

APPENDIX E

SAMPLING PROCEDURES USED AT SRS FOR MONITORING ATMOSPHERIC TRITIUM RELEASES

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ABSTRACT

This appendix describes the monitoring instrumentation, sampling procedures, and data analyses used to measure or estimate the reported releases of [tritium](#). The [accuracy](#) of reconstructing the atmospheric tritium [source term](#) is largely dependent on the type of monitors used to measure the releases. Atmospheric tritium releases have been monitored using dehumidifiers, silica gel monitors, Kanne ionization chambers, the stack tritium monitor (STM), the Berthold tritium monitor (BTM), the stack monitor integrator (SMI), and the FORMS in the [reactor](#) and tritium facilities on the Savannah River Site (SRS). This appendix describes the monitoring equipment and how it was used. We have performed an extensive review of documentation, but it has been difficult to determine the exact times that each type of monitor operated at each of the facilities.

Radiological Assessments Corporation (RAC) researchers identified no clear deficiency in the methodologies used at SRS. Therefore, we initially believed that no correction factors should be applied to the SRS data in producing the final source term estimates presented in [Chapter 4.1](#). To verify that this was an acceptable conclusion, we used an [uncertainty](#) assessment to evaluate the effectiveness of the monitors. This appendix presents the estimated uncertainty of the tritium measurements by type of monitor.

Using the results of the uncertainty assessment, we concluded that the “best estimate” values should be used as the atmospheric tritium source term without correction, and the range of uncertainty should be applied to release data by facility and year. If more information is found about the monitors or when they were used in each facility, we can apply these results more specifically.

INTRODUCTION

Our annual or monthly release estimates of tritium to the atmosphere are based on Site documents, memos, and logbooks. To develop the tritium facilities and the reactor areas atmospheric source term and its associated uncertainty, it was necessary to determine the methods by which the reported releases were measured or estimated. This appendix describes the monitoring instrumentation, sampling procedures, and data analyses used to measure or estimate the reported releases of tritium.

The equipment used at the SRS to monitor tritium, in most cases, was developed at the Site using existing components. Usually, off-the-shelf equipment that could fulfill the monitoring requirements was not available. The work being done by the scientists and engineers at the SRS was at the forefront of tritium technology, so new approaches and instrumentation were developed as the need arose. Letters that were found from other facilities requesting advice from the SRS staff confirm this fact. It appears from old documentation that the adequacy of the monitoring capability was evaluated regularly. The development of new technology can be

tracked in the memos that propose, justify, and evaluate the new monitors as they were implemented onsite.

It has been difficult to determine the exact times that certain equipment was installed and operated at each of the facilities. In reviewing the documentation, it is possible to find dates on equipment operation procedures or reports of plans or prototype tests. However, it has not been possible to reconstruct an exact chronological history of which monitoring equipment was used in each facility to measure and report the atmospheric releases. Unless more specific information is found for each SRS facility, [Table E-1](#) provides as a general guide to tritium monitoring systems that were the primary source of the reported tritium releases since the 1950s.

Table E-1. Guide for Use of Tritium Monitors at SRS

Monitor	Facility	Time period in use
Dehumidifier	Reactors	1954–shutdown
Silica gel	Reactors and disassembly areas	during 1954–1958 or until shutdown in some cases
Kanne ionization chambers	Tritium facilities	1954–present
Stack tritium monitor (STM)	Reactors	1970–1988
Berthold tritium monitor (BTM)	Reactors	1988–present
Stack monitor integrator (SMI)	Tritium facilities	1974–present
FORMS	Tritium facilities and reactors	1985–present

We made assumptions about which monitors were used in specific facilities based on the reactors' years of operations. For example, BTMs only came into use after the R-Reactor was shut down, so we concluded that BTM monitors were never used in the R-Reactor facility.

The documentation we found that describes testing or operation of prototypes showed that monitoring equipment was tested in parallel to provide some redundancy and check results. Therefore, we could not identify exact dates, by month, when specific monitoring equipment was used. In the cases where data sheets reported the results of multiple monitoring devices, it is possible to check the measurement results of one system against the reported monthly value and determine which monitor results were used. However, we could not determine the exact date that the primary monitoring device replaced an older system, or when it was used during unplanned malfunctions of equipment.

The overall uncertainty includes the representativeness of the sample, the frequency of sampling, efficiency of sampling, the effects of environmental conditions, the accuracy of the measurements, and errors in reporting results. This appendix describes each monitoring system or component of the system. The latter part of this appendix addresses the uncertainty associated with the results from each method of monitoring.

In many cases, existing systems were coupled to create “new” monitoring systems. For example, silica gels or dehumidifiers were combined with the Kanne chambers to create the STM. The SMI is a Kanne chamber coupled with an electronic method of integrating the continuous monitoring data. The FORMS monitor installed in the Tritium Facilities appears to be similar to

the STM because the Kanne chambers that are part of the FORMS monitor measure the stack releases with and without water vapor being removed.

EXHAUST AIRFLOW AND AIR STREAM SAMPLING

Estimates of exhaust airflow rates and additional errors introduced because of sampling could possibly be a major influence on the final estimated stack releases. Detailed descriptions of methods of exhaust airflow measurements taken at the SRS facility stacks have not been found; however, it is clear that there was some capability to measure these flows. A historical copy of Health Physics Procedures ([Du Pont](#) 1965) presented average exhaust air stream values to be used for various stacks. These changed slightly through time, indicating that some measurements were actually made. Measurements were definitely made in later years.

In health physics work area sampling documents, we found a description of a system designed to individually sample widely separated work areas or simultaneously average the air [activity](#) from any work areas. By using electrically or manually operated valves, samples could be selected from one or more points at a work site or the size of the area being monitoring by one system could be changed. The piping was sized to correct for the pressure drop in long piping runs or restricting orifices ([Morris](#) 1967). Although the sampling documents described how these types of measurements were made, in general, we found no specific evidence on how stack measurements were actually made.

There were, however, airflow diagrams of the reactors and Tritium Facilities Buildings 232-H, 234-H, and 244-H were found in a Radioactive Air Emissions Monitoring document ([Westinghouse](#) 1991e). [Chapter 4.1](#) presents these diagrams, which also include the exhaust monitoring sampling locations. For different areas, it seems that airflow in the stack was either measured directly or estimated based on building and exhaust system designs; the method of determination is not clear. For example, [Table E-2](#) presents the ventilation airflow rates for reactor stacks provided in a written procedure for silica gel monitoring written in the 1950s.

Table E-2. Ventilation Airflow Rates for Reactor Stacks^a

Location	Pounds of air per day
Purification exhaust	4,220,000
148-ft stack	35,300,000
Process room near exhaust	8,780,000
Process room for exhaust	7,660,000
-40 pump rooms	12,800,000

^a Source: [Du Pont](#) (1954).

We found some discussion indicating that in 1989, the airflow in the stack was determined by measuring the pressure drop across the carbon filters. This was measured in inches of water and was converted to airflow using a vendor [quality assurance](#) test of the filters. The error in the stack flow estimates was estimated to be about 10% based on experience and review of stack flows. The pressure drops are only accurate to the nearest 0.05 in., which introduces a potential additional error of $\pm 5\%$. Therefore, the conservative estimate of airflow measurement accuracy is $\pm 15\%$ ([Smith](#) 1989). However, exactly how knowledge of this accuracy was used in day-to-day operations is not clear.

A data sheet for August 17, 1990, shows that data were collected daily on reactor ventilation systems. Airflow was checked five times a day and showed no variation for the main exhaust stack ([Westinghouse 1990a](#)), but methods used were not outlined.

In *SRS Alternative Methods Information* ([Westinghouse 1991b](#)), rotameters were the principal method of regulating and measuring sample flow rates for the Kanne chambers in the reactors. It was indicated that the rotameters were calibrated annually and at any other time that maintenance or changes occurred. Unfortunately, no information was found about how often rotameters were calibrated or the range of variability.

In 1991, a report indicated that for K-Reactors and L-Reactor ventilation systems, five hot wire anemometers were used to monitor the total [effluent](#) air discharge, and the flow was indicated in the central control room ([Suttinger and Burgo 1991](#)). Data sheets found for reactors in the 1970s and late 1980s list airflow values that vary, indicating measured values were being used. From this information, we concluded that when possible (and in later years) actual measurements were used daily.

To sample the air leaving the stack, pumps were installed to withdraw effluent and route it to the samplers. In the earlier years, the samples were not necessarily continuous, which affected the overall accuracy of the release estimates. We describe the sampling regimes in the following sections for each type of monitor used. For tritium gas and vapor, we assumed that sampling air directly from the outflow stack did not introduce measurement errors.

TRITIUM LOSSES

Moderator Loss

The earliest monitoring operations were mostly concerned with measuring loss of [moderator](#). To keep the reactors running, there needed to be sufficient moderator in the system to ensure the core of the reactor was cooled. The moderator is comprised of [heavy water](#) and was produced at SRS or elsewhere (Dana Plant). Because of the bombardment of the moderator by fission [neutrons](#), tritium was created in the moderator. Because tritium has a relatively long [half-life](#), its [concentration](#) in the moderator built up over years of continuous reactor operation. Moderator could be lost to the atmosphere or by direct spills because of evaporation from leaks in the system, spills during maintenance, or planned liquid discharges. These losses were economically important because moderator was costly to replace and [exposures](#) to the environmentally mobile tritium oxide were a potential cause of human health and environmental impacts. For these reasons, the loss of moderator was routinely monitored. By measuring moderator losses and the concentration of tritium, total tritium losses were measured. Daily releases to the stack would vary depending on the type of operations and maintenance carried out in the reactor.

Initially, “releases of tritium to the atmosphere from the 105 (reactor) building are computed from the per cent of D₂O contained in water samples condensed from the stack stream and from moderator losses as determined by inventory” ([Caldwell 1958](#)). The method of tracking the moderator inventory reflected all phases of loss; therefore, stack discharges could not be differentiated from losses through sump collection, losses to the disassembly basin water during reactor discharge, or other [inventory](#) unaccountability. We did not find an indication of the exact time period that this method was used.

[Chapter 5](#) of this report, which deals with aquatic releases, addresses moderator losses as direct liquid releases. [Caldwell](#) (1958) indicates that individual inventory loss measurements were considered accurate to within a factor of 2.

It appears that the first direct method used to determine moderator loss was measuring the inventory of moderator in the tank. When the volume was measured visually on a sight glass attached to the tank, it was possible to discriminate by 1/8 in. or ± 149 lb in the reactor tank. The amount that evaporated was dependent on the bulk moderator temperatures. It was reported that the effect of a fluctuation of the moderator temperature of $\pm 1^\circ\text{C}$ would lead to a change of ± 300 lb in apparent inventory ([Longtin et al.](#) 1973; [Jacober et al.](#) 1973). The reports states, without more explanation, that the inventory [precision](#) was about ± 550 lb. Assuming a density of 1.105 g cm^{-3} for heavy water moderator ([Shleien](#) 1992), 1 lb of moderator is approximately 410 mL.

The moderator samples were analyzed for tritium content using a liquid scintillation counter. Using the estimated volume of moderator lost (pounds or liters) and the concentration of the tritium in the moderator (curies per pound or curies per liter), the total loss of tritium (curies) could be estimated. The efficiency of counting (measuring) the concentration of tritium in the moderator sample was quoted as 57%, and it was calibrated over a range of 5×10^{-5} to $0.4\ \mu\text{Ci mL}^{-1}$. The cited variation at $0.038\ \mu\text{Ci mL}^{-1}$ was determined to be 0.8% relative at the 95% confidence level ([Longtin et al.](#) 1973; [Jacober et al.](#) 1973). This information is included in this report for possible use in understanding the uncertainty associated with the early values reported for atmospheric tritium releases.

Limited data found for the early years showed that in 1955, the average total loss of moderator per reactor month was 1453 lb. If the liquid discharge is not taken into account, the loss was 740 lb. The 1956 losses were estimated at 1197 lb mo^{-1} , with no discharge ([Rubin](#) 1958). The average concentration of tritium in the moderator was about $1\text{--}3\text{ Ci L}^{-1}$, which was relatively low because the reactors had just begun operation (1955–1957). By the early 1980s, the concentration was about 9 Ci L^{-1} . [Kiger](#) (1955) indicates that the losses from R-Reactor ranged from 3 to 80 lb d^{-1} in January and February 1955.

Because of the limitations of this method, other methods of determining moderator loss were developed using silica gel, cold traps, dehumidifiers, and the STM. The monitors, described below, were used to determine the tritium losses as well as total moderator loss.

Silica Gel

Silica gel was used, since start up, to determine the amount of moderator that was lost from the reactor stacks ([Miller](#) 1956). The description of the procedure for measuring moderator loss to the stack was given in [Du Pont](#) (1954), dated July 23, 1954, and is presented below.

Moderator loss was to be measured three times a week during the day shift, with other samples taken as required if high moderator loss was indicated. Moisture from the exhaust stack was absorbed on the silica gel desiccant during a sampling period. The gel was desorbed, the water collected, and tritium content determined (counted). Environmental releases of tritium were monitored at the +148-ft stack sampling point. The silica gel sampling line was left in place for 3 hours to collect the required 25 g of water. On dry winter days, more time was needed because of the relatively low humidity in the stack exhaust. This confirms that the level of humidity in the

outside environment did have an impact on the humidity levels in the stack at the sampling locations.

The temperature and humidity that was needed for this calculation was read three times during the sampling period using a hydrothermograph in the air supply duct at the +33-ft level. The average of these values was used. The health physics laboratory extracted water from the silica gel and submitted the samples to the 100-R laboratory of D₂O (moderator) and P-10 analysis. Although P-10 was not defined in [Du Pont](#) (1954), RAC believes that “P-10” in this document refers to tritium, based on the procedure outlined in this and other documents from the 1950s ([Anonymous](#) 1950). The procedure called for the laboratory to determine the difference between the sample and natural abundance of D₂O ([Du Pont](#) 1954). The airflow rate values presented in Table E-2 were used to calculate tritium released depending on sampling location.

As part of the procedure given in [Du Pont](#) (1954), a calculation sheet ([Table E-3](#)) was provided to calculate the loss of moderator to the stack. Using the temperature and humidity measurements (three per sampling period), the absolute humidity was determined using a chart provided with the procedure. If the chart was in grains water per pound air, the value was to be divided by 7000 to convert to pounds of water to pounds of air. If the chart was in units of moles water per mole of dry air, then the value was to be multiplied by 0.62 to obtain pounds of water per pounds of air ([Du Pont](#) 1954). The sample analysis (in mole percent) minus the natural abundance of D₂O (0.0147) was multiplied by 1.11 to provide weight % D₂O in the exhaust air. This value times the absolute humidity (pounds H₂O per pound air) by ventilation airflow (pounds air per day) divided by 100 gave the moderator loss per day (pounds per day) ([Du Pont](#) 1954). [Table E-3](#) summarizes these steps.

Table E-3. Area Procedure P–Moderator Loss to Stack Data and Calculations^a

Column	Data
1	Date and time sampling of air sample point
2	Temperature (°F)
3	Relative humidity (%)
4	Absolute humidity (lb water lb ⁻¹ air) A ^b
5	Ventilation airflow (lb air d ⁻¹) B ^b
6	Sample analysis—lab result (mol %) C ^b
7	Sample analysis—wt % above natural 1.11 (C–0.0147) = D ^b
8	Moderator loss (lb d ⁻¹) E = A × B × D/100 ^b

^a Source: [Du Pont](#) (1954).

^b Letters correspond to columns on data sheets.

The amount of moisture a silica gel monitor could absorb was limited, and the monitors could become saturated. When the monitors were saturated, moisture (and therefore tritium) would be lost if the sample was not changed when needed.

[Kiger](#) (1955), in presenting the use of a better method (the dehumidifier), states that the silica gel monitor has been “very erratic and non-reproducible with little correlation to system losses as determined by process water inventories.” The dehumidifier set-up did not have the limitation of becoming saturated. [Kiger](#) (1955) presents 6 days of data from R-Reactor using the dehumidifier method and P-Reactor using the silica gel method. Although the data presented show the silica gel to have produced a much wider range of daily data, they cannot be used to

make a quantitative judgment of the accuracy of the dehumidifier method because of cross-contamination of samples results from using the sample bottles to sample high purity moderator. It is not clear whether the reported tritium losses from the reactors were derived from the dehumidifier or the inventory method.

Dehumidifier

In 1955, dehumidifiers were first used in the R-Reactor to measure tritiated water vapor (HTO) releases from the reactor facilities (Kiger 1955). No specific date of the transfer from the silica gel method to stack dehumidifiers was found, but it appears that they were used after the beginning of 1955. Reports indicate that dehumidifier use continued until the shutdown of the reactors, although other systems were installed over time. Dehumidifiers appear to have been the primary monitors for tritium releases (Murphy et al. 1991). Air effluent monitoring by the dehumidifier method also was used to monitor areas other than the reactors, such as daily stack dehumidifier sampling of the 772-D, 420-D, and 421-2D stacks in the 400-D area, (Westinghouse 1991a).

The methodology for the dehumidifier was essentially the same as for the silica gel sampler, except that the moisture (and, therefore, HTO) was condensed out of the air on a set of cold coils rather than being absorbed by desiccant. The exhaust air was passed over the cold coils and water in the exhaust stream was condensed and frozen. The collection was interrupted daily (usually at night) to thaw for about 30 minutes to collect the 60 mL of sample needed for tritium analysis by liquid scintillation counting (Westinghouse 1979). Other documents (Anonymous 1950) indicate that another method collected the condensate immediately as it dripped into a collection pan. The unit was then dried by infrared heat lamps.

In 1958, Caldwell (1958) indicated that the dehumidifier was run three times a week for 24 hours, but it was not operated during reactor outages. Therefore, it was reliable for computing releases during normal operations of the reactor (Caldwell 1958) but not necessarily during outages. Westinghouse (1982) indicates the dehumidifiers were sampled daily in 1982.

The amount of tritium released from the stack was calculated based on the relative humidity of the air in the exhaust stream, the exhaust flow rate, and the concentration of tritium in the water sample collected from the exhaust (Longtin et al. 1973; Jacober et al. 1973) (Equation E-1). Tritium concentrations in air were measurable over the range of 10^{-5} to $5 \mu\text{Ci cm}^{-3}$ (Albenesius and Meyer 1962). The assumption was made that no significant incidents occurred while the dehumidifier was not running.

Health Physics Standard Operating Procedure (DPSOP) 193-HPP 241 (3/16/65 and 7/14/65) (Du Pont 1965) describes the method used to calculate the tritium activity loss in stacked air from the 100 Areas using the dehumidifier samples. The stack losses were determined using Equation (E-1):

$$H_3 \text{ (Ci)} = (H) (A) \text{ (days/month)} (K) (D) \quad (\text{E-1})$$

where

- H_3 = tritium discharged from stack (Ci)
- H = average absolute humidity of stack air (grains lb^{-1})
- A = tritium in dehumidifier sample (average daily) ($\mu\text{Ci HTO L}^{-1}$)

- K = conversion factor; obtained from a calibration curve of K factor related to the air flow in [cfm](#) (converts μCi to Ci ; cfm [from the x-axis of the curve] to cfd [unit retained in conversion factor]; and grains lb^{-1} to l ft^{-3}) ([Du Pont](#) 1965)
- D = air density correction factor (this factor was not introduced until the 3/16/65 version of the DPSOP): below 58.5°F (14.5°C) = 1.035; $58.5\text{--}77^\circ\text{F}$ (14.5 to 25°C) = 1.000; above 77°F (25°C) = 0.967.

Stack airflow for normal operating conditions in the reactors was indicated to be 110,000 $\text{cfm} \pm 20\%$ and for shutdown conditions usually 350,000 $\text{cfm} \pm 20\%$ ([Du Pont](#) 1965). It appears that the stack flow was not directly measured because the same exhaust air flow chart was used for many years.

In later published procedures, these same methods seem to have been adapted ([Westinghouse](#) 1979). However, the average airflow for the exhaust stream appears to be determined based on measurements at the time the air sample was obtained (this was usually the average flow for the day). The grams of moisture per unit of dry air was determined using a computer program.

In [Westinghouse](#) (1979), we see the dual use of the dehumidifier results to determine both moderator loss (pounds per day) and tritium released up the stack. The moderator loss was determined using the tritium data for the vapor in the air sample and in the moderator using Equation (E-2). This was independent of the type of tritium monitoring method used.

$$\text{Tritium loss per day (D}_2\text{O d}^{-1}\text{)} = F \times W \times 0.0168 \times \text{T-ratio} \quad (\text{E-2})$$

where

- F = exhaust stream ($\text{ft}^3 \text{min}^{-1}$)
- W = moisture content (grains lb^{-1})
- T-ratio = tritium concentration in the water extracted from the air sample by the dehumidifier, divided by the tritium concentration in the moderator
- 0.0168 = conversion of $\text{ft}^3 \text{min}^{-1} \times \text{grain lb}^{-1}$ to lb d^{-1} .

The “T-ratio,” which is the ratio of the tritium content of the water condensed from the air sample to the tritium content in the reactor moderator (the most recent value known), was introduced. The tritium values are obtained from laboratory analysis of the water collected (A in Equation E-1). Using these data, the loss of D_2O (in pounds per day) was calculated for the +148-ft elevation stack exhaust, 0-ft level, -40-ft level, and the purification exhausts (see [Figure 4.1-1](#) for air flow schematic of a reactor). The results of the +148-ft elevation were used to determine the total releases of tritium from the stack. The next step was to use the same laboratory analysis of tritium in the dehumidifier sample to calculate the tritium released to the stack. DPSOP-105-1886 PLCK ([Westinghouse](#) 1984a) outlines the procedures for analyzing the amount of tritium in the moderator. The water samples were sent to 735-A for tritium analysis ([Suttinger and Burgo](#) 1991). The conservative estimate of the error in determining tritium in the water sample because of equipment counting statistics is 5%. The precision of the tritium level in the sample at $0.038 \mu\text{Ci mL}^{-1}$ was 0.8% relative at the 95% confidence level ([Smith](#) 1989).

Although the dehumidifier monitoring method was used until 1986, other methods eventually were used simultaneously, and the newer methods were probably the primary sources

of atmospheric tritium release data after they were installed. DPSOL 105-1878-PLK ([Westinghouse](#) 1979) determined the loss of D₂O from the STM (which was a more modern monitor that was put into use in 1970 and is described in a [later section](#)) rather than laboratory analysis of the dehumidifier sample, even though the dehumidifier sample results were still reported on the data sheets. Later, the data sheets showed that the STM was used to determine the amount of tritiated water vapor that was released.

The final step of this procedure was to calculate the tritium loss in curies per day for the +148-ft elevation stack using the tritium value from the water condensed from the exhaust stream or the STM measurement. This same procedure was followed whether the tritium release data were from the dehumidifier; the STM in DPSOL 105-1886 PLCK ([Westinghouse](#) 1984a); or the latest monitor type, the BTM (which was put into use in 1988 and is described in a [later section](#)) in DPSOL 105-1887-PLK ([Westinghouse](#) 1988). Data sheets from 3 weeks in 1990 ([Westinghouse](#) 1979, 1990b) for one of the reactors (P, L, or K) include BTM sample results as well.

We have found no indication of how often the tritium concentration in the moderator was measured. Intermittent monthly data was found over the years ([Lee](#) 1998 - these data are included in [Chapter 4.1](#)). During the late 1950s, at the beginning of reactor operations, the tritium concentration gradually more than doubled over the course of 15 months. In the 1980s and 1990s, the monthly values stayed virtually the same. Additional evidence found on data sheets from the 1990s ([Westinghouse](#) 1990b) shows the value did not vary during the month.

Kanne Chambers

A method to compute releases of [radioactive](#) gases from the 105 building (reactor) stacks was required to compensate for the drawbacks of the dehumidifier (e.g., inactive during reactor outages) and the moderator inventory methods (e.g., differentiating between atmospheric releases and other types of moderator losses) ([Caldwell](#) 1958). The Kanne chamber (ionization chamber) was considered a good monitor for use following reactor shutdowns or when activity increases in the stack air were known to result from tritium releases rather than other [noble gases](#). Also, a method was needed that could measure tritium gas and vapor online in the Tritium Facilities where the releases were not predominantly tritiated water vapor.

[Ellett and Bulter](#) (1963) indicated that the only practical method of monitoring stack exhausts for tritium was an ion chamber called the Kanne chamber. Kanne air monitors were used as permanently mounted instruments for air monitoring. Two sizes of ionization chambers were used (51.5 and 18.5-L), and the current was amplified and measured with a Beckman micromicroammeter or a vibrating reed electrometer. The measurements were continuously recorded. [Longtin et al.](#) (1973) indicated, without providing reasons, that use of the smaller 18.5-L stainless steel chamber was preferable to the older 51.5-L aluminum chamber. A schematic of the Kanne chambers can be seen in [Figure E-1](#).

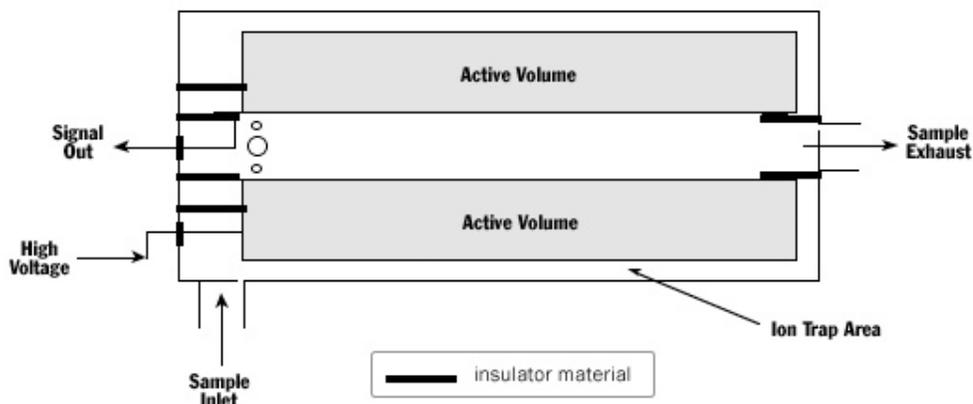


Figure E-1. Functional layout of a 18.5 and 51.5-L Kanne chamber (Litrell 1989).

The ionization chambers that were used consisted of a high-voltage electrode and a collector electrode, each separated from ground by a high-resistance insulator (Teflon). The current was generated as a tritium atom emitted a [beta](#) particle in the chamber. As the beta moved through the surrounding gas molecules, electrons were knocked off to form positive ions, and the electrons quickly formed negative ions. The positive and negative ions migrated toward the opposite charge elements in the chamber, creating a flow of current that could be measured by an electrometer or a picoammeter (Litrell 1989).

A linear relationship exists between the concentration of tritium in air and the current from the Kanne chamber if enough potential is present to collect all of the ions formed by radioactive decay of tritium in the chamber. Therefore, it is possible to estimate the active volume of an ion chamber to calculate the conversion factor of microcuries per cubic centimeter per ampere. By introducing a known amount of tritium, this factor can also be determined. Calibration curves were provided for each chamber size, and once determined for a type of ion chamber, this value will not change. It will be the same for identical chambers. The response of the 51.5-L chamber was $2 \times 10^7 \mu\text{Ci}/\text{mL}/\text{ampere}$ and the response in the 18.5-L chamber was $6.85 \times 10^7 \mu\text{Ci}/\text{mL}/\text{ampere}$. The measured accuracy was quoted as $\pm 10\%$ for low energy beta-emitting radioisotopes such as tritium. An external [gamma](#) source can also be used to check the ion chamber, so it is not necessary to use known concentrations of tritium in air to calibrate the chambers (Longtin et al. 1973; Litrell 1989).

Reactor process stack releases were monitored by Kanne chambers at an elevation of +148 ft (Hoy 1961). Here, the Kanne chamber detected releases to the atmosphere while a dehumidifier method was used to measure the releases quantitatively. An alarm was set to signal at a

predetermined release level. Because of noble gas activities present in the stack effluent, the Kanne does not give a quantitative measure of tritium released.

The Kanne was installed at the +50-ft level in buildings 232-H and 234-H. The basic configuration of the Kanne chamber in Buildings 232-H and 234-H can be seen in [Figure E-2](#). An air sample was drawn continuously through the ionization chamber. [Hoy](#) (1961) indicates that at SRS, the flow rates of the Kanne systems were set between 2 and 4 ft³ min⁻¹.

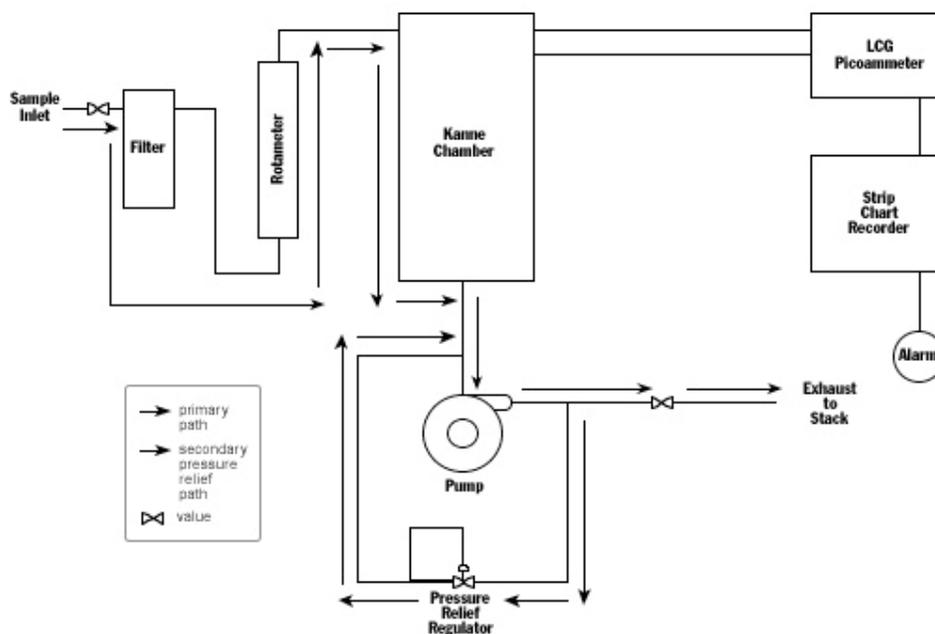


Figure E-2. Typical Configuration of the Kanne chamber monitor ([Litrell](#) 1989).

Difficulties in operating Kanne chambers included high gamma [background](#) fields and high noble gas activity in the air sample. Kanne chambers are affected by background [radiation](#) and cannot differentiate between radioisotopes (noble gases versus tritium releases in the reactor stacks). This caused problems with using them in the reactor but not for the Tritium Facilities stacks. Another drawback was that their sampling capacity was quite small. However, the great advantage of the Kanne ionization chamber was that it provided continuous sampling at a sufficient flow rate to provide real-time information. It was also found that the internal surfaces of the chamber could experience tritium buildup that required decontamination ([Longtin et al.](#) 1973; [Jacobson et al.](#) 1973). Consistency of the results using this method could be affected by system pressure ([Litrell](#) 1989).

Stack calibration, rather than only checking the monitor itself, was carried out in the Receiving Basin for Offsite Fuel (RBOF) and the four stacks in the Tritium Facilities. This was done by releasing spikes of about 100 Ci (in Building 238-H only 20 Ci was released because of the much lower height of the building stack) through process hoods into the system ([Davis](#) 1977). The results, presented in [Table E-4](#), show that the Kanne did an adequate job detecting the tritium released.

**Table E-4. Calibration Test on the 200-H Tritium Facilities Exhaust Stacks
Performed during Late February and Early March 1977**

Date	Stack	Tritium released (Ci)	Release detected by stack
			Kanne (estimated) (Ci)
2/28/77	234-H	110	105 ^a
3/3/77	232-H	104	104
	Lines I & II		
3/15/77	232-H	100	97
	Line III		
3/17/77	238-H	20	22

^a 234-H stack integrator was online during the test to compare results. It registered 100 Ci.

In 1989, the calibration methods for the ionization chambers that were used at SRS for measuring tritium in air were reevaluated. This is important because it may indicate that the calibration factors used during the earlier years were incorrect. [Table E-5](#) lists the documented calibration values that have been used since the late 1950s. The 1989 study confirmed that the factors being used at that time were adequate. Reviewing the factors used through time showed that there was a small decrease in the values. This change does not appear to be significant ([Litrell](#) 1989). If corrections were to be made to the tritium release estimates based on this information, it would be to decrease the earlier estimates slightly. The propagation of errors in the calibration factors used for the ion chambers was discussed in [Metlin](#) (1989). Metlin's calculations show that for the 51.5-L ionization chamber, the measurement uncertainty results in a calibration uncertainty of 2.3% of the reading; for the 18.5-L ionization chamber, it is 2.0% of the reading; and for the high volume chamber, it is 2.6% of the reading.

Table E-5. Documented Calibration Factors for Three Types of Kanne Chambers

Year	High range ($\mu\text{Ci cm}^{-3} \text{A}^{-1}$)	18.5-L Kanne ($\mu\text{Ci cm}^{-3} \text{A}^{-1}$)	51.5-L Kanne ($\mu\text{Ci cm}^{-3} \text{A}^{-1}$)
1959 ^a		6.85×10^7	2×10^7
1960s ^b		6.25×10^7	2×10^7
1971 ^a	3.3×10^9		
1974 ^c		6.25×10^7	2×10^7
1988 ^a	3.2×10^9		
1989 ^a	3.152×10^9	6.319×10^7	1.866×10^7
	$\pm 4.3\%$ at 3σ	$\pm 4.0\%$ at 3σ	$\pm 4.1\%$ at 3σ
1993 ^d	3.15×10^9	6.32×10^7	
	$\pm 11.5\%$	$\pm 11.5\%$	

^a Source: [Litrell](#) (1989).

^b Source: [Du Pont](#) (1965).

^c Source: [Westinghouse](#) (1974).

^d Source: [Westinghouse](#) (1993).

The discussion of [Hoy](#) (1961) on the calibration of ionization chambers used at the SRS for measuring tritium in air is useful for evaluating the uncertainty of the Kanne monitoring results. The following numbered list summarizes the main points:

1. During **calibration**, tritium absorbs on to the walls of the calibration system and there is a loss of tritium in the chamber. The calibration factors are not corrected for this, so the curves used provide pessimistic or higher tritium values. The article indicates that for the larger volume Kanne, there could be up to an 11.5% loss to the walls, and in the smaller chambers, a 28% loss compared to calculated responses.
2. The mixture of other beta-emitting gases in the sample can also cause an error in the measured values. The results are based on the average beta energy versus response of the system, which would be increased by the presence of increased beta emissions.
3. **Pressure and temperature** variations (flow rate) can cause errors (in 1961, the sample flow rate was set at 2-4 ft³ min⁻¹). *Stack Monitor Integrator System Pressure Factor Calculation* ([Westinghouse](#) 1989a) indicates that the Kanne chamber data are based on the chamber operating at atmospheric pressure. This procedure was developed in the late 1980s to calculate a pressure factor. Unfortunately, it does not explain how it was applied or how much difference it makes.
4. **Humidity** up to 96% seem to be OK. If condensation occurs in the instrument, there is a problem and it must be dried out. The determination of absolute humidity and temperature might also be important factors in determining the uncertainty of these measurements
5. If the air is filtered, there seem to be few problems with [contamination](#) in the sample; however, residual activity may build up. This may require air purging.

We considered these influences when conducting the uncertainty assessment ([discussed later](#) in this chapter).

Manual Integration of Stack Recorder Charts

Initially, the Kanne chamber results were recorded on a paper strip chart that was manually integrated by the health physics staff. The height of the trace on the strip chart was proportional to the [radioactivity](#) in the airflow. The area under the trace, minus the background, was related to the radioactivity release. The Kanne would measure tritium and ⁴¹Ar, so this method was best used when the source of the activity was known. The tritium releases from the stack were computed using the Kanne recorded chart as follows in Equation E-3 ([Caldwell](#) 1958):

$$Q = c [\sum (k_s A_s) - a_{bg} T] * 1.7 \times 10^6 \quad (E-3)$$

where

- Q = amount of tritium (Ci)
 T = the release time period (taken from chart) (hours)
 c = the air exhaust rate during time, T. This may be computed from the rated capacities of the exhaust fans (ft³ m⁻¹)
 a_{bg} = background tritium air activity (obtained by purging the Kanne with clean air) (Ci cm⁻³)

- A_s = area between trace and zero axis on recorder chart generated during time period, T, during which its amplifier was set at a particular setting, s ([sensitivity](#) setting of the amplifier) (in² or % of scale x in)
- k_s = proportionality constant depending on s, the chart speed, and chamber calibration (see [Table E-6](#))
- $\Sigma (k_s A_s)$ = sum of all products of k_s and A_s for each amplifier setting, s, that occurred during time period, T.
- 1.7×10^6 = conversion of time and area units.

[Caldwell](#)'s (1958) procedures for the reactors indicated that the area measurements A_s could be carried out using one of the three methods:

1. Straight-line trace method, where the chart is sectioned into rectangles or trapezoids and the area of each section is added to get the total.
2. Approximate integration method, where the sections are created at equal lengths along the time scale (base section); the heights of the right side of the sections are added and multiplied by the length of the time section.
3. Equal area method, where within a section, a line is drawn through the trace so that about the equal amounts of area are enclosed below and above the rule. The area between the zero axis and the drawn line is determined by the height of the line at the midpoint of the base (time) section; these sections are summed.

Table E-6. Summary of Proportionality Constant, k_x , Chart Speed, and Chamber Calibration^a

	s		k_s	
	Beckman settings (A)	% of scale measurements	% of scale measurements	inch measurements
I A	3×10^{-13}	3×10^{-14}	3×10^{-14}	2.7×10^{-13}
II A	10×10^{-13}	10×10^{-14}	10×10^{-14}	9.0×10^{-13}
II B	3×10^{-12}	3×10^{-13}	3×10^{-13}	2.7×10^{-12}
II C	10×10^{-12}	10×10^{-13}	10×10^{-13}	9.0×10^{-12}
II D	3×10^{-11}	3×10^{-12}	3×10^{-12}	2.7×10^{-11}
III A	10×10^{-11}	10×10^{-12}	10×10^{-12}	9.0×10^{-11}
III B	3×10^{-10}	3×10^{-11}	3×10^{-11}	2.7×10^{-10}
III C	10×10^{-10}	10×10^{-11}	10×10^{-11}	9.0×10^{-10}
III D	3×10^{-9}	3×10^{-10}	3×10^{-10}	2.7×10^{-9}
IV A	10×10^{-9}	10×10^{-10}	10×10^{-10}	9.0×10^{-9}
IV B	3×10^{-8}	3×10^{-9}	3×10^{-9}	2.7×10^{-8}
IV C	10×10^{-8}	10×10^{-9}	10×10^{-9}	9.0×10^{-8}
IV D	3×10^{-7}	3×10^{-8}	3×10^{-8}	2.7×10^{-7}

^a Source: [Caldwell \(1958\)](#).

If a logarithmic amplification system was used the following relationship (Equation E-4) was used ([Caldwell 1958](#)):

$$Q = c [k_e \Sigma A_e - a_{bg}T] * 1.7 \times 10^6 \quad (E-4)$$

where, in addition to the parameters defined above:

A_e = effective area below the trace in a section of the chart. The height of the area is taken as the ampere reading of the trace (A) and the base is measured inches (in)

k_e = chamber calibration $2.0 \times 10^7 \mu\text{Ci cm}^{-3} \text{A}^{-1}$

ΣA_e = sum of all A_e s during the tritium release of interest.

The area measurements could be made either as a constant activity trace (so it was a rectangle) or using the approximate integration method described above (referred to as the variable activity trace) (Caldwell 1958). Stack recorder charts for the Tritium Facilities (B232-H, 234-H, and 238-H) Kanne chambers were manually integrated to determine the tritium releases from the stacks. In later years, when the electronic SMI was being used, it was used to verify that the SMI and FORMS monitors (monitor used to determine the ratio of tritium vapor to tritium gas) were working correctly. The stack recorder charts were integrated for each shift at the beginning of the next shift. Special integration rulers were used for each stack to take into account the different airflow characteristics (Westinghouse 1994, 1991c).

The integration rules that were specific to the Tritium Facilities are summarized as follows (Westinghouse 1994, 1991c):

1. If the release rate is $<100 \text{ Ci min}^{-1}$, use one of the following:
 - use the constant rate
 - average the rates measured at the beginning of minute and the rate at the end of the minute
 - if rate increased and decreased, take into account the portion of the minute involved in the increase and decrease to use 1(b) and then add the portions together for the total 1-min release.
2. If the release rate is $>100 \text{ Ci min}^{-1}$, break the minute into three parts, determine the amount released during each third, and then take the average to determine the release for the 1-min period.
3. The actual activity release in all peaks must be corrected for stack background. This is done by multiplying the background by the time in minutes for each operation or peak shown on the chart and subtracting the background reading for the period from the uncorrected release totals determined in Steps 1 and 2 above.
4. All peaks must be identified and attributed to the process that would have caused it.

Stack Tritium Monitor

In the early 1960s, a STM system was developed by Du Pont engineers at the SRS (Ray 1991). The systems used up until this point were neither adequate for measuring rate and release or time of release (the dehumidifier) nor could they differentiate between tritium and other radioactive gases (the Kanne chamber). This new method was designed to allow for measuring tritium online in the presence of noble gases by combining the dehumidifier with a compensated ionization chamber (Kanne chamber). A prototype of the system was described in 1961 using a vibrating reed electrometer, an Esterline-Angus recorder, a control circuit, and a mechanical

counter to record the amount of tritium released ([Kincaid 1961](#)). The initial version is shown in [Figure E-3](#), and an updated version, put to use in later years, is shown in [Figure E-4](#).

In August 1965, the STM description was slightly different than the prototype developed in 1961. The stack gas sample was split in two: one-half going directly to a 4-L ion chamber measuring elemental tritium, tritium oxide, and noble gases, and the other half passing through a cold trap first for the removal the tritium vapor (HTO) and tritiated water (T_2O) before flowing through another identical ion chamber measuring elemental tritium and noble gases. The resulting current was proportional to the difference in activity between the two chambers, which is equal to the tritium activity removed by the cold trap, thus, allowing for an estimate of the amount of tritium oxide released ([Rampey 1965](#)). This method eliminated background and noble gas interference, but it measured only tritium in the form of HTO. This could be corrected if a catalytic recombiner cartridge was inserted ahead of the moisture trap to convert the T_2 to HTO and, therefore, include it in the measurement ([Longtin et al. 1973](#); [Jacobson et al. 1973](#)).

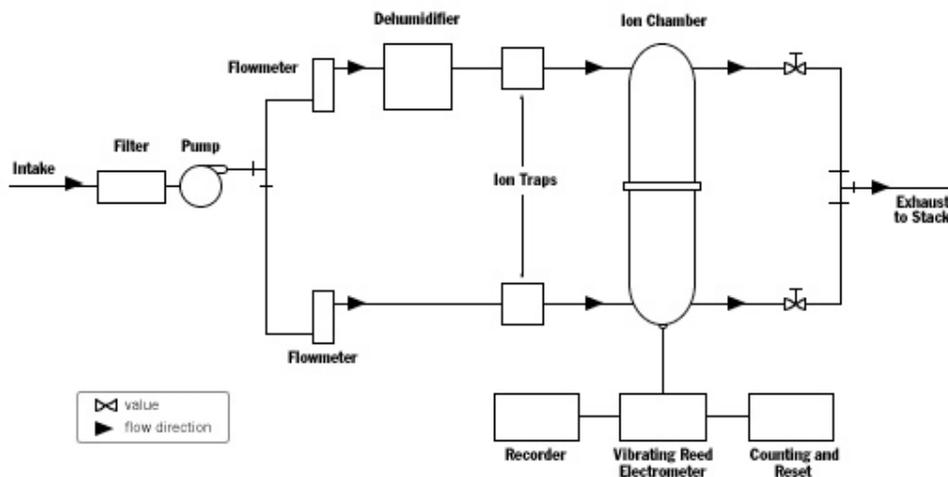


Figure E-3. Configuration of the early stack tritium monitor ([Rampey 1965](#)).

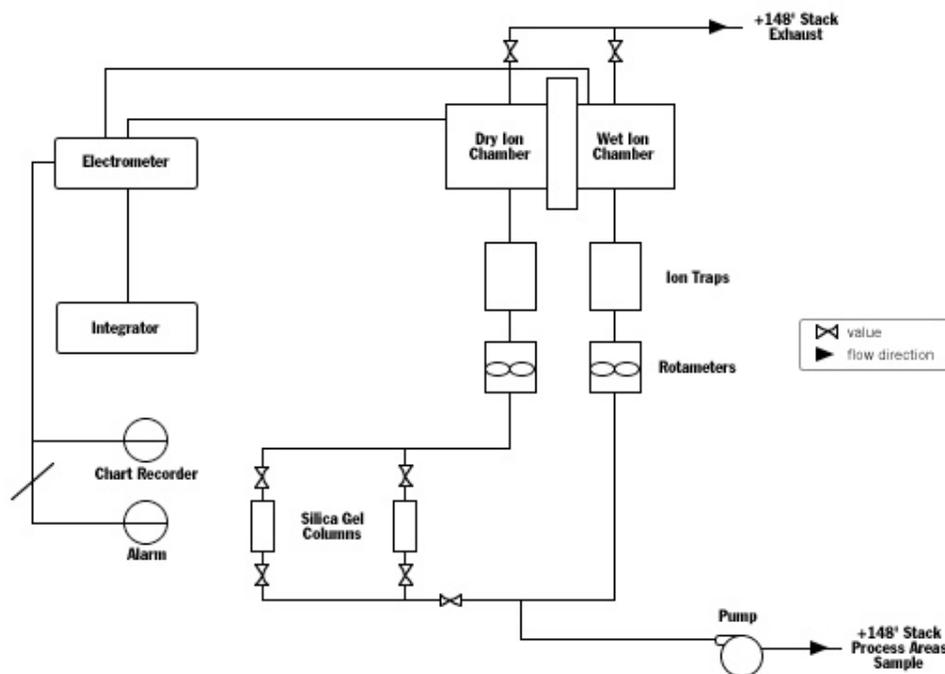


Figure E-4. Configuration of the later stack tritium monitor ([Westinghouse 1990c](#)).

Two Kanne ionization chambers were used, and the sample of exhaust gas flowed continuously through the chambers. A Beckman logarithmic micromicroammeter measured the current that was produced. A digital computer was used to integrate the ion chamber current data. The output signal from the micromicroammeter was converted to pulses, with the frequency of the pulses proportional to the rate of discharge of tritium. The computer automatically integrated this data and allowed for integration of tritium releases for up to 1 month. The Kanne chamber and integrator system covered a range of tritium concentrations from 6×10^{-6} to $60 \mu\text{Ci cm}^{-3}$. Tests showed that at the levels of 25 to 130 Ci of tritium, the accuracy for the monitoring system was better than 10% ([Albenesius and Meyer 1962](#)).

The minimum detectable tritium level in air when there were no other [radionuclides](#) present was $0.5 \times 10^{-5} \mu\text{Ci cm}^{-3}$. Therefore, the minimum detectable level in 105 buildings stack gas, when other noble gases were present, was reduced to $1.0 \times 10^{-5} \mu\text{Ci cm}^{-3}$ at full reactor power ([Rampey 1965](#)). The STM could be used to monitor stack exhaust during normal operations and shutdown. One disadvantage was that if there were a partial core melt accident, the higher levels of releases would render the system useless.

The stack monitor operated under slight positive pressure so that if there was a leak, air would not dilute the gas sample. Direct readings of gas stream activity could also be monitored. There might have been an increase in response time, but pulse indications caused by changes in noble gas activity were prevented ([Rampey 1965](#)). All components (except for the dehumidifier) were mounted in a 4-ft rack cabinet. The prototype of this instrument was demonstrated in 105-K ([Rampey 1965](#)).

The amount of tritium oxide released from the exhaust stream was calculated using Equation (E-5) ([Rampey 1965](#)):

$$C = K_1 N \quad (E-5)$$

where

- C = curies tritium oxide released (Ci)
 K_1 = constant determined from the stack monitor calibration and exhaust stream flow rate (Ci/counts)
 N = number of counts recorded.

On the daily data sheets, K_1 was determined from the use of a Keithly Amplifier Factor based on the multiplier range, and instantaneous calibration factor based on field calibration, K_2 , and the air exhaust rate (Westinghouse [1984a](#), [1990c](#)).

The rate of tritium released was determined by Equation E-6:

$$R = K_2 / t \quad (E-6)$$

where

- R = rate of release (Ci h⁻¹)
 K_2 = constant determined from the stack monitor calibration and the exhaust stream flow rate (Ci)
 t = time required for count (h).

Although the STM was able to discriminate tritium from other radioactive noble gases, it was only effective for monitoring tritium oxide. It was insensitive to elemental tritium, which was why it was used in the reactors and not in the Tritium Facilities. Another drawback of the STM was that it was more difficult to calibrate because tritiated water had to be handled.

In later years, the cold trap that was used to remove the tritiated water vapor was replaced by a 60-in.³ silica gel drying column. The silica gel provided a more efficient method for removing water than the cold trap, which fluctuated with changes in relative humidity and temperature ([Smith](#) 1989). An ion trap was also added and set at the inlet of each of the ion chambers to remove ions produced in the gas outside the chambers that might disrupt the proper tritium measurement. These traps consisted of 14 parallel plates with a potential difference of 70 V between adjacent plates ([Smith](#) 1989).

Stack tritium monitoring DPSOLs (Westinghouse [1984a](#), [1984b](#)) indicate the monitors were calibrated annually, when required by supervision, or when there was a question about the accuracy of the existing calibration factor. The calibration documentation provides equations to calculate the rate of drift of current measurements; a calibrations procedure print list; and the recommended initial installation procedure for prototype design, including routine operation and maintenance (Westinghouse [1984a](#), [1984b](#)). It was not easy to calibrate the equipment because tritiated water had to be handled by the technicians ([Merz](#) 1988).

A strip chart was used as an integrator/recorder of the average daily moderator loss rate. With the strip chart, it was possible to determine the instantaneous loss rate at anytime ([Smith](#) 1989). The total releases were integrated manually.

In 1965, releases from the STM and the existing dehumidifier in the reactor stacks were compared during normal operations ([Rampey 1965](#)). [Table E-7](#) provides the data. The releases are for a 24-hour sampling period ending at 2 p.m.

**Table E-7. Stack Monitor and Existing Dehumidifier System
Estimated Releases during Normal Operations of a Reactor in 1965^a**

Day	Estimated release (Ci d ⁻¹)	
	Stack monitor	Dehumidifier
Wednesday	218	336
Thursday	221	372
Friday	211	413
Saturday	162	341
Sunday	156	346
Monday	152	321
Tuesday	240	390
Wednesday	240	320

^a Source: [Rampey](#) (1965).

Although the trends seem to be the same for this reported comparison, the STM release estimates are, on the average, about 40% lower than the dehumidifier estimates. An illustration ([Rampey 1965](#)) of the hourly rate of loss over 5 days shows a variation of up to a factor of 2 (i.e., 6 to 12 Ci h⁻¹) over the course of 1 day. However on some days the release rates were more uniform ([Rampey 1965](#)). If the STM releases are about 40% lower than the dehumidifier estimates, the tritium release estimates based on the dehumidifier might have overestimated the actual tritium releases from the reactors. [Lee](#) (1998) reports data that indicate that the STM may have overestimated releases compared to the newer BTM; this would support the view that the STM overestimated the results rather than the dehumidifier. Insufficient evidence exists to draw a conclusion.

In July 1972, a memo was written to recommend a few changes to the newly installed STMs in each of the three reactor buildings ([Ray 1991](#)). The instruments were designed based on the prototype that was operating in K-Area, and there had been some problems in installation. The recommended changes were mainly to find better locations for equipment and improvements on instrument piping. We did not find documented evidence about how these changes might have improved measurement efficiency.

By 1991, it was recommended that the three remaining STM systems be replaced by a new BTM system for the following reasons:

- The STM chambers required extreme care and cleanliness in assembly to ensure that leakage current was not greater than the signal current.
- Condensation in the “wet” chamber could cause slight negative readings; the amplifier must operate in “fast” mode and is noisy; therefore, there was a slower response time.
- Releases to the environment would be underestimated if the recorder went offscale during a major reactor incident because of inaccessibility of the instrument.
- The amplifier was sensitive to temperature and vibrations that caused significant error.

- STMs were difficult to keep calibrated. The STM did not require mechanical calibration, but it required a “calculational” calibration using two factors: instantaneous (K_2) and daily loss rates (K_3) of process water, where $K_3 = K_2/60$. The K_2 was dependent on the response of the specific STM and the tritium concentration in the moderator; therefore, known quantities of tritium needed to be introduced into the chamber. This factor should be fairly constant, but historical data from the reactor areas do not support this ([Table E-8](#)). The error in the estimates of tritium releases would vary with the error of these factors ([Ray 1991](#)).
- These factors should be fairly constant over time for a particular STM in one of the areas ([Ray 1991](#)). If the calibration factors were off by 100%, the moderator loss rates and tritium releases rates would be incorrect by the same factor.
- The STM and BTM loss rate calculations were required to yield values within 25%. However, for low release days (during shutdown periods), the losses were below the STM sensitivity; therefore, it was not possible to test the BTM.
- There was a greater uncertainty of the STM-calculated moderator loss rates because of poorer counting statistics under conditions of high noble gas concentrations.

Table E-8. Calibration factors (K_2) for Stack Tritium Monitors in P-Area from 7/87 to 3/90

Date	Stack tritium monitor A	Stack tritium monitor B	Stack tritium monitor C
3/8/90	0.00452		0.00286
2/26/90		0.00156	
2/23/90			0.00179
2/20/90	0.00167		
2/11/90		0.00266	
2/9/90	0.00417		0.0125
1/22/90	0.00146		
1/10/90			0.00175
1/8/90		0.00172	
1/4/90	0.00106		
12/22/89	0.00490	0.00325	
11/15/89	0.00321		
11/6/89		0.00240	
8/3/89	0.00356		0.00128
7/3/89		0.00428	0.00179
6/18/89	0.00277		
5/18/89			0.00696
5/17/89	0.00229	0.00368	
5/12/89		0.00506	
5/11/89	0.00126		0.00940
4/15/89	0.00185	0.00600	0.00200
3/8/89		0.00183	
10/28/88	0.00229		0.00032
7/30/87	0.00078	0.00151	0.00087

Berthold Tritium Monitor

During the late 1980s, BTMs were added to monitor the moderator and tritium releases from the reactor areas. The first BTM was installed in L-Area in 1985 (Ray 1991). The BTM was developed because the threshold sensitivity of detection for the STMs was too low to detect the small stack moderator losses expected from the restarted L-Reactor, and the BTM could discriminate between the tritium and the radioactive noble gases released from the stack. The tritium activity in the moderator of L-Reactor was 175 times less than the activity in the P-Reactor and K-Reactor. As mentioned earlier, the tritium concentration in the moderator increased with reactor operation time, so a newly started reactor had much lower tritium releases. With such low activity, the existing STM was unable to detect a moderator loss of less than 2000 lb d⁻¹. The newly developed BTM was able to detect losses of tens of pounds per day. It was first installed in 105-L (Merz 1988). The BTM was going to be the main source of monitoring stack moderator loss rates in the +148-ft stack in L-Area and the dehumidifier would provide a redundant (or backup) measurement.

The BTM system was comprised of a tritium monitor and a radiation monitoring system, along with associated flow monitoring and control devices (Figure E-5). The gas flow proportional counter could discriminate between tritium and noble gases using two single channel analyzers employing rise time discrimination methods. The sample air was mixed with counting gas before it passed through a 1.3-L gas flow-through proportional counter. (In this chapter, the counting gas is called P-10. It is composed of 10% methane and 90% argon. This appears to be different from the P-10 referred to in documents from the 1950s, which appear to be referring to tritium gas).

The lower energy tritium beta particles, with a short range, interacted with the counting gas in a limited volume in the counter tube. The noble gases in the exhaust air reacted in a larger volume of the tube because of higher beta particle energies. These higher energies could be discriminated by using two single channel analyzers because of the difference in the pulse rise times (Merz 1988). Pulse rise time discrimination depended only on the energy of the emitted beta particle, not the chemical composition in which the isotope of interest was found. The BTM was equally useful in monitoring elemental tritium as tritium oxide.

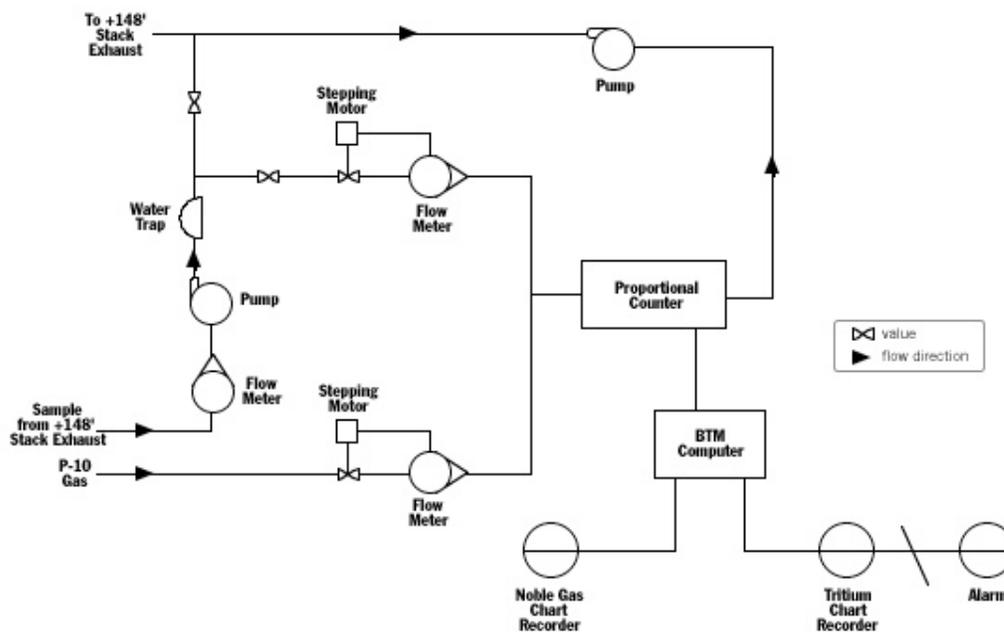


Figure E-5. Schematic of the Berthold tritium monitor (Merz 1988).

The accuracy of the tritium measurements from the BTM depended on the ratio of exhaust air to counting gas mixture. A control unit regulated this ratio with a light barrier control. The measurement was accomplished using a floating body flow-through meter using photoelectronic position scanning. The detector automatically shut down if the counting gas flow was lost (Smith 1989). After counting, the gas and sample were pumped out to the exhaust air system. A water

trap was added to the exhaust air system to alleviate the buildup of moisture in the sample lines and the counter tube.

The Berthold Analytical Instruments Company quoted a sensitivity that was not achieved in the field (0.01 μCi tritium per cubic meter air or between 0.01–0.04 lb moderator per day depending on the reactor). In fact, a sensitivity of $<0.5 \text{ lb d}^{-1}$ was found with operating experience. This was attributed in part to the uncertainties in ventilation airflow rates and moderator tritium content. In calibration exercises, the BTM was able to measure tritium in the presence of a simulated noble gas background to an accuracy of $<5 \text{ ppm}$ (Merz 1988). The threshold of detection for stack releases was improved by a factor of about 200 to $10^8 \mu\text{Ci}$ of tritium per cubic centimeter of air with the shift from the STM to the BTM (Westinghouse 1985; Merz 1988).

In 1988, a BTM was also installed in K-Reactor (Merz 1988), and there were recommendations to replace the existing BTMs with newer systems. It was also recommended that the STMs currently installed in each reactor building continue to be used to monitor tritium concentration of below grade, purification, and process room air. It was suggested that the use of the dehumidifiers could be discontinued because the STMs could be used as a redundant measure for the BTMs, but the data sheets we found indicate that all three systems continued to operate.

The procedure for BTM operation (DPSOL 105-1887-PKL) (Westinghouse 1988) describes how the equipment worked, and the Special Procedure SP-2381 (Westinghouse 1985) provides a Daily BTM Stack Loss Data Sheet requiring measurements at 4-hour intervals starting at 2:00 a.m. The daily stack loss (in pounds per day) was calculated by the product of the concentration of tritium in the moderator (microcuries per milliliter; weekly lab results), a conversion factor, the integrated counts in Channel B, and the average duct flow rate (cfm). Data sheets were used to calculate the instantaneous loss rate (Rp) using Equation (E-7):

$$R_p = k \times F \times B_c / T \quad (\text{E-7})$$

where

- k = conversion factor from DPSOL 105-1885A-PLK, *BTM/STM Calibration* (Westinghouse 1989b)
- F = flow in duct being sampled (cfm)
- T = tritium content in moderator, $\mu\text{Ci mL}^{-1}$ from latest laboratory result
- Bc = [Ba-Bo] – S[Ag-Ao]

where

- Ba = count rate from recorder or count rate meter Channel B
- Bo = Channel B background, Data Sheet 1 DPSOL 105-1885A
- Ag = count rate from recorder or count rate meter Channel A
- Ao = Channel A background, Data Sheet 1 DPSOL 105-1885A
- S = spillover factor Data Sheet 1 DPSOL 105-1885A.

As was described in the earlier section on dehumidifiers, the calculated loss of moderator in pounds per day was used to calculate the amount of tritium oxide released to the atmosphere. However, in this case, both elemental and oxide forms were included in the release estimate.

Some BTM data were available for the newly installed equipment in P-Area from May 1988. The BTM was reported to show an accuracy of measurement of $\pm 3\%$. It was suggested that a conservative estimate of $\pm 5\%$ be assumed to take into account 2% drift in the calibration measurement. At the time of the report, it indicated that an accuracy performance check had been shown to be adequate for every 6 months for the BTM; however, it seems that the check was done every 3 months (in accordance with DPSOL 105-1885A-PLK [[Westinghouse](#) 1989b]) ([Merz](#) 1988).

Two more upgraded BTM systems were installed in L-Area (June 1989 and July 1989) and three in KA-Areas (September 1990 and January 1991). The sensitivity quoted for the systems was $2 \times 10^{-8} \mu\text{Ci cm}^{-3}$ in 1-hour measuring time as compared to $1 \times 10^{-6} \mu\text{Ci cm}^{-3}$ for the STM ([Ray](#) 1991). In 1989, it was recommended that a third BTM be installed to increase the capabilities of the BTM system by dedicating a monitor to the 148-ft stack. This would allow increased availability for the other existing detector sampling points ([Terronez](#) 1989).

By 1991, there were three BTMs in each reactor area monitoring the total stack, at 0-ft elevation for the process and purification areas, and at +40-ft elevation areas. [Westinghouse](#) (1991d) provides the instructions for starting up the BTMs. At anytime the process water system contained moderator, at least two tritium monitoring systems should have been operable and monitoring the +148-ft level ventilation exhaust; one of the two monitors should have been a BTM. The third tritium monitoring system should have been operable and monitoring any ventilation exhaust air streams in the 105 building. The three BTMs were connected to six ducts and the atmosphere.

STM and BTM Redundancy

The STMs and BTMs could provide adequate surveillance of the release of process water because the reactor ventilation system was designed so that the process areas were maintained under negative pressure. During reactor operation, the air from the process room, purification, and -40-ft pump room passed through the filter compartments and released to the stack. By monitoring at the exhaust stack, the combined tritium levels from the process areas were measured. By having two tritium monitors at the +148-ft ventilation stream, it was possible to switch one of the detectors to monitor different ventilation streams to determine the source of the releases. It also provided a backup to the +148-ft monitor ([Merz](#) 1988).

The STMs were not removed and remained in use because of the broader range over which they could detect tritium in the presence of other radioactive gases. At the low end, this corresponds to very small process leaks throughout the reactor building to small and medium size leaks (the type that normally occur). A very large spill of tritiated water in the reactor building could overwhelm the existing BTM system by exceeding the existing measurement range. The gas flow-through ionization chambers saturate at higher concentrations than the more sensitive proportional chambers of the BTMs. The reported BTM ranged from $2 \times 10^{-8} \mu\text{Ci cm}^{-3}$ air to $2 \times 10^{-3} \mu\text{Ci cm}^{-3}$ air for a 1-hour counting period ([Merz](#) 1988).

The results of the STM were compared to the BTM daily (at a minimum of every 48 hours). The STM calibration required that it be within $\pm 10\%$ of the BTM reading. A conservative estimate of the STM accuracy was $\pm 15\%$, which included $\pm 10\%$ agreement and 5% drift error in the calibration measurement. The STM was to be calibrated every 3 months or whenever the calibration check warranted it ([Merz](#) 1988). However, data sheets for the 1990s indicate that in

many cases there were large differences between the STM and BTM results (see [Table E-9](#)). This is also true of the dehumidifier results that were reported. More of this duplicate measurement data can be found in [Lee](#) (1998).

Table E-9. Data for Tritium Releases from an Unknown Reactor (P, L, or K)^a

1990 date off	Moderator content ($\mu\text{Ci mL}^{-1}$)	+ 148 Moisture content (g lb ⁻¹)	+ 148 Tritium content in exhaust air ($\mu\text{Ci mL}^{-1}$)	+ 148 Dehumidifier D ₂ O loss (lb D ₂ O d ⁻¹)	+ 148 STM D ₂ O lost (lb D ₂ O d ⁻¹)	+ 148 BTM D ₂ O lost (lb D ₂ O d ⁻¹)	Tritium lost to the environment per day (Ci)
15-Apr	11600	70	0.40	10.0	208.3	21.4	102.5
16-Apr	11600	49	0.42	7.4	210	21.1	101
17-Apr	11600	58	0.26	5.3	198.3	11.7	51.8
18-Apr	11600	32	0.92	10.6	142.9	35.5	170.1
19-Apr	11600	56	0.88	17.9	60.3	37.5	185.9
20-Apr	11600	64	0.53	12.1	40.7	28.9	138.5
21-Apr	11600	10	0.43	1.7	33.3	22.7	108.8
22-Apr	11600	6	0.32	0.5	2.4	22.4	103.76
23-Apr	11600	6	0.36	0.75	9.6	18.4	88.2
24-Apr	11600	6	0.44	1.2	3	18.7	90
25-Apr	11600	6	0.36	0.8	0	17.9	83
26-Apr	11600	6	0.36	0.78	0	17.2	79.7
27-Apr	11600	6	0.40	0.8	25.9	17.5	81.1
28-Apr	11600	12	0.33	1.4	76.8	19.8	94.9
29-Apr	11600	11	0.29	1.1	24.4	17.6	84.3
30-Apr	11600	106	0.26	10.0	23.2	16.9	78.3
1-May	11600	38	0.24	3.4	23.9	18.1	83.9
2-May	11600	110	0.30	10.6	2.9	21	98
3-May	11600	76	0.23	6.4	30	20.5	95
4-May	11600	86	0.26	7.9	46.9	20.1	93.1
5-May	11600	84	NA ^b	NA ^b	184.1	15	71.9

^a Source: ([Westinghouse](#) 1979).

^b NA = indicated as not available on data sheet.

It is clear from the handwritten data sheets that the BTM data were used to determine moderator and tritium losses. The data sheets from the 1990s ([Westinghouse](#) 1990b) used the data from the BTM monitor, multiplied it by the concentration in the moderator, a conversion factor of 0.000413 (for pounds per day \times microcuries per day to curies per day), and the air density correction factor (dependent on temperature). The apparent differences in the measurements introduce some doubt as to how well and often the systems were actually calibrated against each other and how the equipment was maintained.

Stack Monitor Integrator

A memo written on 4/2/68 ([Du Pont](#) 1958) announced that the prototype SMI was delivered to Building 232-F, and preliminary tests showed it performed in a satisfactory manner. The equipment was designed to eliminate the difficulties encountered while manually integrating the curve drawn by the seven-decade semi-log chart paper recorder set up to record the Kanne chamber measurements. Its features included an instantaneous display (in curies of tritium released since midnight), alarm capabilities with selectable alarm thresholds, and automatic data printouts with time and date. A current-to-frequency conversion technique was used to convert the ionization current from a Kanne chamber into electrical pulses ([Figure E-6](#)). Digital techniques were then used to process the signals. The Health Physics group suggested calibration and maintenance procedures to be implemented to test if the integrator could be used to replace manual integration.

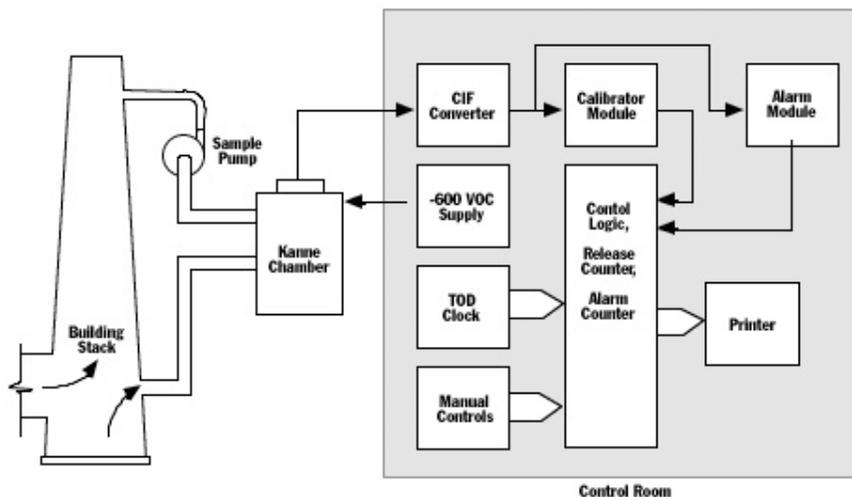


Figure E-6. Schematic of the stack monitor integrator ([Reinig](#) 1980).

In 1972, there was a request to get vendor bids to develop three SMI instruments for the current output of ion chambers in the 200-H Area. The instruments were to continuously record the output from the ion chamber and integrate it to be printed every 8 hours and reset after 24 hours. During an alarm, it was to continuously integrate the output and printout at 1-minute intervals. The accumulated reading was not to be reset, and a final printout was to be delivered at the end of the alarm condition ([Kilpatrick](#) 1975).

Another memo written 8/6/75 indicates that a prototype SMI was installed in Building 234-H in April 1974 and that field tests showed it accurate and reliable for measuring tritium releases. The long-term accuracy of the integrator electronics that was expected was better than $\pm 5\%$ of reading. However, the overall accuracy of the measured losses was dependent on the calibration of the integrator electronics, the Kanne chamber, and the measurement of the stack airflow. It was recommended that three SMI systems for Buildings 232-H and 234-H be fabricated for the

primary stack monitoring instruments and the existing monitoring systems be retained to keep track of the rate of releases.

A manual for the SMI dated July 2, 1980, (Reinig 1980) indicates that one integrator was installed in Building 234-H in January 1980 and two others were operating in 232-H by late 1980. This was confirmed in a Works Technical Monthly report (Du Pont 1982b). This system was compatible with a proposed FORMS monitoring system that could determine both the amount of HTO and HT released by the stacks. Exhaust airflow up the stack was indicated to be $6.31 \times 10^7 \text{ cm}^3 \text{ s}^{-1}$. Calibration was required every 6 months.

The calculation to determine the counts per curie released is as follows in Equation E-8 (Gelsie 1981a):

$$R = [x/y] \times [z] \times [1/k] \quad (\text{E-8})$$

where

- R = counts per curie released
- x = Kanne chamber constant = $1.6 \times 10^{-8} \text{ A } \mu\text{Ci}^{-1} \text{ cm}^{-3}$ (for 18.1 L)
- y = stack flow (232-H Line 3 = $131,000 \text{ ft}^3 \text{ min}^{-1}$ or $6.22 \times 10^6 \text{ cm}^3 \text{ s}^{-1}$; 232-H Line 1 and 2 = $104,000 \text{ ft}^3 \text{ min}^{-1}$ or $4.93 \times 10^7 \text{ cm}^3 \text{ s}^{-1}$; 234-H = $133,000 \text{ ft}^3 \text{ min}^{-1}$ or $6.31 \times 10^7 \text{ cm}^3 \text{ s}^{-1}$)
- z = current-to-frequency transfer function ($0.984 \times 10^{11} \text{ Hz A}^{-1}$ except for Line 3 in 232-H, which is $0.289 \times 10^{11} \text{ Hz A}^{-1}$)
- k = normalization constant.

Background must be subtracted from the total tritium released as displayed by the counters. It was necessary to compensate for error caused by the current-to-frequency (C/F) converter module (which linearly converted ionization current from a Kanne chamber into curies of tritium); leakage current; and Kanne chamber background. This was done by running room air through the Kanne chamber and measuring the frequency of the C/F converter. The C/F conversion period was measured automatically to within 100 microseconds by internal times. Background in terms of curies per minute was calculated from this conversion period. The normal background current was about 0.1 Ci min^{-1} . Most of the error was contributed by the C/F converter and was eliminated by subtracting background measured with the integrator (Reinig 1980). The amount of tritium released per minute was calculated per conversion period and the value of K was determined to provide the curies per minute measured by the SMI in C/F conversion. This procedure is described further in a memo dated January 20, 1981 (Gelsie 1981a).

In February 1982, the Works Technical Report (Du Pont 1982b) reported a 100-Ci test release to compare the SMI and charge recorder system in 234-H. A 4.5% difference was seen. Three more controlled release experiments were planned to continue to test the system.

A memo written in April 1981 (Gelsie 1981b) states that the SMI systems had been installed in 232-H and 234-H. A large stack release on March 27, 1981, was recorded by the SMI and provided release data that agreed with the manual integration of recorder traces within 10%. Both rate and quantity during the peak release period were recorded accurately, but there were some problems with the total display counter, which were quickly resolved.

Tritium FORMS Monitor

To monitor the form of the tritium (elemental tritium and tritium oxide form) released from the Tritium Facilities stacks, a monitor was installed in Building 234-H early in 1982. Like the STM, the monitor consisted of two Kanne chambers simultaneously measuring samples of stack effluent. One measured only elemental tritium after the removal of the oxide, and the other measured both forms of tritium (Du Pont [1982b](#), [1981](#); [Jacobsen](#) 1976).

The monitor used a pressure-swing dryer with self-regulating desiccant beds to remove tritium from the sample. A microcomputer-compatible electrometer measured total and elemental tritium. The difference between the two, as determined by the microcomputer, was available to tritium facility personnel and by telephone link. After a 1-week test in early February, it was determined that total tritium data agreed with Health Protection estimates of the stack release during that time. On February 12, a test release of 100 Ci validated the results with other measurement methods.

The gas stream sample was drawn into the sampler and mixed with tritium-free hydrogen carrier gas. The air moisture (both water and tritiated water vapor) was collected on a molecular sieve (A in [Figure E-7](#)). The elemental tritium gas was mixed with the tritium-free hydrogen carrier gas that passed through Column A. A palladium catalyst in Column B converted the hydrogen gas and tritium to oxide form. The water that was formed was collected on the molecular sieve in Column B. The moisture collected from the two columns was kept separate (samples A and B) and analyzed for tritium content. The results for Column A represented the tritium that was in oxide form and the water from Column B was the elemental tritium content in the air. The content of water samples A and B related the tritium concentration in air to the absolute humidity, air sampler flow rate, length of sampling time, and the flow rate of the tritium-free hydrogen gas.

An Air Effluent Monitoring Procedure (Westinghouse [1994](#), [1991a](#)) instructed facilities personnel that the FORMS measurements should be compared with the manually integrated stack release data from the Kanne chambers and the SMI results for each shift. This comparison was made to verify that the SMI and FORMS data were within 20% of the chart recorder data. If it was not, then the health physics officer responsible for the stack release data was contacted to determine which system was in error. A description of the samplers and flow monitoring devices at SRS ([Westinghouse](#) 1991b) indicates that the FORMS and Kanne chambers operated redundantly in the Tritium Facilities stacks. However, there is no indication of which data were used to determine the reported atmospheric tritium releases.

Hold Volume Monitor—Integrator for Tritium Facilities

In June 1982, a 6500-ft³ hold volume tank was installed in Building 300-H to contain accidental releases of tritium from Building 234-H ([Du Pont](#) 1982a). Sensors in the process hoods automatically triggered diversions to the hold volume tank. The monitoring capabilities of tritium diverted to the hold volume before that time were limited to approximately 0.05 g (about 470 Ci). A high-range ionization chamber accurately measures and integrates tritium in the range of 0.1 to 10.0 g. The monitor-integrator was designed to provide sufficient information to allow for decisions on tritium recovery after an accidental release. A C/F converter processed the signal

from the ion chamber to a totaling scale by fiber optic cable. We found only one mention of this tank in the SRS document database and no indication of when or if it was used.

UNCERTAINTY

Uncertainty analysis is an important part of a source term calculation. Because reconstructing the tritium source term at the SRS required using incomplete historical information, we used an uncertainty analysis to account for parameters that were either not completely understood or are defined in the literature with ranges, not specific values.

The uncertainty analysis combines all of the best scientific knowledge available about each parameter in a calculation to provide a distribution of results that is realistic given what we know about each variable. While a distribution is sometimes more difficult to understand, it is certainly a more accurate representation of releases than a single value.

We used the Crystal Ball[®] uncertainty analysis software package ([Decisioneering](#) 1996). The Crystal Ball[®] package is used within Microsoft Excel and allows the user to define the distribution of possible values for an input parameter about a defined mean. This assumption cell within Excel is used as a parameter in an equation cell. This equation is contained in a forecast cell. The results of the equation are calculated using a [Monte Carlo](#) analysis. A Monte Carlo simulation uses randomly generated numbers to measure the effects of the uncertainty expressed in the probability distribution in the assumption cell. A single trial generates a random number based on the probability distribution for each assumption, recalculates the equation within the spreadsheet, and displays the result of the calculation in a forecast chart. This forecast chart represents the results of the many calculations for the series of random numbers as a probability histogram. The more Monte Carlo trials run, the more continuous this histogram appears. This histogram can then be fit to a more conventional distribution, and the statistics describing that distribution are given by the Crystal Ball[®] software, accounting for the uncertainty in the input variables.

The results of this analysis can be used in the next phase of the [dose reconstruction](#) for SRS to estimate the range of possible releases using the source term data presented in this report. We also used the uncertainty analysis to determine whether there was a need to correct the source term data reported by the Site using different types of monitors.

Uncertainty for all monitor types was determined in generic terms. That is, the uncertainty was calculated about a mean of 1 so it could be applied to any value for the tritium released during the time period for which uncertainty was calculated. If the uncertainty for a given calculational technique has two components (each having an uncertainty of $\pm 10\%$), that uncertainty would be applied to a [median](#) of 1, the two factors would be multiplied, and the uncertainty would be calculated using Monte Carlo techniques to produce the total uncertainty for that technique. The final uncertainty can then be applied to all release estimates determined using that measurement technique.

When uncertainty is presented or was discovered in the literature to be represented by plus or minus some percentage value, the distribution was assumed to be normal, with the percentage value representing two standard deviations.

The uncertainty analysis is presented by type of monitor below. Because of the use of similar or identical components in the different equipment, in some cases the discussion of the different monitoring techniques overlap.

Dehumidifiers

In principle, dehumidifier measurements should be a reliable way of collecting tritium water vapor. As long as the coils were kept cold at all times and the sampled air was well mixed, the method should provide reliable and consistent results because it relied on collecting tritium vapor and measuring the amount of tritium in that water. Unfortunately, this method does not collect tritium gas. Therefore, there was no way to estimate total tritium releases to air, only releases that occurred from moderator loss.

The uncertainty inherent in the technique for measuring moderator loss by collecting water vapor with the dehumidifier was subjectively estimated at $\pm 15\%$ (2 standard deviations or 5 and 95% limits on a triangular distribution).

It is nearly impossible to correlate moderator loss to production. In fact, it is possible that moderator loss may have been higher during shutdown or maintenance because the reactor would have been open and moderator would have been more likely to evaporate. Dehumidifier readings were not commonly collected during shutdown because the technique was assumed to be reliable only during routine continuous operations ([Caldwell 1958](#)).

Mixing Within the Stack

The [aliquot](#) of stack effluent to be passed through the dehumidifier was removed from the exhaust stream. Sufficient mixing should have taken place by the time the exhaust stream reached the level where the sampling takes place because no inflow or outflow points exist in the stack between 0 and 148 ft. This was considered a negligible source of uncertainty.

Calculational Method

The uncertainty related to the tritium release obtained from the dehumidifier measurement was based upon the factors in the following equation used to estimate the tritium release to the atmosphere:

$$H_3 = H \cdot A \cdot (\text{days / month}) \cdot K \cdot D \quad (\text{E-9})$$

where

- H_3 = tritium discharged from stack (Ci)
- H = average absolute humidity of stack air (grains lb⁻¹)
- A = tritium in dehumidifier sample (average daily dehumidifier stack) ($\mu\text{Ci HTO l}^{-1}$)
- K = conversion factor; obtained from a calibration curve of K factor versus airflow in cfm; converts μCi to Ci, cfm (from the x-axis of the curve) to cfd (unit retained in conversion factor), and grains lb⁻¹ to l ft⁻³
- D = air density correction factor.

Absolute Humidity

Absolute humidity was also averaged for use in this equation. Absolute humidity was not usually measured; it was calculated from measurements of temperature and relative humidity. These two quantities were measured three times daily at each change of an 8-hour shift, and averaged before conversion to absolute humidity. As temperature or relative humidity increases, so will absolute humidity. We located humidity data from the SRS corresponding to stack measurements from 1989. These data indicate that humidity in the stack may have been dependent on outside humidity, with higher values for humidity appearing in the summertime. Outdoor temperature may have had a similar impact on stack temperature.

Historical weather data provided by the National Oceanic and Atmospheric Administration (NOAA) from their web site (<http://www.noaa.gov>) showed that relative humidity during summer months usually reached a maximum of 95%, with evening lows of 55%. During winter months, relative humidity reached an average high of 80% and an average low of 45%. Although these average values changed from day to day, over the course of a month they remain relatively consistent. In fact, NOAA's compilation of weather data from 1961–1990 showed that the average monthly relative humidity at the Augusta, Georgia, airport (the closest NOAA station to the SRS) was consistently around 70%, with the lowest average in April of 64% and the highest average in August of 76%.

Temperature fluctuated by about 20°F throughout the day during all seasons. The variation in temperature in the summer went from a high around 90°F to a low around 70°F, in winter from a high around 56°F to a low around 36°F, and during the spring and fall from a high around 75°F to a low around 55°F. Monthly averages compiled by NOAA from 1895–1996 reflect these same ranges.

[Figure E-7](#) shows the relationships between temperature, relative humidity, and absolute humidity. Absolute humidity varies more at higher temperatures with changes in relative humidity than it does at lower temperatures. During the summer months, with a daily change in relative humidity from 95 to 55% and a temperature change from 90 to 70°F, absolute humidity may have varied from 0.3 to 0.9 g ft⁻³. For winter months, variation in humidity from 80 to 45% and in temperature from 56–36°F may have varied absolute humidity from 0.05 to 0.2 g ft⁻³. Using the average value of the extremes may have resulted in uncertainty ranging from 50% to a factor of 2. Because it was not possible to determine at what time of the day measurements of humidity and temperature were taken, the [bias](#) in absolute humidity values cannot be determined. The uncertainty distribution was assigned a uniform shape, with minimum and maximum values of 0.05 to 0.2 g ft⁻³.

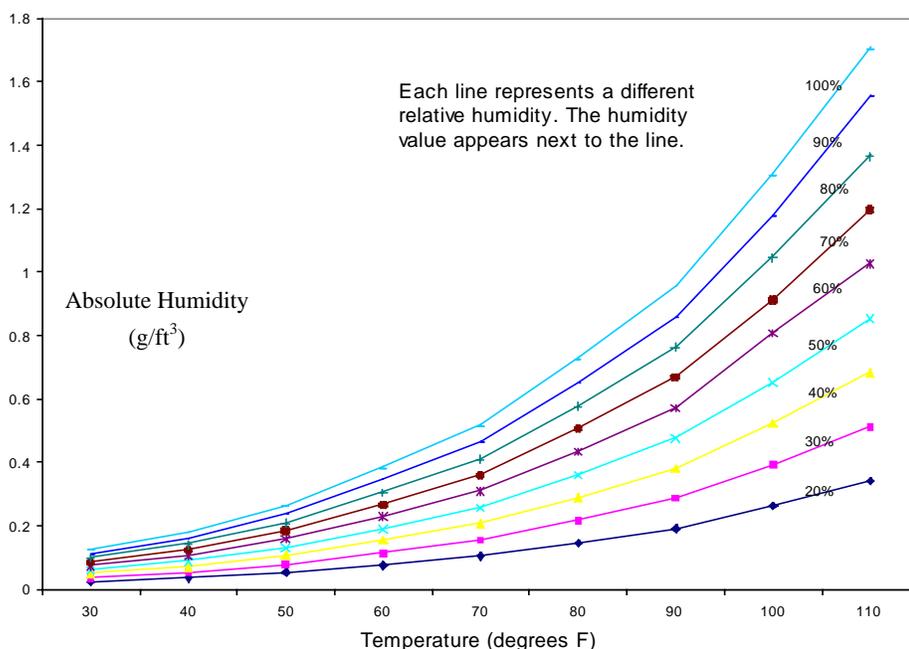


Figure E-7. Temperature, relative humidity, and absolute humidity relationships. Each line on the figure represents a different relative humidity and shows the absolute humidity as it varies with temperature.

Tritium Concentration in Dehumidifier Sample

The average daily dehumidifier stack sample condensate was counted using liquid scintillation counting techniques ($\mu\text{Ci HTO L}^{-1}$). The dehumidifier water was counted in a liquid scintillation counter with a sensitivity of $3 \times 10^{-7} \mu\text{Ci mL}^{-1}$ ($\pm 10\%$) (Longtin et al. 1973; Jacober et al. 1973). Estimates of error from the SRS low-level tritium laboratory predict error in liquid scintillation counting of $\pm 15\%$ (Ostland and Berry 1968). For this uncertainty calculation, liquid scintillation counting uncertainty was estimated at $\pm 20\%$ and assumed to be normal based on indications in the supporting document.

Conversion Factor

The uncertainty in the conversion factor K was directly related to the uncertainty in the airflow through the stack, which could have varied up to 20% for any given condition and by over a factor of 3 between shutdown and normal operations. For normal conditions, with airflow variations up to 20%, K could have ranged from 7.0×10^{-4} to 1.0×10^{-3} , with a median value of 8.5×10^{-4} . For shutdown conditions, the variation in K was from 2×10^{-3} to 2.95×10^{-3} , with a median value of 2.45×10^{-3} . This range in K is described graphically in a July 1965 Du Pont operating procedure (Du Pont 1965).

Although each of these uncertainties represents about a $\pm 15\%$ variation from its respective median, it is important to estimate the uncertainty in the total variation in K . Monthly tritium release values come from both normal and shutdown conditions, and there is no way of

distinguishing between the conditions. The total uncertainty in K is a factor of 4, or approximately a factor of 2 in either direction from the median of the two central values for normal and shutdown conditions. The distribution of K was assigned a triangular shape, with minimum, median, and maximum values that represent this factor of 4 of 0.42, 1, and 1.8, respectively. This quantity is unitless and serves only as a calibration factor.

Air Density Correction Factor

The air density correction factor, D , is dependent on temperature. For temperatures below 58.5°F, the correction factor is 1.035; for temperatures between 58.5 and 77°F, the correction factor is 1.000; and for temperatures above 77°F, the correction factor is 0.987. To create a change in the air density factor would require a rather large change in temperature. At most, the air density factor resulted in about a 3% change from the median value of 1. We considered this an insignificant source of uncertainty and did not consider it further.

Reporting a Monthly Release Value Based on Selected Daily Measurements

Equation (E-9) calculated monthly tritium release estimates by averaging the dehumidifier measurements taken three times during the week and humidity measurements taken at the same time. Daily measurements would have been available had the dehumidifier been run and collected from each day, but it was difficult to discern whether this was the case. However, there were indications that it was not run during reactor outages. Documents were found that show that the dehumidifier was run only 3 days a week for 24 hours ([Caldwell](#) 1958). Data from 1989 were examined and showed that dehumidifier readings did not exist for each day; however, because the dehumidifier was not the preferred measurement technique in 1989, it is difficult to draw any definitive conclusions from this data set. It is clear that these data were probably not used to report releases but merely existed as an emergency measurement backup.

Although [Westinghouse](#) (1982) indicates that during later years the dehumidifier was run every day, references from the time period during which dehumidifier measurements were used to estimate releases indicate that once every 3 days was the operating procedure ([Caldwell](#) 1958). For this reason, we have chosen to use the conservative assumption that the dehumidifier was only operated once every 3 days and include this source of uncertainty in our calculations.

We examined the 1989 data, however, to explore the effect of averaging a few daily values to obtain monthly totals. If a dehumidifier measurement for each date in a month existed, the monthly average closely reflects a median value between the maximum and minimum value in a month. The average multiplied by the number of days in the month was a good prediction of the sum of the measurements. When 4 or more days of data were blanks, however, the average was generally much smaller than the maximum monthly value. This may indicate that higher dehumidifier readings had a tendency to be damped out when a number of data points during a month were missing. Additionally, nothing was known about the dates for which no measurements are taken. For months during which numerous measurements were missing, little else could have been done other than averaging existing points and multiplying by the number of days in the month.

Using the data from the later years of operations, the month of August 1989 reflected a number of data values that might be expected from the period of time when measurements were taken for 3 days a week. [Table E-10](#) summarizes the data for August 1989.

Table E-10. August 1989 Dehumidifier Data^a

Number of existing data points	17
Average of existing data points	9.49
Average × number of days in month (31)	294
Sum of existing data points	161
Maximum value	50.3
Minimum value	1.4

^a Source: ([Westinghouse](#) 1979).

For this month, only 17 data points existed. The average of the 17 values clearly reduced the effect of the maximum value during the month, which was 50.3. If 3 of the 14 missing points were in the same range as the maximum, this quantity added to the existing sum of the values would about equal the existing average spread over the month. That still leaves 11 days without data points, indicating that the average may underestimate the sum over all days. Alternately, minimum values could have existed on each day, making the average an overestimate. In the worst case, the uncertainty in the average may have been a factor of 2 in our analysis. This uncertainty was reflected as the maximum and minimum values of a uniform distribution of possible values.

Overall Uncertainty

The total uncertainty in monthly dehumidifier values was calculated using Monte Carlo statistical techniques and 3000 generations of random numbers. The shape of the uncertainty factor results for a monthly source term value was fit to a lognormal distribution with a [geometric mean](#) (GM) = 0.99 and a [geometric standard deviation](#) (GSD) = 2.3. To determine the range in the monthly source terms that this uncertainty represents, the median source term value should be divided and multiplied by the GSD to obtain one standard deviation on either side of the median value, respectively.

Silica Gel

The operation of the silica gel is quite similar to that of the dehumidifier, and many of the same parameters affect the uncertainty. Moderator loss is calculated from the silica gel using Equation E-10.

$$E = \frac{A \cdot B \cdot D}{100} \quad (\text{E-10})$$

where

E = Moderator loss (lb day⁻¹)

- A = Absolute humidity (lb water (lb air)-1)
- B = Ventilation airflow (lb air day-1)
- D = $(C - 0.0147) 1.11$ (weight %)
- C = Sample analysis lab results (mol %).

Silica gel samples were taken out of the ventilation airflow of the stack 3 days a week for approximately 3 hours at a time.

Absolute Humidity

The absolute humidity was described in detail in the section above on dehumidifier uncertainty. The major difference between how this parameter affects the moderator loss calculated from the silica gel as opposed to the dehumidifier regards the sample duration. Silica gel samples were only collected for 3 hours on any given day rather than 24 hours at a time.

Data from the NOAA web site were again used to examine the change in temperature and relative humidity over shorter time periods. It appears that it is reasonable to assume that temperature will not vary by more than 20° and humidity by 20% over any 3-hour period. From the NOAA data, this is probably a conservative assumption. Even at high temperatures and relative humidities, the fluctuation in the absolute humidity is no more than about a factor of 2 total. This will be used as the uncertainty estimate for the equation parameter A , distributed in a triangular fashion.

Ventilation Airflow

The ventilation airflow has been previously defined to have an uncertainty of $\pm 20\%$ during a given operating condition and by over a factor of 3 between shutdown and normal operations. Because there is no way of knowing for certain during which condition any measurement was made or whether measurements were always taken during a single condition, the uncertainty must be assumed to be a factor of 3 for the parameter B , triangularly distributed from the median of the two central values for normal and shutdown conditions.

Sample Analysis Result

The sample analysis result, C , is the only parameter with any impact on the uncertainty of parameter D . Sample analysis uncertainty is affected by the duration of the sample as it relates to averaging effects and the sampling procedure and analysis uncertainty.

Silica gel moisture collection is still used today as an effective means of collecting water vapor contaminated with tritium. This method is still used because it is accurate when used properly and without exposing the gel to more moisture than it can collect, resulting in missed sample. The sampling procedure and analysis uncertainty was subjectively estimated to be $\pm 20\%$, normally distributed.

The effect of averaging 3 hours of data to represent a day and 3 of these days a week to represent a month introduces some uncertainty into the calculation. Dehumidifier data were examined to explore the effects of averaging daily values to obtain monthly totals. The uncertainty in this technique was estimated to be a factor of 2. It is more difficult to assess the

impact of averaging 3 hours to represent 24 hours, but we can assume this averaging may have a similar impact to the monthly averaging and assign a factor of 2 uncertainty. This makes total uncertainty in the averaging techniques around a factor of 4. This uncertainty is reflected as the maximum and minimum of a uniform distribution of values.

Total Uncertainty

Total uncertainty in the silica gel values for moderator loss was calculated using Monte Carlo statistical techniques and 3000 generations of random numbers. The shape of the uncertainty factor results for monthly moderator loss values was fitted to a lognormal distribution with a GM = 1.15 and a GSD = 2.16.

Kanne Chamber and Stack Monitor Integrator

Kanne Chambers

The underlying principle of a Kanne chamber is good as long as the Kanne chamber's size is sufficiently large to allow an ionization from a low energy beta emission to take place and be absorbed within its volume. The SRS reported the efficiency of this counting system as $\pm 10\%$ for low energy beta emitters such as tritium. This is used as the uncertainty for the basic Kanne chamber measurement technique.

Mixing of Air within the Stack. The aliquot of stack effluent was removed from the exhaust stream at the 148-ft level. Sufficient mixing should have taken place by the time the exhaust stream reaches this level because no inflow or outflow points exist in the stack between 0 and 148 ft. This is a negligible source of uncertainty and is not considered further in the uncertainty estimates.

Calibration. Kanne chambers must be calibrated before use to determine the ionization current produced per unit of radioactivity in the volume of air passing through the chamber. After a calibration was completed, the calibration value should not have changed for the detector. The complicating factor would be if a calibration were performed incorrectly and permanently skewed the results of the chamber.

[Hoy](#) (1961) related operational experience with calibrating Kanne chambers at the SRS and identified some possible problems with the calibration and use of Kanne chambers for accurate measurements.

The calibration factor for the Kanne chamber may have been miscalculated because adsorption of tritium onto the walls of the Kanne chamber may not have been accounted for. This had the effect of decreasing the experimental response of the chamber during calibration (less current per unit tritium in air); thus, overpredicting total concentration of tritium in air for the life of the detector. Experimental results from the calibration of Kanne chambers at SRS indicated that wall losses could have been as high as 28% for the smaller chamber or 11.5% for the larger chamber ([Hoy](#) 1961). These wall losses would have increased the Kanne chamber measurements by the same amount, requiring a reduction in the calibration factor to achieve the actual result.

Because calibration was done by measuring tritium in the chamber, the general measurement uncertainty of $\pm 10\%$ also applies to the calibration uncertainty. The total uncertainty in calibration was calculated by combining the measurement uncertainty and the wall loss

uncertainty and fit to a normal distribution with a mean of 0.91 and a standard deviation of 0.11. This corresponds to 5 and 95% values -20% and +17% of the mean, distributed relatively normally.

An operational disadvantage of the Kanne chamber was that all molecules creating ionizations in the chamber looked the same because the chamber operated within the ionization region and did not distinguish between energies of the particles deposited. The fraction of the average beta energy deposited in the chamber for different radionuclides and, therefore, the response for each nuclide could be estimated for a Kanne chamber. The fraction of the energy that the ionization chamber captured decreased as the average beta energy increased. The ionization current caused by radioisotopic decay was proportional to the average energy deposited per disintegration. If the relative abundance of each isotope in the gas mixture was known, the amount of ionization because of decay of each isotope could have been estimated. A calibration factor was determined for each beta-emitting gas that was likely to have been present within the Kanne chamber. This method of apportioning the energy deposited was highly dependent on knowing the isotopic ratios of the gases in the chamber, and it involved calculations instead of actual experimental measurements. This source of uncertainty is difficult to quantify.

Some guidance was gained from a 1989 document that revisited the Kanne chamber calibration ([Litrell 1989](#)). This work outlined the discovery that the calibration factors established before the use of a Kanne chamber were essentially good values. If the ratio of the gases was known, the impact of noble gases on the measurement for tritium was reasonably well understood. The primary noble gas component in stack air that interfered with tritium was ^{41}Ar . Uncertainty depended on the ability to estimate the ratio of these two gases. This must have been difficult because it was recommended that the Kanne chamber be used only following reactor shutdown or when activity could be attributed to tritium ([Caldwell 1958](#)). Supporting documentation seemed to imply that this was a large source of uncertainty and should be avoided if possible. Currently, there is no way of knowing if the Kanne chamber was used under ideal conditions. The uncertainty related to noble gas interference was subjectively estimated as a factor of 2, uniformly distributed about the mean, because no quantitative information was available and some estimate of uncertainty resulting from this factor was necessary.

Temperature and Pressure. Variations in temperature and pressure of the chamber may have had an impact on the consistency of the results. System pressure affected the ionization rate. The calibration memo ([Litrell 1989](#)) indicates that this could have easily shifted the Kanne results by $\pm 10\%$. The variations because of temperature and pressure were quantified as normally distributed about the mean with an uncertainty of 10%.

Relative Humidity. Relative humidity above 95% had the potential to cause condensation within the chamber, but Kanne chambers have been known to operate well even at 100% humidity. Although relative humidity in the SRS area could have been high during the summer, historical records indicated that humidity above 95% was rare, even as a daily high. The condensation factor was assumed to have a negligible effect on the Kanne chambers, and no uncertainty was applied.

Manual Integration. The Kanne chamber measurements were manually integrated to produce tritium stack release results before digital integration techniques were in place. The area under the curve of changes in current with time, as recorded by the chart recorder, was proportional to activity passing through the chamber. The amplifier used on the Kanne system

was logarithmic, as was the paper charting the trace, so Equation (E-11) was used for logarithmic integration of the stack charts:

$$Q = c \left[k_e \sum A_e - A_{bg} T \right] \cdot 1.7 \times 10^6 \quad (\text{E-11})$$

where

- Q = amount of tritium (Ci)
- c = air exhaust rate during time T ($\text{ft}^3 \text{ min}^{-1}$)
- k_e = chamber calibration ($\mu\text{Ci cm}^{-3} \text{ A}^{-1}$)
- A_e = area under curve during time T (A hours)
- T = release time period (hours)
- A_{bg} = background air activity (Ci cm^{-3})
- 1.7×10^6 = conversion from hours to minutes and from cubic feet to cubic centimeters.

The value of k_e was the chamber calibration for the appropriate Kanne chamber and would have varied with the uncertainty described previously. There should be no additional uncertainty added to the constant than the factor of 2 inherent in the chamber collection of noble gases and the uncertainty related to wall losses.

Air exhaust rate, c , was assumed to have an uncertainty of $\pm 20\%$ at the 95% confidence level as identified in previous sections. For our analysis, a triangular distribution was assumed, with 5 and 95% values set to $\pm 20\%$ of the median value.

The estimate of area under the Kanne chamber current trace had units of ampere hours because the area is calculated by multiplying some vertical distance on the chart (ampere) by the horizontal distance (hours). The uncertainty in area was related not only to the estimate of area using prescribed methods but also to operational uncertainty. This uncertainty was described earlier in this section as $\pm 10\%$ under standard operating conditions and an additional $\pm 10\%$ contributed by variations in pressure within the chamber. The area estimate should have been reasonably good, particularly because the current traces produced by the Kanne chambers were somewhat constant through time. The method used to calculate area under the trace depended on the appearance of the trace. If the trace was quite regular and straight, the straight-line method was used, dividing the trace into a series of rectangles and trapezoids, calculating the area for each section, and adding up the areas of the segments. If the trace was more irregular, the approximate integration method was used. The approximate integration method involved marking off sections of equal length along the x-axis, adding together the heights of the right sides of the segments, and multiplying by the distance (in units of hours) between the reference lines. If the appropriate area method was used with the appropriate graph type, this area estimate should be fairly certain. Because the uncertainty was negligible, we did not make a subjective estimate for this factor.

Stack Monitor Integrator

In later years, the manual integration technique described above was abandoned for a more reliable digital one, referred to as the SMI. The signal from the amplifier was sent directly to a digital processor, and results were available continuously and immediately. The digital signal was converted from current to frequency and normalized to 1 count per curie released so the number

of counts over the integrated period of time was related one-to-one with the curies of tritium released from the stack.

The conversion factor for count per curie released is shown in Equation (E-12). This factor took the chamber calibration, stack flow rate, and the current to frequency transfer function into account.

$$R = \left[\frac{x}{y} \right] \cdot z \cdot \left[\frac{1}{k} \right] \quad (\text{E-12})$$

where

- R = counts per curie released
- x = Kanne chamber constant ($\mu\text{Ci cm}^{-3} \text{ A}^{-1}$)
- y = stack flow ($\text{cm}^3 \text{ s}^{-1}$)
- z = current to frequency transfer function (Hz A^{-1})
- k = normalization constant.

If R is set to 1, the normalization constant can be solved for. Electronically, normalization was accomplished using a calibration unit attached to the SMI that calibrated the signal and normalized it to 1.

The same uncertainty that applied to the Kanne chamber collection, chamber calibration, and stack flow also applied here. Additionally, electronics used in the digital signal conversion had some associated uncertainty.

The C/F converter linearly converted the ionization current from the Kanne chamber to a user-defined frequency. The input current to the machine charged a capacitor, which produced a voltage ramp. For a given increase in voltage, a trigger was fired that produced a pulse whose width was equal to the user-defined frequency. The trigger fire also discharged the capacitor and restarted the cycle. The uncertainty in this setup was related to the time it took the capacitor to discharge and any current that was lost during that process, as well as the leakage current inherent to the equipment. This source of uncertainty was negligible, and it was not included in the calculation.

In summary, the normalization technique had uncertainty in the airflow and chamber calibration factor, but no additional uncertainty was added for the nominal amount that would have been related to the electronics.

Overall Uncertainty

Monte Carlo techniques were used to combine the different sources of uncertainty in Kanne chamber measurements with 3000 generations of random numbers. Because no uncertainty was applied to the area estimation or electronic integration techniques, the uncertainties for the two different types of Kanne stack chart integration were the same. Data from the uncertainty calculations were widely spread and not well fit by any distribution, but they were reasonably approximated by a lognormal distribution with a GM = 0.9 and a GSD = 1.8. To calculate uncertainty in release estimates, the GM is multiplied by the Kanne chamber estimate to provide a better estimate of a median value. One standard deviation of that median value is calculated by

multiplying and dividing the median by the GSD. Five and ninety-five [percentile](#) estimates are obtained by multiplying and dividing by the square of the GSD.

Stack Tritium Monitor

The STM simply adapted the use of the Kanne chamber and the silica gel into a piece of equipment that could more readily distinguish between tritium and noble gases in the reactor areas. Tritium in elemental form was not much of a concern in the reactor areas, and tritium vapor was the release of interest.

Uncertainty in the STM readings inherently had the uncertainty associated with collection of activity using a silica gel column and two Kanne chambers. Although it might appear at first glance that the uncertainties in the three methods might have canceled each other out, they instead became additive and increased the total uncertainty in the final value.

Additionally, the calibration of the STM was more difficult than simple calibration of a Kanne chamber. The increased hardware left extra room for error. The calibration of a Kanne chamber was discussed in a previous section of this appendix. This calibration uncertainty applied to a similar constant used in the STM calculations, called K_I . This constant had units of curies per cubic centimeter per ampere and became a factor in the series of calibration equations described below.

$$A = K_2 \cdot V \quad (\text{E-13})$$

$$K_2 = \frac{F \cdot C}{K_1} \quad (\text{E-14})$$

$$R = \frac{dA}{dt} = K_2 \cdot \frac{dV}{dt} \quad (\text{E-15})$$

where

- A = total integrated stack activity released (Ci)
- V = voltage across the capacitor, created by current through capacitor (V)
- F = flow rate of air out stack ($\text{cm}^3 \text{min}^{-1}$)
- C = capacitance (farads)
- capacitance (in farads) multiplied by voltage (in V) equals current (in A)
- K_I = chamber calibration ($\text{Ci cm}^{-3} \text{A}^{-1}$)
- R = rate of release of activity (Ci min^{-1}).

Since K_I in the above equations depended on the calibration of two Kanne chambers, it is reasonable to assume that uncertainty in K_I is greater than the uncertainty in the calibration of a single chamber. The measurement uncertainty for a single Kanne chamber, including chamber calibration, is +17% and -20% of the mean (see Kanne chamber section). This uncertainty factor is applied twice in the calculation of uncertainty for the STM by multiplying the total uncertainty by this factor squared.

The other source adding uncertainty to the calibration factor for the rate of release calculation (K_2) is the airflow rate, F . As before, we assigned a flow rate uncertainty of $\pm 20\%$ based on measurement uncertainty (see previous sections).

The silica gel has an uncertainty that is lognormally distributed with a GM = 1.15 and a GSD = 2.16. This uncertainty represents the efficiency with which the silica gel collects total water vapor in the airstream (see silica gel section).

A major source of uncertainty in Kanne chamber measurements is the inability of the chamber to distinguish between noble gas activity and tritium activity. That source of uncertainty is removed by this new experimental setup because subtracting the two Kanne results gives a value for only tritium vapor in the airstream if all sources of uncertainty in collecting activity are accounted for in the calculation.

Pressure fluctuations may still have been an important source of uncertainty because two ionization chambers had to remain under stable pressure throughout measurements. The pressure difference between the two chambers was zeroed before use of the instrument, but that does not imply that pressure remained constant. This source of uncertainty, identified as $\pm 10\%$ earlier, is included twice to account for possible fluctuations in both chambers.

The total uncertainty in STM releases is calculated using Monte Carlo techniques and 3000 trials. The results of the calculation fit a lognormal distribution with a GM = 0.91 and a GSD = 2.19.

A test of the STM system compared to the existing dehumidifier system was performed in 1965 when the STM was first introduced. The test monitored stack releases for 24-hour periods over 1 week. The measured results are shown in [Table E-11](#). The parenthetical values following the measured results are the 5 and 95% uncertainty limits of the values based on the uncertainty factors presented here.

Table E-11. Release Measurements and Uncertainty for a 1965 STM Versus Dehumidifier Test

Day	Release (Ci d ⁻¹)	
	Stack tritium monitor (5-95% uncertainty limits)	Dehumidifier (5-95% uncertainty limits)
Wednesday	218 (41-950)	336 (64-1800)
Thursday	221 (42-960)	372 (70-2000)
Friday	211 (40-920)	413 (78-2200)
Saturday	162 (31-710)	341 (64-1800)
Sunday	156 (30-680)	346 (65-1800)
Monday	152 (29-660)	321 (61-1700)
Tuesday	240 (46-1000)	390 (74-2100)
Wednesday	240 (46-1000)	320 (60-1700)

For all release days, the uncertainty bounds of the release values overlap, indicating that although the values appear different, they are statistically the same given what was known about the operation of each measurement system.

Berthold Tritium Monitor

The BTM was the most recent evolution of stack monitoring systems at the SRS, and the use of the gas proportional counting principle made it the most robust. In gas proportional counters, like ionization chambers, charged particles were produced by ionization in the gas. Unlike ionization chambers, radiation that interacted in a gas flow proportional counter produced ion pairs in a quantity proportional to the amount of energy it deposited. This allowed the proportional counter to distinguish between tritium oxide and elemental tritium, the forms of tritium released from SRS stacks. The counter not only distinguished between the two, but it also measured the activity of each isotope passing through, something no counter before this one could have done. Two single channel analyzers collected the two different forms of tritium.

General Operation

In the reactor areas, the tritium releases were still estimated by estimating the total amount of moderator lost and then converting back to total curies released. Available documentation referenced two equations that may have been used to calculate total moderator lost from the stack ([Westinghouse 1988](#)). However, [Westinghouse \(1988\)](#) identified one equation whose solution was transferred to the data sheet from which tritium loss was calculated (Equation [E-16]).

$$A = C \cdot IB \cdot \frac{F}{T} \quad (\text{E-16})$$

where

- A = daily stack loss (lb D₂O day⁻¹)
- C = conversion factor
- IB = daily integrated total counts in channel B, channel which collects tritium oxide ionization (counts)
- F = duct flow (cfm)
- T = tritium content of moderator from latest lab result (μCi tritium [mL D₂O]⁻¹).

The units of the conversion factor were not found in any documentation, but they were determined based on the calculation. The conversion factor presumably converts microcuries of tritium to counts, minutes to day, and milliliters D₂O to pounds D₂O, where 1 mL = 1 g water.

To convert from this value of pounds D₂O per day to curies of tritium per day, the following equation was required:

$$Q = A \cdot T \cdot 0.000413 \cdot D \quad (\text{E-17})$$

where

- Q = tritium released to atmosphere (Ci)
- A = daily stack loss (lb D₂O d⁻¹)
- T = tritium content of moderator from latest lab result (μCi tritium [mL D₂O]⁻¹)
- D = air density correction factor
- 0.000413 = lb d⁻¹ × μCi d⁻¹ to Ci d⁻¹.

The air density correction factor, D , used in Equation (E-17) is the same factor explained in the dehumidifier section, with uncertainty equal to $\pm 3\%$ of the mean.

Equations (E-16) and (E-17) were combined to remove a source of uncertainty, the tritium content of the moderator. Because this value was only measured once a week, it would have been a source of uncertainty for the stack loss calculation. The same value was used again in the conversion of the moderator loss data back into tritium release, effectively removing this source of uncertainty. Combining the equations produced Equation (E-18).

$$Q = C \cdot IB \cdot F \cdot 0.000413 \cdot D \quad (\text{E-18})$$

where all values are as described previously.

The uncertainty in the BTM tritium release values was centered around the stack flow rate; the accuracy of the BTM measurement (resulting in the IB value); and the conversion included in the factor C , which converted counts from the BTM to microcuries tritium. The uncertainty in D is so small as to become negligible in the uncertainty calculation.

Uncertainty in the stack flow rate has been described in all previous sections as $\pm 20\%$ of the mean, with a triangular distribution.

The conversion factor was related to the calibration of the BTM. Because calibration of this instrument was much easier than calibration of the STM or previous stack monitoring equipment, it was probably done more frequently. A small quantity of tritium in nitrogen calibration gas was injected directly into the BTM, and a long check sheet for calibration was established to ensure more accurate stack results. The sensitivity of the BTM was quoted in manufacturers' specifications as much lower than achievable in the field, with a [lower limit of detection](#) lower by a factor of 10.

This inconsistency was blamed on the uncertainty in stack flow rate and in moderator tritium content (Merz 1988). Calibration experiments showed that the BTM could measure tritium in the presence of a simulated noble gas to accuracy of less than 5 parts in 1000. Because moderator tritium content had been removed from the tritium release equation and uncertainty in stack flow rate was accounted for, the large inconsistency between predicted sensitivity and achieved sensitivity was ignored. The accuracy of the calibration experiments was noted (0.5%), but we assumed the calibration factor exhibited a small degree of uncertainty, subjectively estimated at $\pm 10\%$. During calibration, extreme measures appeared to have been taken to ensure accuracy. Calibration was highly controlled, but not so controlled as to eliminate operator error. A small uncertainty appeared appropriate in this case, however, because the technique was so finely tuned and carefully practiced.

The accuracy of the BTM readings was probably quite good, given operation in ideal conditions, but stack conditions were never ideal and a small degree of operational uncertainty was assumed to account for possible inconsistencies. Radioactive decay is a statistical process, and not all radiation that entered the chamber was likely to have been detected by the equipment. Still, these factors account for a relatively small uncertainty when compared to uncertainties inherent in the other measurement techniques. Operational uncertainty of the BTM was subjectively estimated at $\pm 10\%$

Total Uncertainty

Total uncertainty in the BTM measurements was calculated using Monte Carlo techniques, with 3000 generations of random numbers. The total uncertainty results were fit to a lognormal distribution with a GM = 0.99 and a GSD = 1.16. To estimate uncertainty in any source term value, the 5 and 95% bounds of the source term estimate can be obtained by multiplying and dividing the median value by the square of 1.16, respectively.

SUMMARY

The accuracy of the atmospheric source term estimates presented in this report is largely dependent on the type of monitors used over time to measure the releases. A detailed description of the equipment and the data used in the uncertainty analysis was presented in this appendix. In reviewing the documentation, it was possible to find dates on equipment operation procedures or reports of plans or prototype tests; however, it was not possible to put together an exact chronological history of monitoring equipment used in each facility. Therefore, there is no clear connection between a reported release value and monitor that was used to measure the release. For example, it has not been possible to determine when the silica gel monitor was replaced by the dehumidifier. Early documentation refers to both monitors.

No clear deficiency in the methodologies used brought us to the conclusion that there was need to apply corrections factors to the data to determine the final source term estimates. To help verify that this was acceptable, we used the results of the uncertainty to evaluate the effectiveness of the monitors. [Table E-12](#) presents a summary of the results of the uncertainty analyses.

Table E-12. Summary of Uncertainty Analysis for Tritium Monitors used at the 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
Kanne chamber	1954–1974	Tritium	Lognormal	GM = 0.9
Stack Monitor	1974–present	facilities		GSD = 1.8
Integrator				
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

^a GM = geometric mean, GSD = geometric standard deviation.

Because there were no other technical indications for the need to correct the estimate release data, we think that the results in the column titled “Range” show that the earlier methods, although less accurate, fall within the same range as the later, better methods with a larger uncertainty range. So, RAC concludes that the “best estimate” values should be used as the atmospheric tritium source term without correction, and the range of uncertainty should be applied to release data by facility and year as indicated in [Table E-12](#). If more information is found defining the time period of use of the monitors in each facility, these results can be applied more specifically.

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