sample probe in the stack at +148 ft carried an aliquot of stack effluent to the Kanne chamber, which was set to alarm at a predetermined point. Because any noble gas activity present in the stack effluent would be added to the tritium activity measured, the Kanne chamber was not used to obtain quantitative measures of total tritium released.

The daily stack tritium discharge was calculated based on a sample of water collected by a continuously running dehumidifier (apparently no longer run only 3 times a week). The water

sample was counted in a liquid scintillation counter with a sensitivity of 3×10^{-7} µCi mL^{-1} (±10%). The daily tritium stack loss was calculated based upon these sample results, humidity, and effluent airflow rate (Longtin et al. 1973; Jacober et al. 1973; Reinig et al. 1973).

A new effluent air monitoring system, designed in the 1960s (Tyson 1965), was evaluated in the early 1970s—the STM. This system consisted of two ionization chambers in parallel with a moisture trap ahead of one chamber to remove HTO. The difference in the signals from the two chambers was proportional to the HTO concentration. This system eliminated background and noble gas interference. It measured only tritium in the oxide form, but nearly all the tritium released from the reactors was in that form (Longtin et al. 1973; Jacober et al. 1973). This system became the primary monitoring method used in the reactors.

In the disassembly bay (in 105-P), no tritium stack monitoring appeared to be implemented. Atmospheric releases were estimated based on area, not the exhaust vent, monitoring (Reinig et al. 1973; Longtin et al. 1973). For the purification makeup room exhaust (105-C and 105-K) no monitoring for tritium was done. In 1973, it was recommended that the air be rerouted to the purification exhaust, which was monitored (Reinig et al. 1973; Longtin et al. 1973). For the purification resin preparation and cold pipe space exhaust (105-P), no tritium monitoring was carried out; it was also recommended that the exhaust be rerouted to the purification exhaust. In the heat exchanger decontamination facility (105-C), no tritium monitoring and grab sampling was recommended (Reinig et al. 1973).

1980s

Documentation from the 1980s showed that losses from the process water system were directly detected by process water loss (volume or pressure drops) and measurement of tritium in the heavy water vapor that evaporated from the leaks in the cooling system to the stacks. The tritium in the D_2O ranged from 3000 to 13,000 µCi mL^{-1}. Tritium concentration in the moderator was measured weekly by laboratory analysis, and daily airflow in the stack was measured by taking the average pressure drop across the carbon filters (Smith 1989).

STM and a new Berthold tritium monitor (BTM) were both being used by the end of the 1980s. Since September 1985, two BTMs had been operating in L-Reactor and K-Reactor for a combined total of 2.5 years of in-service operation (Merz 1988). The BTM was recommended as the primary method to measure tritium releases.

Smith (1989) reported on a leak before break program (LBB), which was established to provide a technical and operational basis for the new design basis loss-of-coolant accident for the emergency cooling system in the reactors. It dealt with the tritium monitors, airflow measurements, and moderator tritium analysis. The leak detection instrumentation consisted of the BTMs, STMs, and their associated equipment. These instruments were used to indicate a leak in the primary coolant system with a 35% error in measurement. The error in the measurements was the result of a 15% error in the detection of the tritium by the tritium monitors, a 15% error in the stack airflow measurement, and a 5% error in the moderator tritium analysis. The BTMs were checked for accuracy every 6 months. Daily comparisons between the BTM and STM results were made to ensure that the instrument readings were comparable (Smith 1989).

In a 1986 review of radiological effluent monitoring, analytical techniques, and reporting, Zeigler (1986) reported on the current status of tritium monitoring at the SRS and the factors contributing to the uncertainty of the measurements, stating that:

> There are a few minor radioactive effluents that are monitored by Area Survey HP personnel, but the releases are not tabulated and reported because they are small . . . Overall the most important improvements needed in SRP effluent monitoring relate to measurement of volumes of air and liquid that are released. In many cases, volumes are estimated based on fan or pump capacities or on measurements that were made at some time in the past. Frequently, there is no documentation of the methods used to obtain the volumes that are used.

In the 1980s, the following measurements were made in the reactor areas:

105-P, 105-L, and 105-K Stacks and Disassembly Area. Department personnel collected a daily dehumidifier sample from the 195-ft stack and a silica gel sample weekly from the disassembly area for tritium analysis (no direct effluent monitoring was done for the disassembly stack). The tritium stack losses were calculated using the STM results to confirm losses determined by the dehumidifier sample. The 195-ft stack flow rate was determined from daily readings of flow measuring instruments. The disassembly building exhaust flow was determined from the manufacturer-cited capacity of the fans, checked by periodic measurements of actual flow. The releases from the disassembly area were based on the area air tritium concentration.

The uncertainty resulting from this method arises because the disassembly area does not have a stack and, therefore, no well-defined exhaust. The facility was maintained under positive air pressure, so the air was exhausted through doors and other openings in the facility. This factor would have introduced some error in the estimated releases. The HT/HTO ratio from these areas was not documented.

105-C Stacks and Disassembly Area, Stack Areas Decontamination Facility, and Sand Blast Facility in 717-C. Silica gel samples were taken 1 day a week in the disassembly area and were assumed to represent the whole week (value multiplied by 7). A daily dehumidifier sample from the stack was analyzed. Stack airflow was determined from weekly readings of flow measuring instruments. Disassembly airflow was determined based on the capacity of the stack fans with periodic measurements. By only periodically measuring the exhaust air, some additional uncertainty was introduced. In addition, there was no characterization and documentation of the HT/HTO ratio.

A schematic of 105-C Reactor shows the airflow to the 200-ft stack and the location of the sampling system (Figure 4.1-1).

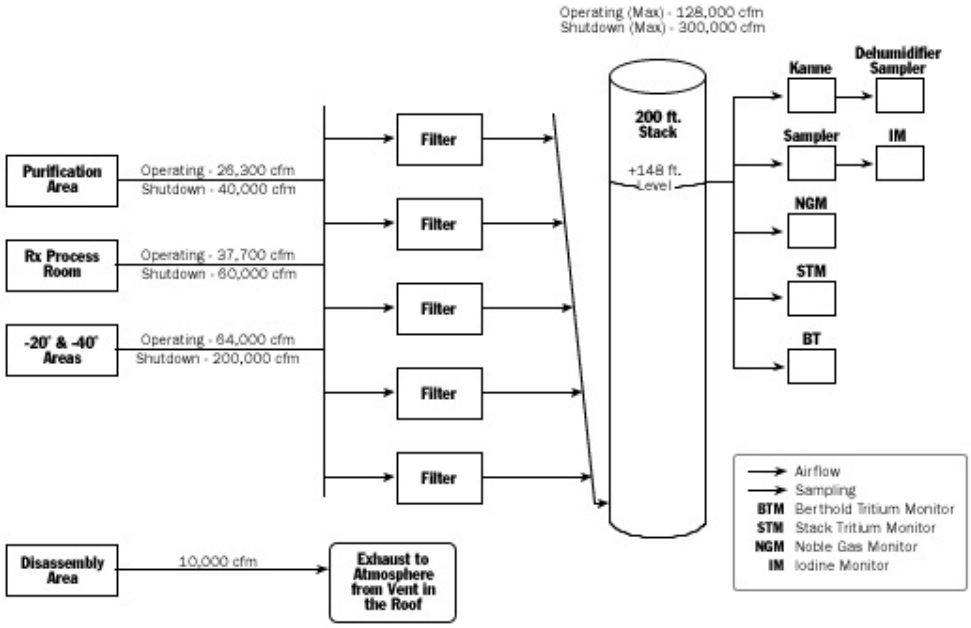

Figure 4.1-1. Airflow schematic of 105-L-Reactor (Westinghouse 1991a).

Reactor Stack Releases

In 1956, it was believed that tritium releases were largely a result of tritium in the moderator and that there were practically no losses from lithium-aluminum target slugs during reactor operations. Nonmoderator losses were believed to occur only on the rare occasions when the slug was cut into accidentally when cutting through control rods. In this event, the release is a small fraction of what is contained in the slug and dissipates rapidly; it could not be measured (Miller 1956a). At that time, the 100-Area tritium releases to the atmosphere were considered low enough to be neglected except in the case of a major reactor failure. Releases of T_2O or DTO averaged 590 curies per month (Ci mo^{-1}) per area during May–August 1956. An increase was expected due to power ascension, but the routine releases were not expected to exceed 5000 Ci mo^{-1}. Compared to the expected Separations Area (200 Area) releases, this increase was considered negligible (Denham 1956).

Original estimates of tritium production in the moderator (from the 100-Area Technical Manual) were 0.5 mCi/mL/y production in the moderator at 700 MW because of neutron capture by the deuterium. Actual results showed a 20% fluctuation about the expected value, so the expected rate was 0.002 μCi/mL/MWd. This estimate could not be checked daily, but monthly it was reported to match reasonably well with tritium concentration estimates (Miller 1956a).

The atmospheric release points from the reactors are the 200-ft stack and ground-level evaporation from the disassembly basin water shown in Figure 4.1-1. An additional source of atmospheric tritium release is the ground-level evaporation of water purged from the disassembly basin to a seepage basin. Tritium in the form of DTO (from the moderator) is released to the ventilation system and, therefore, to the stack by evaporation of process water exposed to air

flowing through the process area. During reactor operations, D_2O containing DTO is evaporated from small leaks in pipe flanges, valves, and from the D_2O process water exposed in the control rod guide tubes. During reactor shutdown, DTO is evaporated when pipes or valves are opened for inspection and maintenance work and when wet fuel, target, and control rods are discharged from the reactor (Dukes and Benjamin 1982).

Although the discharged fuel and target assemblies are rinsed with water before being placed in the disassembly basin, some tritium is transferred to the disassembly basin with process water that adheres to the assemblies and from DTO contained as water of hydration in the aluminum oxide on the assemblies. Disassembly basin water was discharged to the seepage basin after being recirculated through filters and deionizers to clarify the water and remove radionuclides. The tritium concentration in the disassembly basin water increased following each reactor discharge. When the set level reached 0.2 to 0.4 $\mu Ci\ mL^{-1}$, the water was purged to the seepage basin through filters and deionizers. Tritium was not removed during this process. It was estimated that about 30% of the tritium purged evaporated from the seepage basin each year (based on average atmospheric conditions) (Dukes and Benjamin 1982).

By 1971, atmospheric tritium releases were 210,000 Ci from the reactors, 400,000 Ci from the processing operations, and 9,000 Ci from D_2O recovery (Jacobsen 1972); 94% of the tritium was released to the atmosphere and 6% to aqueous effluent streams. In 1973, the situation was about the same; 63% of SRP tritium releases resulted from processing the irradiated lithium and uranium and 37% from various sources associated with the reactor moderator.

The possibility of decreasing the loss of D_2O (coolant) to offset the increase of tritium with time was discussed in 1972. The heavy water loss reduction was shown to have partly offset the increasing tritium concentration in the moderator. Some of the activities taken to reduce heavy water losses were described. The average tritium concentration in the reactor stack air was noted as $5 \times 10^{-4}\ \mu Ci\ cc^{-1}$ (Jacobsen 1972).

In the 1980s, further efforts were made to reduce the reactor releases with an intensive leak reduction program and provisions for recovery of D_2O carried on discharged fuel. The D_2O loss rate was reduced, but the increase of tritium in the moderator caused a lesser reduction in the tritium releases. It was too expensive to introduce fresh D_2O or remove the tritium from the moderator, so improvements were planned to the leak detection and recovery systems for the processing of irradiated lithium. About a 25% reduction in tritium releases was expected to be accomplished at a reasonable cost (Dukes and Benjamin 1982). Further reference of this program was not found in the document database.

In 1981, the reactor areas released 1.3×10^5 Ci to the atmosphere. Of this number, 1.2×10^5 was from stack releases, 5.4×10^3 Ci (92%) was from disassembly basis, and 5.6×10^3 Ci (4%) was attributed to seepage basin evaporation (Crawford and Roggenkamp 1983).

Westinghouse (1991b) indicates that although the reactors were not working, stack monitoring of tritium, ^{41}Ar, and particulates continued at all times. Releases from the reactors were due to the evaporation of tritiated water; therefore, tritium released from these facilities was in the form of tritiated water (HTO). Virtually all the tritium was released to the atmosphere through the stack. Some very small fraction (approximately 1%) was released through evaporation from fuel and target storage basins (Murphy et al. 1991). Tritium losses from the reactor area were estimated to be 100% oxide (Miller and Patterson 1956).

SEPARATIONS AREAS

History

Tritium target processing operations began in October 1955 in a small facility (232-F) and moved to larger quarters in July 1957 (232-H and 234-H). In 1957, tritium was first loaded into reservoirs that are components of nuclear weapons. Tritium was also recovered, purified and reloaded into weapons components. By 1958, the capacity of the recovery operation was doubled. Today, there is no tritium production at the SRS. Tritium that was produced in the past is recycled, mixed, and reloaded in the new Replacement Tritium Facility, which began operations in 1994. This new facility has replaced most of the SRS facilities that processed tritium during the last 35 years.

Tritium Processing Facilities Stack Monitoring

Early years

In the mid 1950s, the Health Physics Group expected that stack monitoring in Building 232-F would be an easy task, but they soon found that more attention to detail was needed to produce good results (Landon 1960). The original system consisted of 1-in. stainless steel sampling line running from the stack plenum into a Kanne system in the air monitor room. The Kanne system monitor measurement output (in curies per meter) was recorded on a paper strip chart that moved at a constant rate. The amount of gas leaving the stack was determined by manually integrating the area under the curve (Landon 1960). Miller and Patterson (1956) reports that based on experimental results (which are not presented in this memo), the figure of 60% oxide and 40% tritium gas is used for the releases from the 232-F facility slow leak in Building 232-F, which went undetected for about 12 days in July 1956. As a result, the reliability of the monitors was carefully investigated and the representativeness of the sampling locations for air sampling points was checked (Miller 1956b). By mid-1956, the adequacy and performance of the system was in question and there was an interest in a more quantitative measurement of stack losses. A test of the system with a series of known releases showed that the system varied as much as ±80% because of two sources of error: the manual integration and inadequate sampling of unmixed air streams. Adding more sampling points showed that activity in different quadrants varied by as much as a factor of 2, so the sampling point was relocated at the 50-ft level in the stack. Comparison between the two sampling points showed that the values were 7% lower at the 50-ft point (Landon 1960).

To improve the accuracy of the quantitative integration of the curve, the paper strip chart speed was increased by a factor of 5 to enlarge the area under consideration and more accurately integrate brief releases. An electronic integrator was installed on July 24, 1957, to digitally integrate the cumulative curies of tritium discharged from the stack. A comparison of the old and new methods showed that electronic integration results were 9% higher than manual integration results. The relatively large releases from furnace changes and discharges were in closer agreement at 2% (Landon 1960).

The electronic integrator, with seven-decade dynamic range, was built to track the total amount of radioactive material released from the 232-F stack. The output current from the Kanne

chamber was recorded by a Beckman logarithmic electrometer and a Brown strip chart recorder. The control shaft followed the position of the Brown recorder indicator by a servomechanism. An oscillator attached to the control shaft oscillated with a frequency that was exponentially proportional to the vertical position of the control shaft. This output frequency was linearly proportional to the Kanne chamber current. A 13-decade counter totaled the cycles from the oscillator to integrate the amount of radioactivity released. Because the oscillator had a dynamic range of only three decades (20 to 20,000 counts per second [cps]), an automatic switching circuit was provided to inject the signal pulses at different decade positions in the counter chain to perform the function of frequency multiplication (Du Pont 1957a).

This integrator was used in 232-F until a few months before shutdown, when it was moved to 232-H. Originally, its maximum range was 6300 Ci min^{-1}. This was expanded after two stack releases exceeded that value (Landon 1960). After the electronic stack integrator was installed in 1958, a 2-week calibration period followed. Manual and electronic integration were compared, after which a controlled tritium release was used to check the monitor (Du Pont 1958a: August 30, 1957–May 9, 1958). The stack integrator was placed in routine operation, and it agreed with the manually integrated values to within 20% (Du Pont 1958a: September 13–19, 1958). Accuracy of these measurements through time was unknown, and it was concluded that the electronic integrator was not yet to be totally trusted (Miller 1959). Unfortunately, no further details were provided. It was not until the mid-1970s that the stack monitor integrator (SMI) was introduced as a primary method of monitoring stack releases (Kilpatrick 1975). Calibration of the stack monitor with known quantities of tritium was discussed in the January 1958 Monthly Progress Report (Du Pont 1958b). It was indicated that the reported losses were 15 to 20% below the actual values. In the July 1958 Weekly Progress Report (Du Pont 1958a: July 1–11, 1958), the accuracy of stack monitor results was questioned during one 2-day period when the Beckman amplifier was known to be drifting as much as 25%. After the amplifier was repaired, the integrated and analyzed results from a controlled tritium release agreed within 2%. By the third week in August, the total discharge to the stack was measured and agreed within 1% of design flow (Du Pont 1958a: August 23–29, 1958).

The stack monitor with controlled releases was not calibrated during the third week in September 1958 because of problems with leaks in sampling equipment (Du Pont 1958a). A controlled tritium release to the stack, carried out during the second week in October 1958, showed that calculated and integrated results agreed within 10%. Stack monitor response to the probe source was within limits the entire week (Du Pont 1958a; Du Pont 1958c).

Miller (1959) is a response to a report by the Atomic Energy Commission (AEC), the U.S. Government agency overseeing the national laboratories, that reviewed the accountability of nuclear materials at SRS. This memo indicates that in February 1959, the amount of tritium that was captured in the zeolite (stripper) beds in 232-H and 234-H was not known. AEC had recommended that better methods were needed to determine the quantities contained in the beds. (Zeolite beds were installed to capture tritium that was released during routine operation. In addition to providing protection to the environment, it enabled the capture and recharged of the tritium so it was not lost.) The method would attempt to complete a mass balance of all tritium produced; the losses determined by pressure, volume, and temperature measurement; and mass spectrometric analysis of the remaining tritium contents of process vessels before discharge to the stack.

The Health Physics Department reported tritium releases hand-integrated from the Kanne monitor paper strip charts (considered to have a ±10% uncertainty) (Miller 1959). When the results of the stack monitor and the spectrometric analysis of the material believed to be released were compared, there appeared to be a discrepancy. The difference resulted from the lack of measurement of all process vessels by the mass spectrometer (resulting in lower values than the Kanne monitor). It was concluded that the releases were recorded by the stack gas monitor (Miller 1959).

In a report on the accidental release of tritium gas from the tritium processing facility on December 31, 1975, the FORMS sampler, designed to differentiate between the forms of tritium released, was used. In addition to the Kanne monitors for total tritium releases, it was indicated that they have been collecting separate samples of elemental tritium gas and tritium oxide since August 1974 (Jacobsen 1976). In 1978, Johnson recommended that the HT/HTO atmospheric release data should be monitored from 232-H and 234-H continuously (Johnson 1978).

No other specific information on changes in the tritium monitoring equipment at the Tritium Facilities was found for this era, other than the full implementation of the SMI in the 1970s.

1980s

In June 1982, it is reported that a 6500 ft^3 hold volume tank was installed in 300-H to contain accidental releases of tritium from 234-H (Du Pont 1982). Sensors in the process hoods were designated to automatically trigger diversions to the hold volume tank. Before this change, the monitoring capabilities of tritium diverted to the hold volume were limited to approximately 0.05 g (about 470 Ci). A monitor-integrator was installed to provide sufficient information on the activity in the hold tank to decide whether to recover the tritium.

Zeigler (1986) presented a review of radiological effluent monitoring, analytical techniques, and reporting. This included the current status of tritium monitoring at the SRS and the factors contributing to the uncertainty in the measurements. He stated that,

> There are a few minor radioactive effluents that are monitored by Area Survey HP personnel, but the releases are not tabulated and reported because they are small. Overall the most important improvements needed in SRP effluent monitoring relate to measurement of volumes of air and liquid that are released. In many cases, volumes are estimated based on fan or pump capacities or on measurements that were made at some time in the past. Frequently, there is no documentation of the methods used to obtain the volumes that are used.

Zeigler (1986) reported the following measurements being made at the Tritium Facilities in 1986:

> To determine the total tritium releases from the Tritium Facilities stacks (232-H [2 stacks], 234-H, and 238-H stacks), the peak areas on Kanne recorder charts were integrated using empirically determined factors. Automatic integration of these releases was also available. The form of tritium is determined by an on-line FORMS monitor in 234-H and by molecular sieve samplers in 232-H. Back-up molecular sieve samples were also collected in 234-H. Molecular sieve samplers were also maintained

in the 232-H stacks by the Environmental Transport Division of SRL. All tritium released from 238-H was assumed to be in oxide form. On-line tritium FORMS monitors are also being installed in 232-H stacks and were expected to be operational by the end of 1986. Stack flows were determined manually when requested specifically, although there is no frequency set for these measurements.

The 772-F stack is not monitored for tritium at this time, but a new system is being installed. The tritium releases from the 244-H, RBOF stacks, are determined from the stack Kanne chamber by integrating areas on the recorder chart. The releases are reported in monthly radioactivity release reports. The F and H areas tank farm waste tanks, diversion boxes and pump pit exhausts are not routinely measured for tritium releases. A HP survey done in 1983 indicated that the annual tritium releases from 11 of the most likely tanks to contain tritium were less than 100 Ci y^{-1}.

An Air Effluent Monitoring Procedure for the FORMS monitor indicated that in the 1990s, FORMS monitors were operating on Lines 1, 2, and 3 in 232-H and in 234-H (Westinghouse 1991c). Further explanation of how the monitoring results from the molecular sieves and FORMS monitors were used was not found. Figures 4.1-2, 4.1-3, and 4.1-4 show the airflow schematics indicating the air sampling points in facilities 232-H, 234-H, and 244-H, respectively.

Releases

In 1956, 232-F tritium loss was determined based on material balance from product input after extraction (Miller 1956b). It seems that there was considerable interest in limiting the releases because of economic considerations. Stack losses from 232-F averaged 30,000–40,000 Ci mo^{-1} under routine operating conditions during 1955 and 1956. These releases were larger than had been predicted. The primary sources identified were miscellaneous process leaks and the extraction furnaces. It was reported that the experience in 232-F showed that ≥60% of the tritium released was quickly converted to the oxide through exchange with water vapor, but no data were presented to support this report (Denham 1956).

Start up of tritium processing in 232-H was scheduled for July 1, 1957 (Du Pont 1957b). In August 1957, a zeolite bed for absorption was installed in 232-H Line 1. It was estimated that a 33% reduction in tritium losses occurred after installation (Harris 1957). Harris (1957) speculated that based on this result, 80% recovery of product could be attained. Estimates were made for the minimum amount of product that would be lost from the 232 Buildings: 232-F 1.5 l d^{-1} or 12 g mo^{-1} (or about 117 kCi mo^{-1}); 232-H 4.5 l d^{-1} or 36 g mo^{-1} (or about 350 kCi mo^{-1}); and 234-H 7.5 l d^{-1} and 60 g mo^{-1} (or about 585 kCi mo^{-1}).

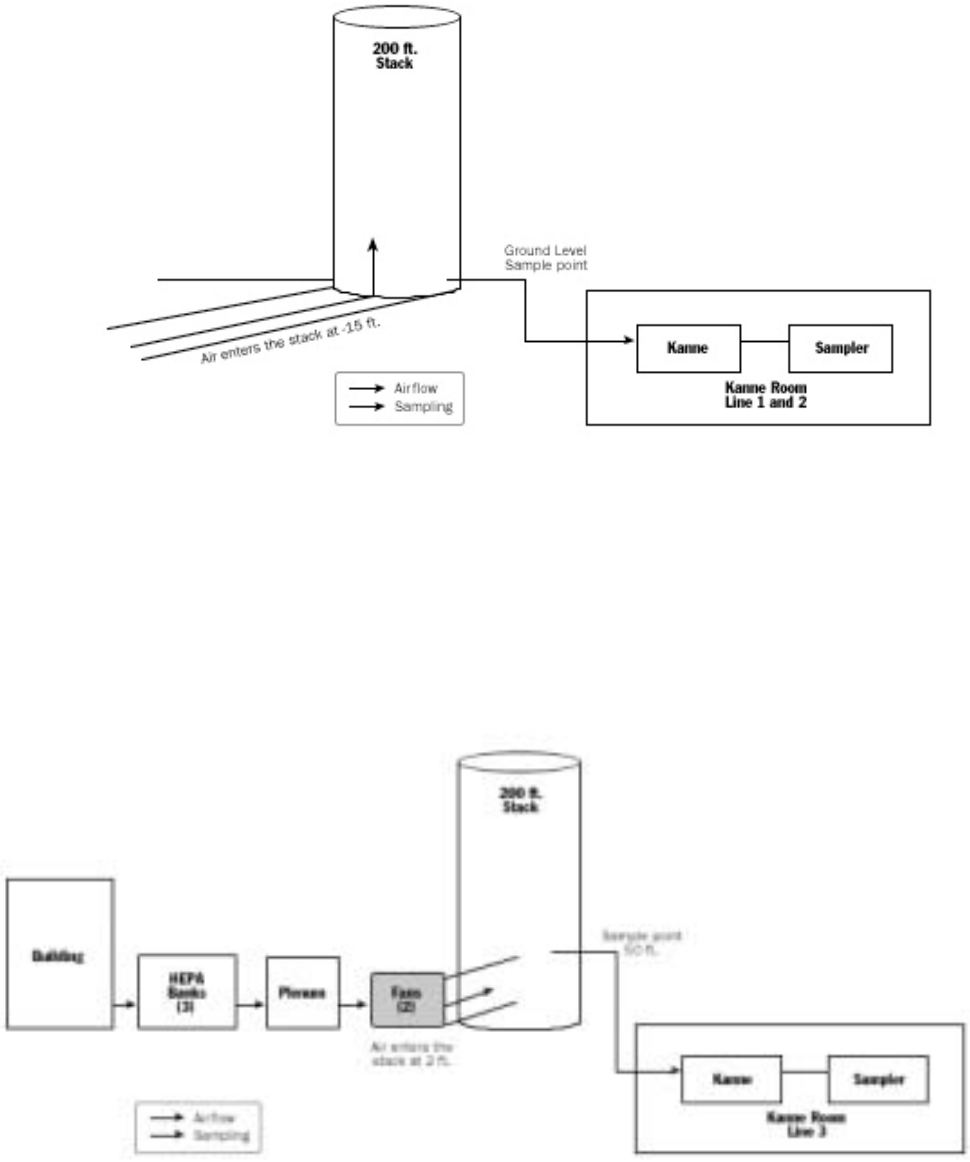

Figure 4.1-2 a and b. Airflow schematic for the lines in Building 232-H (Westinghouse 1991a).

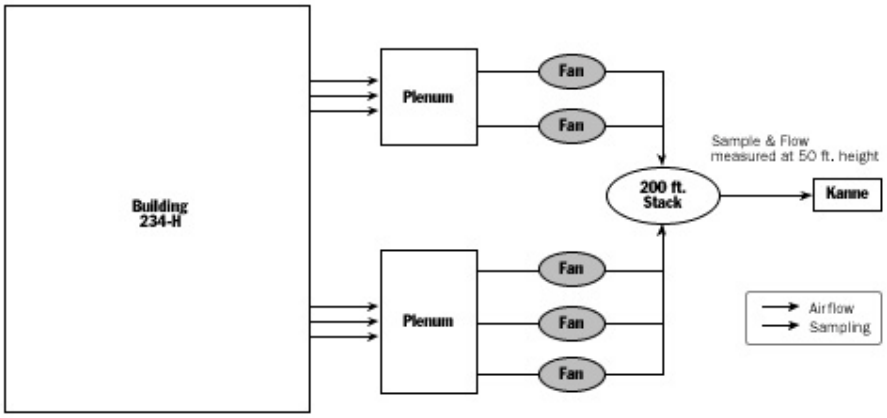

Figure 4.1-3. Schematic of air flow in Building 234-H.

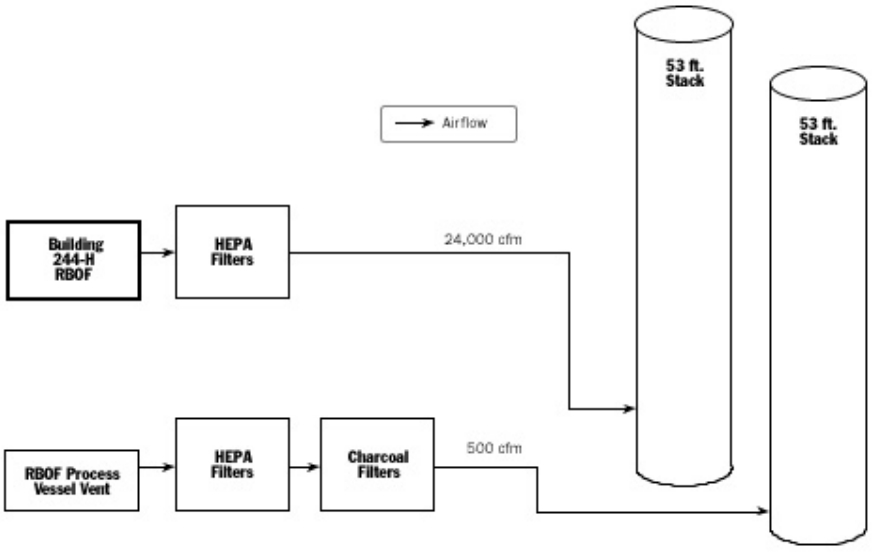

Figure 4.1-4. Schematic of airflow in Building 244-H.

In September, stack losses from 232-H were reported to be high during the first 20 runs of the month, but they were reduced (McKeller 1957). Most of the releases were due to by-product and first evacuation gas. McKeller (1957) indicated that the use of a stripper brought about a 70% reduction in the first evacuation and a lesser reduction in the by-product loss. This appears to be referencing the zeolite beds installed, but the memo also indicates that "the process design for a permanent stack stripping system is being developed."

During the early operation of 232-H, the product losses shown by the stack monitor were higher than expected, so the accuracy of the stack monitor's response was questioned (Butler et al. 1957). A series of tests was carried out to look for possible reasons for the discrepancy. These tests included

- Surveys for particulates
- Effects of argon introduction
- Release of known quantities
- Measurement of the presence of oxides during different operations.

Known releases of 25 Ci and 130 Ci were introduced into the main air exhaust and showed that the monitor results were within 10% of the amount released, confirming the calibration of the monitor. Additional work showed that about one-half of the product released was in oxide form. It is not clear if this determination of the chemical form of the tritium released was a result of the same work mentioned in other documents. Tests showed that the addition of argon gas acted only as a dilutant in the Kanne chamber (Butler et al. 1957).

Moisture in the furnace appeared to be a main cause of high stack losses in 232-H. An additional zeolite bed was added to the test stripper assembly. It had one copper oxide (CuO) and two zeolite beds in series. Data from one run indicated that 50% of the losses came from the last part of the run when the effluent had not normally been stripped. The memo also indicated that the following determinations were planned as part of the permanent stripper evaluation; however, documentation of the results was not found:

- Oxide conversion efficiency
- Oxide absorption efficiency
- Measurement of total water from the system
- Recovery form zeolite
- Oxide decomposition.

The largest losses occurred during the charge-discharge operation in 232-F. The drop in releases is attributed to cleanliness of the system. Increased losses during zeolite bed trials could have been associated with gradual buildup of condensed metal distillate on the furnace cover. The losses in 232-H gradually decreased after the zeolite beds began operation (Bird 1957a). In December 1957, it was reported that an efficient permanent stack stripper was installed on Line 2 (Bird 1957b).

The variation of the form of the release of tritium (elemental, HT, or tritiated water vapor, HTO) depended on the nature of the work being done in the facilities. Murphy et al. (1991) cites studies that show the fraction of HTO, the form of greater concern to this study, was 56% over a 3-year period in the 1970s. In a 3-year period in the 1980s, the fraction of HTO was cited to be 48% (Murphy et al. 1991). The fraction HTO released was likely dependent on the type of operation carried out at the facility. Because this information is not available, it would not necessarily be valid to assume the fraction HTO released on a given day, week, or month; however, at least some data are available to assume about 50%. Unless other data are found, it

can be assumed that about one-half of the tritium releases from these facilities was HTO, which is representative of operations after 1970. The section on inadvertent releases in this report presents cases of large inadvertent or accidental releases where the ratio of tritium to tritiated water (HT/HTO) was known.

TRITIUM SOURCE TERM RECONSTRUCTION

The releases of tritium from the SRS are generally very well documented on a monthly basis in published internal reports. Records of releases have been kept since almost the beginning of the operations. Yearly data are very well summarized for all main release points in two summary documents published in 1991 (Cummins et al. 1991; Murphy et al. 1991). In an effort to corroborate the summarized data, we conducted a record search to find original data to reconstruct the monthly releases wherever possible. We reviewed hundreds of documents, including numerous facility report series. The main sources of information were SRS weekly and monthly reports. The best estimate of the source term has been compiled on monthly where possible (in some of the earlier years, it was compiled biannually). Atmospheric tritium releases were separated into reactor releases and tritium facility releases. Detailed data are available electronically via hyperlink by selecting either release type above.

The documents used to develop the source term compiled in the above data files have not been cited in connection with the individual release values, but they are listed in the reference list at the end of the chapter by year, in the ”Reference - Source Term” section. Lee (1998b) contains even more detailed tritium release data.

Whenever possible, we checked the release values with shift or daily data found on datasheets or handwritten laboratory logbooks for both the reactors and tritium facilities.

In general, there is quite good agreement between the different release values reported in the different sources. In fact, in most cases, the published aggregated values are larger than the sum of the logbook or datasheet values reported for shorter time scales. Because in most cases the summary values were within 10% of the published values, it is likely that most “unchecked” monthly values are also valid. It is possible to conclude that other small sources were not published independently in regular reports, but they were included in the Site-wide totals. It is also possible to speculate that the reported values were “rounded-up” when published. It is, however, impossible to accurately reconstruct a daily or weekly history of releases.

No consistent monthly release trends across the year were found for the reactor data, so it was not considered prudent to artificially break down the 6-month and annual totals by month (when monthly data were unavailable). When data for releases from the reactor disassembly area were available, they were included (at the end of each reactor section). The total annual releases from each reactor are plotted over time in Figures 4.1-5, 4.1-6, 4.1-7, 4.1-8, and 4.1-9. Figures 4.1-10, 4.1-11, 4.1-12, 4.1-13, and 4.1-14 provide plots of the annual releases from the Tritium Facilities.

Lee (1998a) concluded that the SRS reactor data found were insufficient to reproduce the releases reported by SRS using original raw data. Based on the comparisons that could be made, evaluation of equipment and methods, and the uncertainty analysis, we have concluded that it was possible to reconstruct a credible set of best estimate tritium monthly release data from the reactors and Tritium Facilities. These best estimates should be used as a range suggested by the

uncertainty analysis and should be considered when performing out future steps in the dose reconstruction process.

Based on the review of the available data sources, we concluded that the SRS reported the monthly tritium release data that were measured at the facilities. The review of the quality of the atmospheric tritium release data reported by SRS has given us no indication that serious or consistent errors were made in monitoring Site tritium releases. Details of this review are provided in Appendix E.

The releases are reported by month because it was not possible to reconstruct daily releases for the whole operational period of SRS for all facilities. During the next phase of the SRS dose reconstruction, it will not be possible to model daily environmental dispersion and human exposures. The next section summarizes the uncertainty calculations that we performed for the atmospheric tritium source term. See Appendix E for details of these calculations.

Table 4.1-2 provides an overview of the data that were used to develop the tritium source term. All documents used to develop the source term are included in the reference list at the end of this chapter (organized by year).

Table 4.1-2. Tritium Release Documentation Used To Reconstruct the Source Term

Year	232-F	232-H	234-H	238-H	244-H	Reactors	Others
1954	monthly; beginning in Nov.					monthly, start in July	
1955	monthly; Jan–Nov					monthly Jan–Sept, biannual	
1956	biannual	biannual (July–Dec only)	biannual (July–Dec only)			biannual	
1957	biannual; monthly (except Nov.–Dec.)	monthly (except Aug.)	monthly			biannual	
1958	CLOSED	monthly	monthly			biannual	
1959	⇓	monthly	monthly			biannual	biannual
1960		monthly	monthly			biannual	biannual
1961		monthly	monthly			biannual	biannual
1962		monthly	monthly			biannual	biannual
1963		monthly	monthly			monthly	
1964		monthly	monthly			monthly (R until July)	biannual
1965		monthly	monthly			monthly	biannual (July–Dec only)
1966		monthly	monthly			biannual	biannual
1967		monthly	monthly			monthly	
1968		monthly	monthly	START OF OPERATION		biannual (L from Jan-July only)	annual
1969		monthly	monthly	monthly (beginning in May)	START OF OPERATION	biannual	annual
1970		monthly	monthly	monthly	monthly (for some months)	annual	annual
1971		monthly	monthly	monthly	monthly (Jan-Feb. only)		
1972		monthly	monthly	monthly		monthly	
1973		monthly	monthly	monthly		monthly	monthly
1974		monthly	monthly	monthly		monthly	monthly
1975		monthly	monthly	monthly	monthly	monthly	monthly
1976		monthly	monthly	monthly	monthly	monthly	monthly
1977		monthly	monthly	monthly	monthly	monthly	monthly
1978		monthly	monthly	monthly	monthly	monthly	monthly
1979		monthly	monthly	monthly	monthly	monthly	monthly
1980		monthly	monthly	monthly	monthly	monthly	monthly
1981		monthly	monthly	monthly	monthly	monthly	monthly
1982		monthly	monthly	monthly	monthly	monthly	monthly

Table 4.1-2. (Continued)

1983		monthly	monthly	monthly	monthly	monthly	monthly
1984		monthly	monthly	monthly	monthly	monthly	monthly
1985		monthly	monthly	monthly	monthly	monthly	monthly
1986		monthly	monthly	monthly		100 areas total	
1987		monthly	monthly	monthly			
1988		monthly	monthly	monthly			
1989		monthly	monthly	monthly		monthly	monthly
1990		monthly	monthly	monthly			
1991		monthly	monthly	monthly		monthly	monthly
1992		tritium facilities total (monthly)	tritium facilities total (monthly)	tritium facilities total (monthly)	tritium facilities total (monthly)	monthly (except Nov.)	monthly (except Nov. and Apr.)

Uncertainty of Source Term Estimates

The accuracy of the atmospheric source term estimates is largely dependent on the type of monitor used to measure the releases. A detailed description of the equipment and the data used in our uncertainty analysis appears in Appendix E. It has not been possible to determine the exact times that each type of equipment was installed and operated in each of the facilities. However, in reviewing the documentation, it is possible to find dates on equipment operation procedures or reports of plans or prototype tests.

For the purposes of this work, general use of the various tritium monitoring systems that were used onsite since the 1950s are documented as

Dehumidifier	1954–1986 possibly longer (reactors)
Silica gel	1954–1958 (reactors and disassembly areas)
Kanne ionization chambers	1954–present (Tritium Facilities)
Stack tritium monitor	1970–1988 (reactors)
Berthold tritium monitor	1988–present (reactors)
Stack monitor integrator	1974–present (Tritium Facilities)
FORMS	1985–present (Tritium Facilities and reactors)

It was not possible to determine exact start and stop dates because systems were tested and then kept running in parallel to provide some redundancy if the main system failed. It is not clearly stated when the measurement results of one system became the primary monitoring device in replacement of an older version. For example, in early years, it is not clear when silica gel was no longer used for stack monitoring (replaced by dehumidifiers) and shifted to general area monitoring for health physics protection purposes. The documentation we found refers to both the silica gel and dehumidifier monitors being used for stack monitoring.

The uncertainty assessments for each of the monitoring systems have taken into account the representativeness of the sample; the frequency of sampling; efficiency of sampling; the effects of environmental conditions; the accuracy of the measurements; and finally, errors in reporting results. Using a Monte Carlo sampling technique, we developed uncertainty estimates, which are reported in Table 4.1-3. Details of the analysis and further explanations can be found in Appendix E.

Table 4.1-3. Uncertainty Estimates for Tritium Releases Reported by Monitors on SRS

Monitor	For results in years	Facilities	Distribution	Range[a]
Silica gel	1954–1958	Reactors	Lognormal	GM = 1.15 GSD = 2.16
Dehumidifier	1954–1970	Reactors	Lognormal	GM = 0.99 GSD = 2.3
Stack tritium monitor	1970–1988	Reactors	Lognormal	GM = 0.91 GSD = 2.19
Berthold tritium monitor	1988–present	Reactors	Lognormal	GM = 0.99 GSD = 1.16
Kanne chamber	1954–1974	Tritium Facilities	Lognormal	GM = 0.9 GSD = 1.8
Stack monitor integrator	1974–present	Tritium Facilities	Lognormal	GM = 0.9 GSD = 1.8

[a] GM = geometric mean, GSD = geometric standard deviation, SD = standard deviation.

To apply these results to a monthly tritium atmospheric source term, use the monthly value for a given facility and select the appropriate uncertainty distribution (based on year and facility). To obtain the median value on the distribution, multiply the monthly best tritium release estimate by the geometric mean (GM). To obtain the 5th percentile value, divide the median value by the square of the geometric standard deviation (GSD). The 95th percentile is calculated by multiplying the median value by the square of the geometric standard deviation. The uncertainty for annual tritium release estimates has been calculated, and is compiled in a reactor release file and a tritium facility release file. These tables contain annual release estimates along with 5th and 95th percentiles and the calculated GM and GSD.

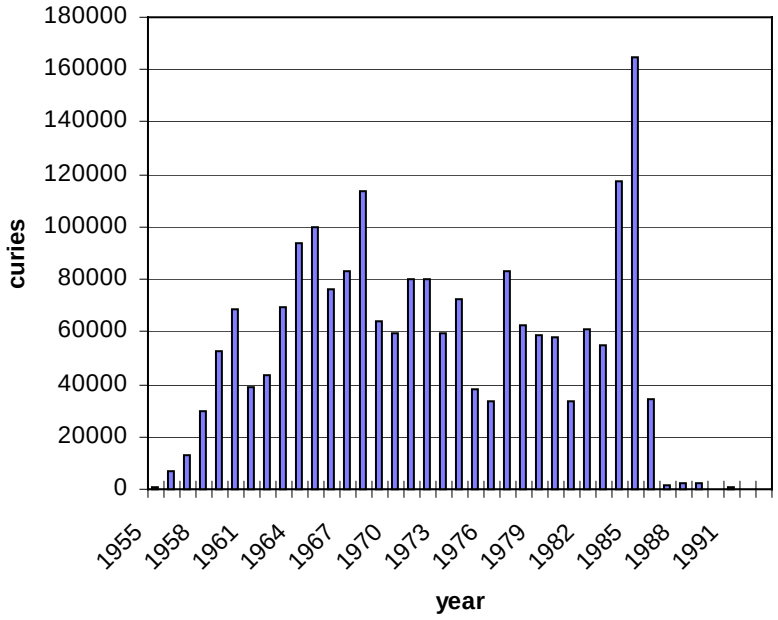

Figure 4.1-5. Annual atmospheric tritium releases from reactor 105-C (Ci). Link to tabulated data.

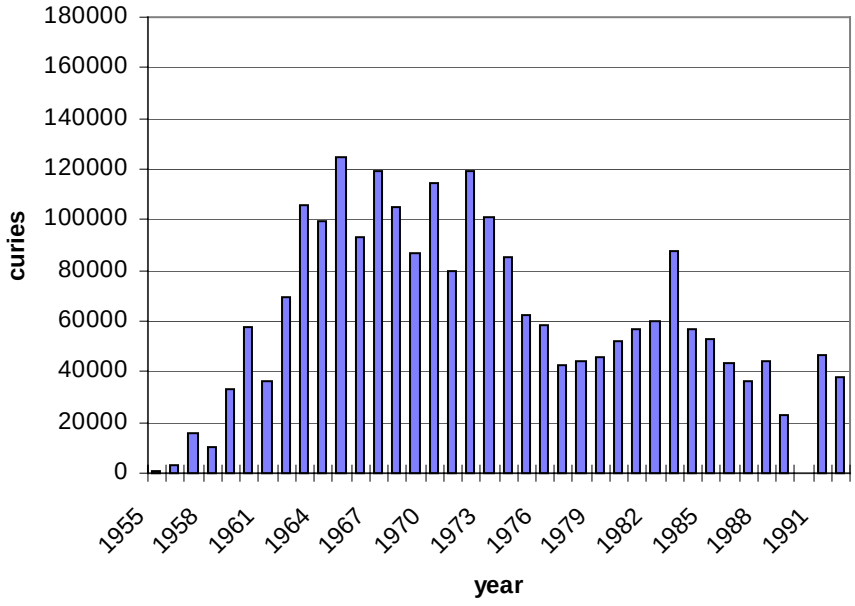

Figure 4.1-6. Annual atmospheric tritium releases from reactor 105-K (Ci). Link to tabulated data.

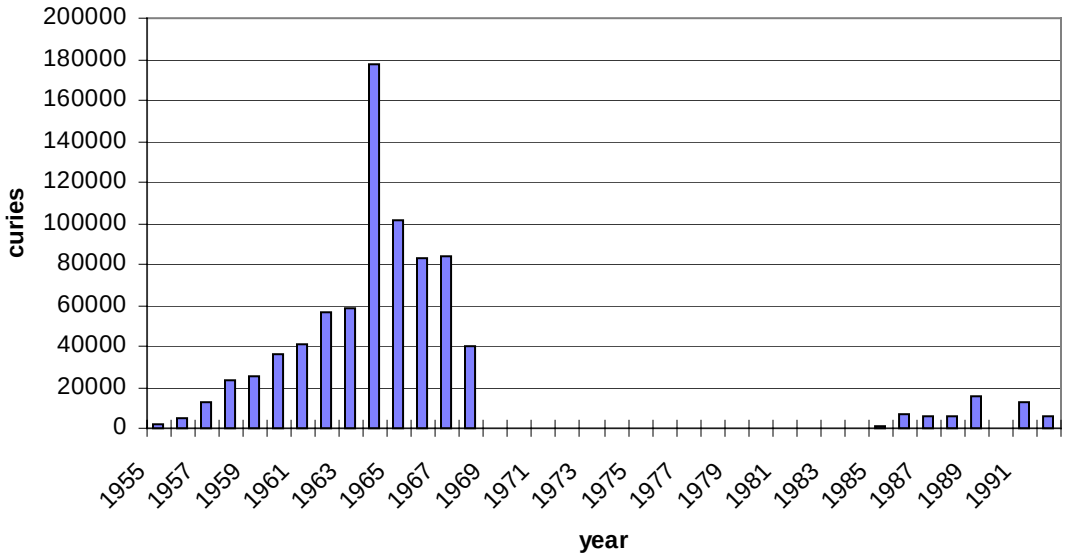

Figure 4.1-7. Annual atmospheric tritium releases from reactor 105-L. Link to tabulated data.

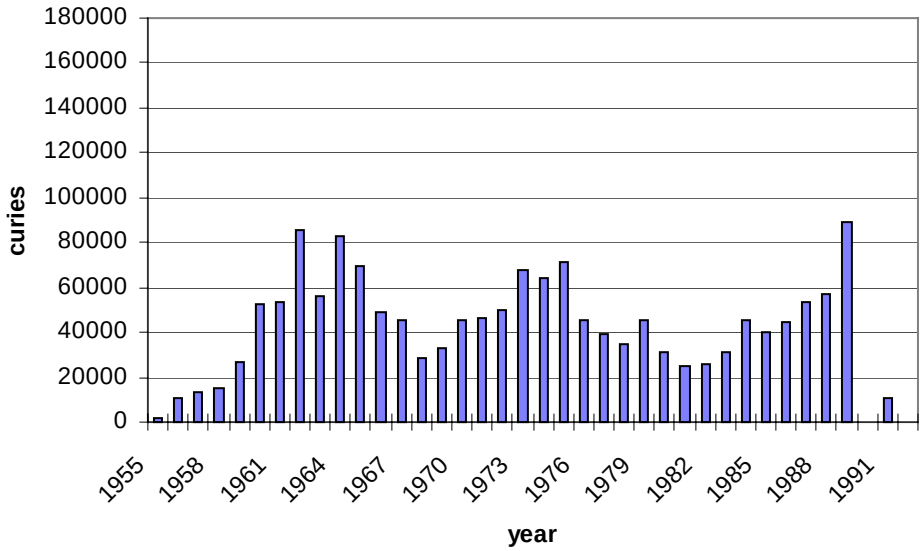

Figure 4.1-8. Annual atmospheric tritium releases from reactor 105-P (Ci). Link to tabulated data.

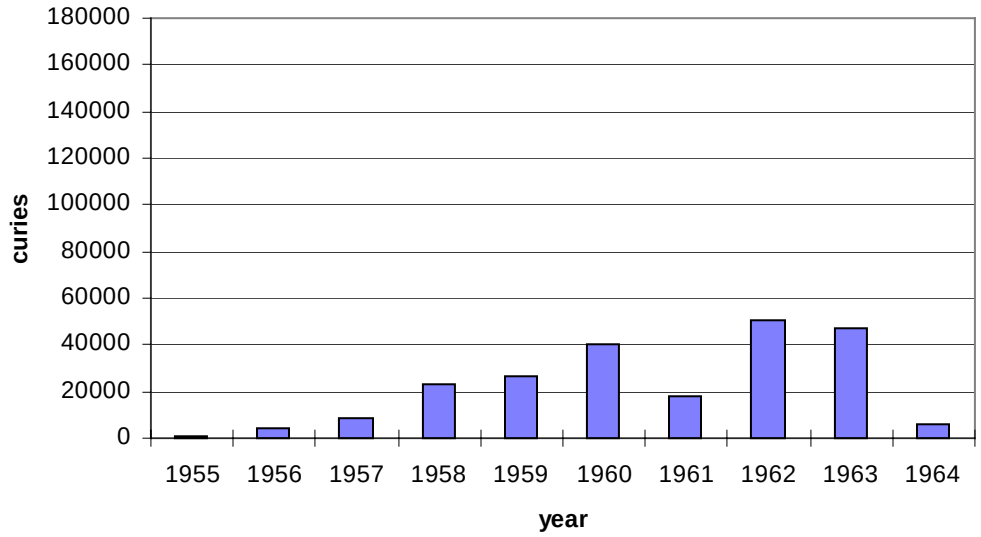

Figure 4.1-9. Annual atmospheric tritium releases from reactor 105-R (Ci). Link to tabulated data.

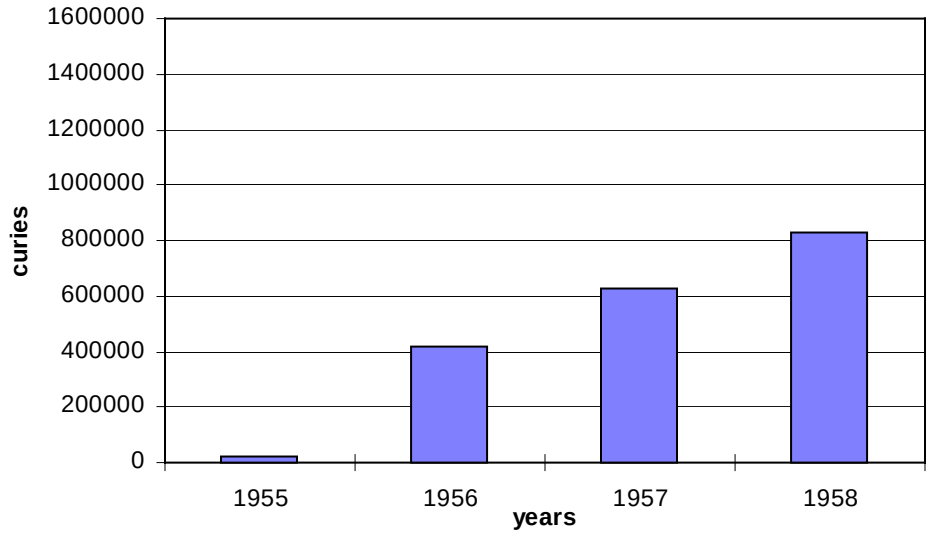

Figure 4.1-10. Total annual tritium releases (Ci) from Building 232-F. Link to tabulated data.

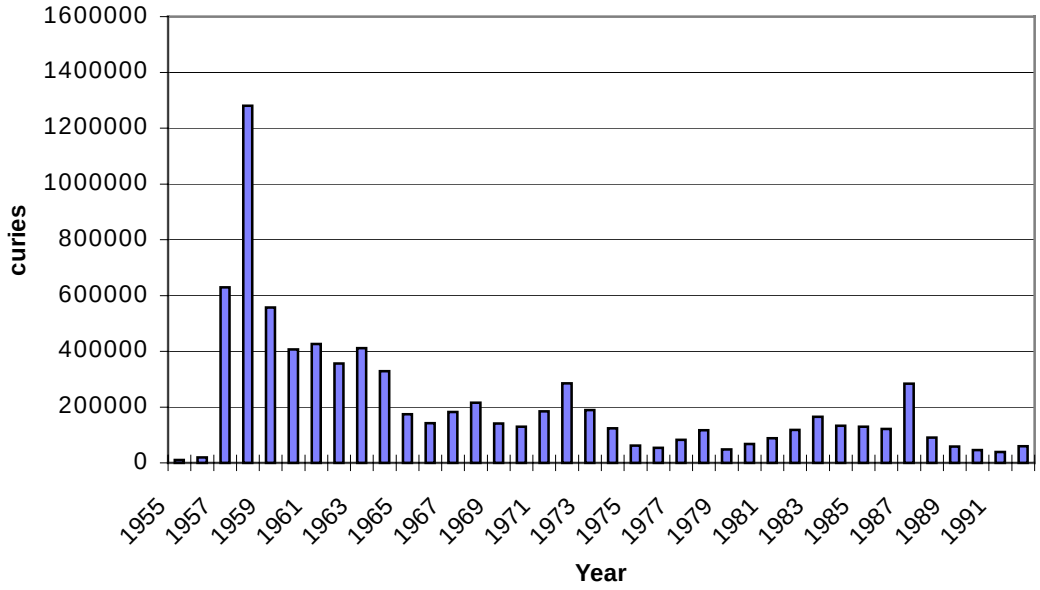

Figure 4.1-11. Total annual tritium releases (Ci) from Building 232-H. Link to tabulated data.

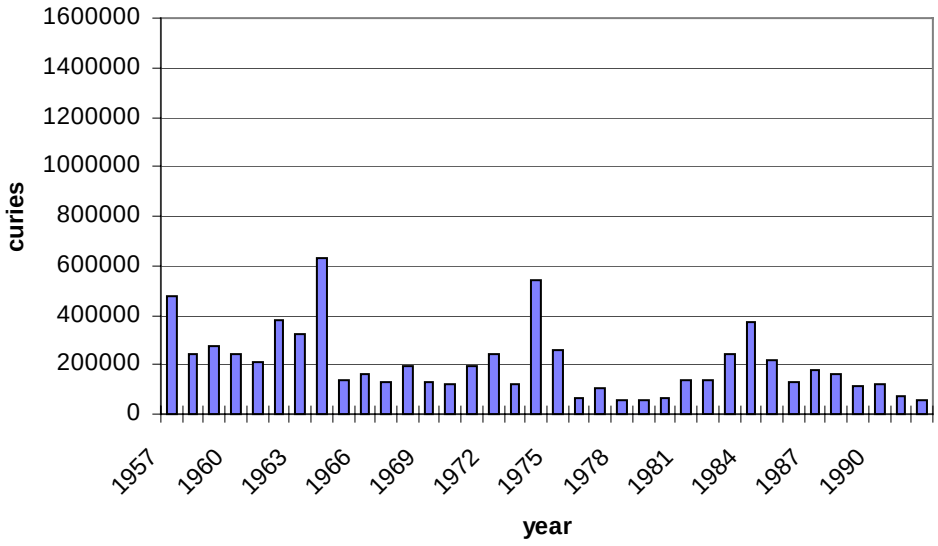

Figure 4.1-12. Total annual tritium releases (Ci) from Building 234-H. Link to tabulated data.

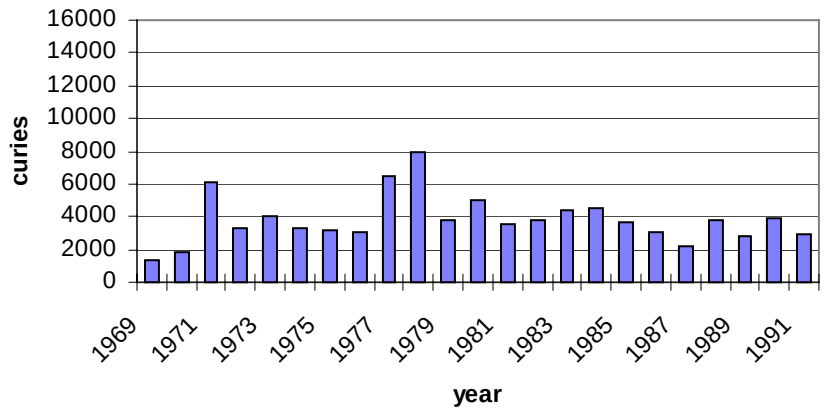

Figure 4.1-13. Total annual tritium releases (Ci) from Building 238-H. Link to tabulated data.

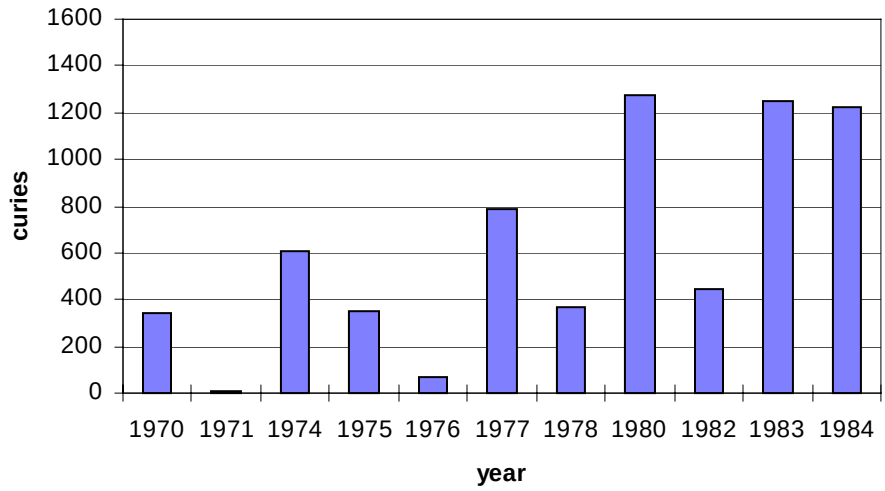

Figure 4.1-14. Available total annual tritium releases (Ci) from Building 244-H. Link to tabulated data.

We believe that the monthly data presented here include all the releases, both routine and inadvertent. However, some of the inadvertent releases were large and occurred over a short period of time, so it is likely that future dose reconstruction work will address these separately. To support this, we have included information we found on the details of inadvertent releases, which are discussed in the following section.

INADVERTENT RELEASES OR INCIDENTS

Introduction

Over the years at the SRS, inadvertent releases were defined as those releases that were not planned and resulted from an unexpected sequence of events or failures in a process. Many incidents occurred, resulting in inadvertent releases of tritium. These were documented in Incident Reports and an incidents report database. Incident Reports were found for releases as early as 1956 (Miller 1956c). All evidence from our record search leads to the conclusion that the inadvertent releases were included in the total monthly and yearly tritium values reported because no large release that were identified seem unaccounted for in the monthly data reported. Starting in the 1970s, any large releases with public health, worker safety, or economic implications, were assessed in special reports written by SRS staff.

We have researched and reported routine and inadvertent releases separately to (1) ensure that the releases (which are expected to be larger than usual) have been included in the release totals published by SRS, and (2) identify these events for possible special treatment during the future phases of the SRS Dose Reconstruction Project.

A report summarizing the results of Phase I of the SRS dose reconstruction project discussed unusual events and accidents that caused environmental releases considered significant to the dose reconstruction process (Meyer et al. 1995). In developing a list of the most important events or accidents, it became clear that the definition of important releases would greatly influence the number of occurrences that would be included as distinct analyses in the future dose reconstruction.

In summaries of the tritium releases from SRS, Murphy et al. (1991) used the term “inadvertent tritium releases” to the environment and designated recorded releases of over 700 Ci to the atmosphere as “significant” releases. This level was chosen because it is roughly 0.1% of the average annual tritium atmospheric releases. In this report, we used the 700-Ci level designated in Murphy et al. (1991) to define significant releases.

Because inadvertent releases were defined as unplanned, the relatively large releases that occurred during the early days of operation as a result of developing new facilities and procedures were not flagged as unusual events. They were included in the routine reporting of releases (Murphy et al. 1991; Du Pont 1955). During later times, much smaller releases that occurred as a result of some malfunction were considered and documented as inadvertent releases. We believe that the comparison of specific incident release with the published monthly release values shows that the tritium release has been included in routine reporting. For example, if you look at the release from B234-H over the year of 1974, you will see the monthly release ranged from 3580 Ci to 8247 Ci, excluding the month of May, during which a large inadvertent release occurred. On May 2, 1974, a release of 479,000 Ci occurred. The May 1974 release was reported to have been 481,954 Ci. This value allows for a “routine” monthly release of about 3000 Ci (which is a bit low but close to the range seen for the rest of the year) plus the inadvertent large release.

Another main source of data for inadvertent releases (as they were defined at the time) was the Tritium Area Facilities Fault Tree Data Storage and Retrieval System, or TAFFTDSRS, (TAFFTDSRS 1994) beginning with the year 1956. Inadvertent releases between 1959 and 1974 are indicated to have been a result of reactor operations and from reactor moderator losses (Murphy et al. 1991), but the few unusual events resulting in increased atmospheric releases of

tritium from the reactors were found in reports or logbooks, not the database. The releases from the Tritium Facilities, which are reported in detail, have been attributed to aging equipment (Murphy et al. 1991).

Search profiles were prepared for the four unclassified data banks during Phase I of the SRS dose reconstruction project (Meyer et al. 1995), and a separate search profile was prepared for the TAFFTDSRS (TAFFTDSRS 1994). The entries range from procedures violations and minor equipment malfunctions to incidents that released radioactivity or chemicals or resulted in injury to workers. The data banks are used primarily for probabilistic risk assessment, training, trend analysis, safety analysis reports, and management and administrative controls analysis. Most of the detail in the incidents listed in the database pertains to the workers, the SRS facilities, and the immediate onsite environment and equipment.

The search profile for the TAFFDSRS (TAFFTDSRS 1994) produced a printout listing 2994 incidents for just the five different curie levels (1 to 100 Ci; 100 to 1000 Ci; 1000 to 10,000 Ci; 10,000 to 100,000 Ci; and greater than 100,000 Ci). For each incident, the database lists the unique incident identification number, source of information describing the incident, date of occurrence, and description of incident. Another list of approximately 3000 incidents was printed out under the remainder of the tritium search. The actual incidents listed in these two printouts are approximately the same (e.g., a release under airborne also would be listed in one of the curie categories). Incidents of tritium releases over 700 Ci were used as the basis for documenting inadvertent releases for this report (link to tabulated data). The data from this printout were augmented by information found in period incident reports and limited information found in logbooks.

Documented Inadvertent Releases

Reactors

The investigation of unplanned and accidental releases of tritium from reactors has uncovered very little specific information. Most of the details that were found are for the Tritium Facilities, where larger amounts of tritium were at potential for release. A list of “Inadvertent Tritium Releases to the Environment from SRS Operations,” found among C. Zeigler’s personal files, notes three specific cases of atmospheric releases from reactors: (1) November to December 1961 (20,000 Ci HTO) from P-reactor stack, (2) March to June 1977 (83,000 Ci HTO) from C-reactor, and (3) early in 1978 (62,810 Ci HTO) from C-Reactor stack (Zeigler 1994). (The notes are varied and are not all dated. The year 1994 depicts the year the notes were reviewed.) However, we found no other specific mention of these releases as unplanned incidents or accidents on specific dates or times in any other report.

The P-Reactor release in 1961 (no date indicated) could have been included in the 6-month releases reported for 1961, which take a jump of 18,500 Ci from the first 6 months to the second 6 months. These biannual values were published and are also shown in the logbook data (Table 4.1-4). Therefore, in this case we cannot draw a clear conclusion regarding the release indicated in the logbook.

The two C-Reactor release values mentioned in Zeigler’s logbook are the same values as were reported for the yearly total atmospheric release. No further information is provided and no

mention of this is indicated in any other source, which would probably have occurred during the 1970s. In this case, it is not possible to interpret this information.

Table 4.1-4. P-Reactor Tritium Releases Noted in Logbook (Zeigler 1994)

	Releases (Ci)	
Year	January–June	July–December
1960	28100	24400
1961	17600	36090
1962	37300	48100

Tritium Facilities

For the Tritium Facilities (Buildings 232-H, 234-H, and 238-H), it became clear upon reviewing the results of the database search, logbooks, and incident reports that there were hundreds of incidents resulting in a wide range of releases. These releases had been individually recorded and included in the monthly release reports. For this report, we have used the level that was defined as significant by Murphy et al. (1991) and only report releases of over 700 Ci to the atmosphere. This is roughly 0.1% of the average annual release to the atmosphere.

With this criterion, we identified 289 events in the time period from July 1956 to June 1990. Figure 4.1-15 plots these incidents and limited details of these events can be found in the next section of this chapter.

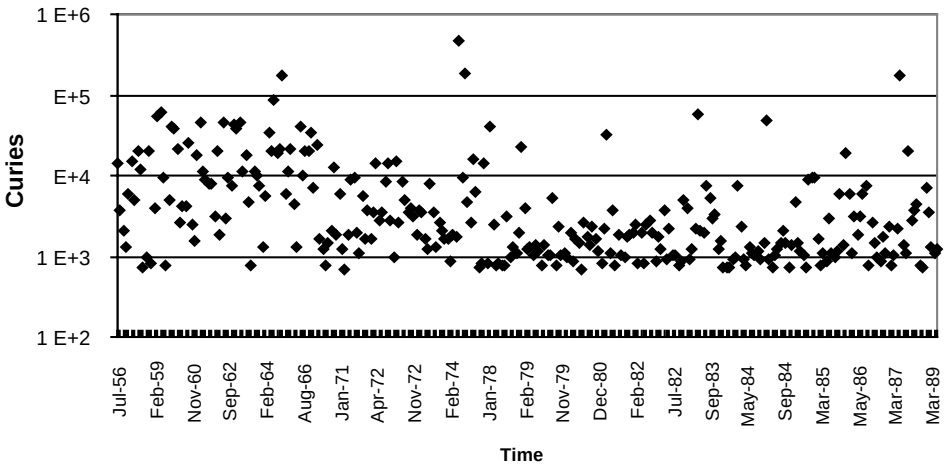

Figure 4.1-15. Summary of incidents of inadvertent or accidental releases of tritium to the atmosphere (over 700 Ci). The y-axis of this figure has values shown in scientific notation. A value such as 1 E+6 is equivalent to 1 x 10^6 or 1,000,000. Link to tabulated data.

To have a better idea of the importance of these releases, Table 4.1-5 groups the data by 5000-Ci increments (above 4999 Ci) and Figure 4.1-16 plots these releases.

Table 4.1-5. Inadvertent Tritium Releases Over 700 Ci from the Tritium Facilities from 1956 to 1990

Curies	Number of events	Curies	Number of events
700–4999	2	55,000–59,999	1
5000–9999	202	60,000– 64,999	1
10,000–14,999	34	65,000–69,999	1
15,000–19,999	12	70,000–74,999	0
20,000–24,999	11	75,000–79,999	0
25,000–29,999	6	80,000–84,999	0
30,000–34,999	1	85,000– 89,999	0
35,000–39,999	3	90,000–94,999	1
40,000–44,999	4	100,000–149,999	0
45,000–49,999	3	150,000–199,999	0
50,000–54,999	3	200,000	3
		479,000	1

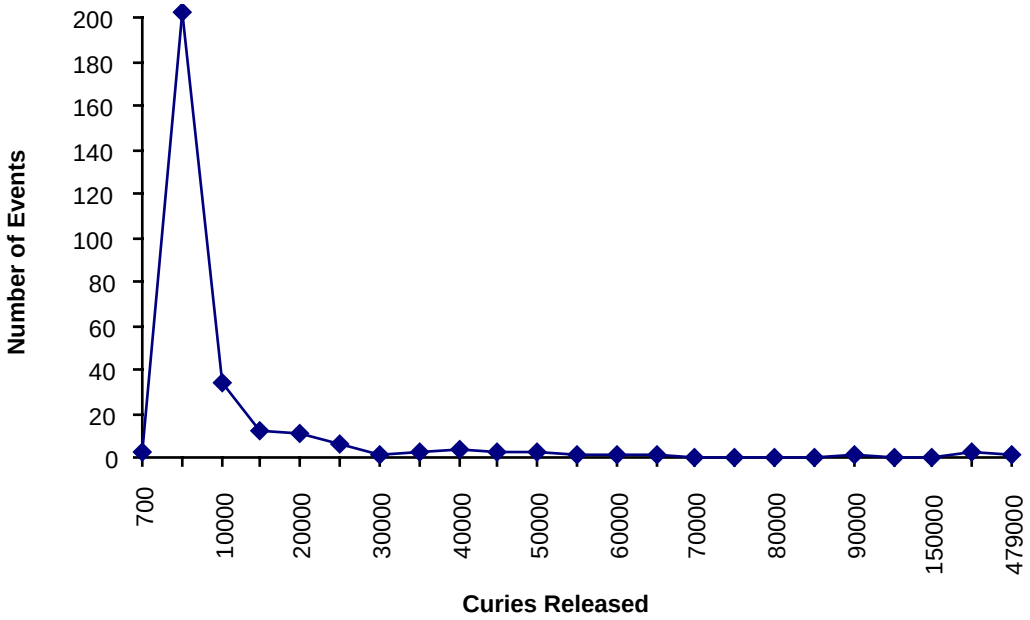

Figure 4.1-16. Activity releases from unplanned incidents and accidents (over 700 Ci releases) (based on data in Table 4.1-5). Link to tabulated data.

Special Incidents

Because of the magnitude of the release, composition of the release, or environmental conditions, more detailed studies were implemented by SRS to investigate the potential impact on

the environment and the public. In the 13 cases where more detailed information was available from SRS reports, the information is summarized below[a].

June–July 1964. Incidents resulting in around a total of 100,000 Ci loss to the atmosphere can be seen in Figure 4.1-15 during mid-1964. Although no detailed description of the incidents were found, the monthly reports describe operational incidents that were not considered to be unusual, such as routine purging of the stack system or during letdown of argon after loading room check operations (Du Pont 1964a, 1964b).

May 2, 1974. In Building 234-H, at 8:01 a.m., tritium monitors for the 200-ft exhaust stack and for the enclosure cabinet exhaust system to the process room alarmed. Over the next 4 minutes, over 99% of the 479,000 Ci of tritium gas (T_2) was released, as indicated by the stack monitor integrator (exhaust monitor) and consistent with the material balances within the measurement accuracy. Approximately 50 g of tritium (about 7 ft^3, STP), mixed with the building ventilation exhaust air, discharged from the stack at rate of 135,000 $ft^3\ min^{-1}$. The total volume released during the 4 minutes was 540,000 ft^3 of air. It exited the 8-ft diameter stack top at a velocity of 2700 ft min^{-1}. Averaged over the 4 minutes, the release was estimated to be a concentration of 13 ppm (approximately 0.25 Ci ft^{-3}) (Morris and Scaggs 1974; Haywood 1974a). Measurements offplant indicated that less than 1% of the tritium was in oxide form (Morris and Scaggs 1974). A Kanne monitor chart is presented in the reports (Haywood 1974a, 1974b; Marter 1974; Morris and Scaggs 1974). The two peaks of the release were estimated to be at 600,000 Ci min^{-1} and 170,000 Ci min^{-1} (the tops were over the top of the chart). A much smaller peak a bit later was estimated to be 9900 Ci min^{-1} (Haywood 1974b).

It appears that the release was estimated using a manual integration technique because the electronic integrator was out of service during the release period (Patterson 1975). Another account written in 1991 indicated that the release started at 07:55 a.m. (Murphy et al. 1991). A summary of the environmental effects of the release was summarized in Marter (1974).

December 31, 1975. At 10:00 p.m. on December 31, 1975, a process failure (vacuum gauge failed releasing about 20 g of tritium) in Building 234-H resulted in a release of 182,000 Ci of tritium via the 200-ft stack. The automatic integrator was out of service because of a malfunction, so the health physics shift supervisor and inspector manually integrated the stack Kanne recorder chart. The graphic integration method they used involves making a calculation for each one-third of a minute during the time the release rate was greater than 100 Ci m^{-1} (Cofer 1976; Webb 1975; Jacobsen 1976).

The extended range stack monitor recorder peaked at a release rate of about 290,000 Ci m^{-1} and about 90 seconds later indicated a release rate of about 600 Ci m^{-1}. About 90% of the release occurred during a 1.5-minute period. The chart is attached to the report (Cofer 1976).

"Tritium absorber samples" from the 234-H stack indicate that approximately 99.4% of the release was in elemental form and 0.6% in oxide form (Webb 1975). Tritium FORMS samplers, which determine the amounts of elemental tritium gas and tritium oxide, had been installed in August 1974 (Jacobsen 1976).

[a] We received information during the review phase of this project indicating that we should have included a tritium release from K Reactor in December of 1991 in this section. We found no detailed information about such a release, and believe that it was likely included in the monthly source term. We do, however, want to draw attention to the possibility that such a release did exist and needs to be studied in more detail in a later phase of the dose reconstruction project.

March 27, 1981. On March 27, 1981, 3.42 g (33,000 Ci) of tritium in the form of water vapor was released via the 234-H stack from 09:45 a.m. to 12:00 noon. Maintenance personnel were attempting to change a plugged inline filter (Anonymous 1981). The stack monitor indicated a loss of 308 Ci min^{-1} (0.03 g min^{-1}) as work to remove an inlet filter was being done (no water was seen). As the work continued, the loss dropped to 12 Ci min^{-1} (0.001 g m^{-1}). However, during the installation of the new filter, water dropped into the hood and the stack monitor activity increased to (2200 Ci min^{-1} [0.23 g min^{-1}]) for about 2 minutes (Green 1981). The tritium FORMS samplers indicated that the release was over 99% tritium oxide (Anonymous 1981). Tritiated water vapor was 99.7% of the release (Murphy et al. 1991).

July 16, 1983. On July 16, 1983, about 56,392 Ci (5.8 g) (Epting 1983) of elemental tritium was released from Building 234-H at 11:16 p.m. (Du Pont 1983). The release took place over a period of about 3 minutes, and only 1% of the release was in oxide form (Murphy et al. 1991; Garrett et al. 1983). The release was monitored by the Kanne chambers.

March 23, 1984. On March 23, 1984, 7673 Ci of tritium was released from Building 234H during a product transfer (Du Pont 1984a). A process line leak occurred in a process hood that began at 5:40 a.m. and continued at decreasing rate until about 9:00 a.m. From 5:40 a.m. to 8:40 a.m., 7400 Ci was released and an additional 100 Ci was released from 8:00 a.m. to 9:00 a.m. as monitored by the Kanne chambers. The tritium FORMS monitor installed on the sampling line indicated that the average composition of the release was 70% HTO/30% HT (Evans et al. 1985).

September 2, 1984. A total of 46,690 Ci of tritium was released from 234-H starting at 10:00 p.m. on September 2, 1984, until 10:00 am on September 3, 1984. The release occurred during preparation for maintenance work on a process pump. The activity was in the form of spilled HTO. Over the next several days, an additional 11,160 Ci was released for a total of 57,850 Ci by the time the cleanup efforts were completed on September 7, 1984. The tritium FORMS monitors confirmed that all the release was in oxide form (Du Pont 1984b; Anonymous 1984). Another report stated that 99% of the release was in oxide form (Murphy et al. 1991).

Another account (Hoel et al. 1990) indicated that 43,8000 Ci was released between 10:00 p.m. September 2 to 3:00 a.m. September 3. The additional 14,000 Ci was released during cleanup operations between September 3 and late in the day on September 7.

January 31, 1985. A total of 9285 Ci of tritium was released from the Tritium Separations Area from 2:00 p.m. until 5:00 p.m. The majority of the tritium (7400 Ci) was released during the first 15 minutes. About 54% of the release was in oxide form and the remaining 46% in hydrogen form (Murphy et al. 1991).

March 27, 1985. An estimated 19,422 Ci of tritium oxide was released from H-Area on March 27, 1985. The release began at 1:53 p.m. and averaged about 90 Ci min^{-1} until 5:30 p.m. (Addis et al. 1985). The rate of release was 76 Ci min^{-1} at 2:14 p.m. and 96 Ci min^{-1} at 2:31 p.m. The tritium FORMS analysis showed that the release was 99.99% oxide.

May 29, 1986. Approximately 5900 Ci of tritium was released from Building 232-H during repairs of process equipment on May 29, 1986. Approximately 95% of the tritium was in oxide form (Lawrimore 1986).

July 31, 1987. A release between 8:17 a.m. and 8:55 a.m. occurred on July 31, 1987, from the separations area stack. The total releases estimate (172,000 Ci) was based on the tritium FORMS monitor, where 2.7% was in oxide form and the remaining 97.8% was in elemental form (Murphy et al. 1991).

March 1, 1988. At 6:18 a.m., a total of 19,980 Ci was released from Building 234-H. The chart recorder peaked at approximately 90,000 Ci min^{-1}. By 6:19 a.m., the stack monitors had decreased to approximately 300 Ci min^{-1}; by 6:25 a.m., the rate was down to about 4 Ci min^{-1}. The tritium FORMS monitor showed that approximately 86% of the release (17,120 Ci) was in elemental form, and the remaining (2860 Ci) was in tritium oxide form (Lott and Mason 1988).

June 7, 1988. About 3650 Ci of 96% tritiated hydrogen gas was released on June 7, 1997, from the separations area tritium facility (Murphy et al. 1991).

October 6, 1988. Approximately 7000 Ci of tritium was released from a separations area tritium facility stack. About 11% was in the form of tritiated water vapor (Murphy et al. 1991).

December 7, 1988. Approximately 3500 Ci of tritium was released from a separations area tritium facility stack. About 99.5% was in the form of tritiated water vapor (Murphy et al. 1991).

The relative importance of the incident releases from the Tritium Facilities is shown in Figure 4.1-17. The larger releases that occurred during the early days of operation were the result of developing new facilities and procedures. They were not documented as unusual events. In later years, these incidents were considered unusual incidents or at least unplanned. Figure 4.1-17 illustrates the percentage of the total annual release attributed to accidental or unusual releases.

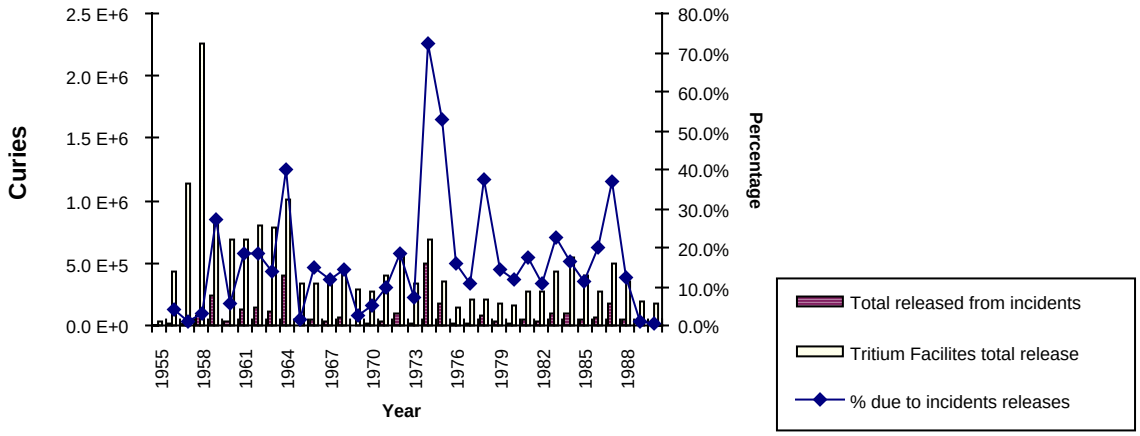

Figure 4.1-17. Annual releases (Ci) attributed to unusual incidents as a percentage of the total release from the Tritium Facilities. Link to tabulated data.

As with the source term releases, several documents that discussed inadvertent releases were identified but not explicitly cited in the text of this chapter. These document citations are included in the "Reference – Inadvertent Releases" section.

SUMMARY

We conclude that the releases of tritium from the SRS are generally well documented in published internal reports. The main sources of information were SRS weekly and monthly reports, published since near-commencement of site operations. We used daily or shift data to check the monthly published data where feasible. In general, agreement was found to be quite good. We have found no indication that there were serious or consistent errors made in SRS tritium release monitoring.

It is not possible to provide a brief summary of the key release data in this summary section. The number of figures provided to summarize the key data is too large to incorporate here. The reader is instead referred to the groups of figures that summarize releases from specific SRS sources. The total annual releases, both routine and inadvertent, from each reactor are plotted with time in Figures 4.1-5, 4.1-6, 4.1-7, 4.1-8, and 4.1-9. Figures 4.1-10, 4.1-11, 4.1-12, 4.1-13, and 4.1-14 provide plots of the annual releases from the Tritium Facilities. Two spreadsheets contain the detailed information, including uncertainty estimates: Annual Reactor Releases, and Annual Tritium Facility Releases. While we believe that these data incorporate the records of inadvertent tritium releases over the years, we have included in this chapter a detailed discussion of documented inadvertent releases to provide the reader with a more complete discussion of this important topic.

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