CHAPTER 4.4

RELEASES OF ALPHA-EMITTING RADIONUCLIDES TO THE ATMOSPHERE

ABSTRACT

This chapter discusses releases of alpha-emitting radionuclides, primarily plutonium and uranium to the atmosphere from Savannah River Site (SRS) facilities. The primary objective of our efforts is to develop release estimates, or a source term, for alpha-emitting radionuclides that can be used to estimate potential exposure and risks to surrounding populations. We have attempted to validate reported releases by comparing data from as many separate SRS sources as possible. In general, reported release values are consistent among the various reports that we have reviewed. Additionally, we have estimated the uncertainty associated with the reported releases, based on sample collection and counting procedures, and estimated possible sample losses during transmission through sampling probes and lines. Annual atmospheric release values for plutonium and uranium and associated uncertainty have been compiled for both F-Area and H-Area stacks.

The data indicate that the majority of plutonium emissions from both F-Area and H-Area stacks occurred during 1955 and 1969. The data also indicate that the majority of uranium emissions from H-Area stacks occurred during 1955, 1968, and 1969, and that the majority of uranium emissions from F-Area stacks occurred during 1955, 1956, and throughout the 1960s. Evaluation of potential impacts resulting from alpha-emitting radionuclide releases should be focused on these years because the relative magnitude of total emissions during other years (including all years since 1970) appears to be quite small.

POTENTIAL RELEASE SOURCES

To assess atmospheric releases of alpha-emitting radionuclides and their potential impacts, it is important to understand the general processes that have historically been carried out at the SRS. This enables release data compilation and analysis to focus on those facilities with the greatest potential for releases as well as those time periods during which the largest releases occurred.

Carlton et al. (1993) and Evans et al. (1992) provide detailed description of the processes that may have led to historical plutonium and uranium emissions. The information in this section was taken primarily from these two references. Most SRS operations focused on the production of plutonium and tritium for national defense purposes. The production of plutonium initially involves the fabrication of fuel and target assemblies (in the M-Area facilities) for use in the five onsite production reactors (R-Reactor, C-Reactor, K-Reactor, P-Reactor, and L-Reactor). Plutonium and other radionuclides are formed in the fuel and target elements, which are reprocessed in one of two chemical separation facilities located in the F-Area and H-Area canyon buildings (221-F and 221-H). Various experiments and analyses with the potential to release alpha-emitting radionuclides have also been conducted in the Administrative Area (A-Area).

Administrative Area

Organizations that supply direct support for SRS operations, including the Department of Energy (DOE) office for the Site, the Savannah River Ecology Laboratory (SREL), administrative offices, the Savannah River Laboratory (SRL), which is now called the Savannah River Technology Center (SRTC), are all located in the Administrative Area, or A-Area (Cummins et al. 1990). The SRTC has historically been involved with analyses of fuel and target material and has dealt with various alpha-emitting radionuclides, including ^{238}Pu and isotopes of americium, curium, californium, and various other transuranics with high specific alpha activities (Evans et al. 1992). Releases from the SRTC are routed through a sand filter, and some of the ventilation streams pass through some combination of HEPA filters, charcoal filters, or caustic scrubbers before reaching the sand filter.

Fuel Fabrication

Most production of fuel and target assemblies took place onsite at the M-Area facilities and involved cladding of uranium with aluminum. Additionally, enriched uranium fuel rods were manufactured by first alloying the uranium with aluminum and were then machined and extruded. These processes have been associated with generating uranium metal filings and dust as well as dissolved and suspended uranium in solution.

Potential releases of uranium from M-Area facilities have included both atmospheric stack releases and liquid effluent releases to Tim's Branch and M-Area seepage basins. The majority of uranium releases from M-Area, however, occurred through liquid effluent releases to Tim's Branch. Chapter 5 addresses these releases in detail.

Reactor Operations

Fuel and target assemblies, heavy water moderator, and control rods comprise the major components of the reactor core. Following fabrication, fuel and target assemblies are transferred to the production reactors. Irradiated target and spent fuel elements are then removed from the reactor core and placed in the vertical tube storage basin of the reactor building. After a delay to reduce radioactivity levels, the elements are relocated to the disassembly basins and prepared for transfer to the chemical separation facilities. Under ideal conditions, all radionuclides would remain contained within the aluminum cladding of the fuel and target elements. However, during normal reactor operations, fuel and target elements can develop defects or ruptures in the aluminum cladding. Uranium, plutonium, other neutron capture products, and fission products have been released to the reactor moderator and basin water during such element failures. The reactor was shut down following a fuel or target element failure, and the failed elements were transferred to a harp container for storage in the reactor basin.

Potential releases of plutonium, uranium, and other alpha-emitting radionuclides were primarily through the discharge of liquid effluent. Atmospheric releases have likely been minimal because all fuel and target handling activities were accomplished under water. The primary mechanism for atmospheric release of radionuclides from the reactor building was resuspension of particulate material from dried liquids. Furthermore, atmospheric releases were minimized through the use of a confinement system that did not allow for air exchange with the gases

present inside the reactor tank, vertical tube storage basin, or disassembly basins. All ventilated air was passed through a demister filter bank, a high-efficiency particulate air (HEPA) filter bank, and a carbon filter bank.

Fuel Processing

From the reactors, irradiated target and spent fuel elements were transferred to the chemical separation facilities located in the F-Area and H-Area canyon buildings for reprocessing. Exhaust stacks from these facilities have historically had the highest measured atmospheric emissions of alpha-emitting radionuclides, primarily plutonium (^{238}Pu and ^{239,240}Pu) and uranium. Therefore, the majority of release data analyses focuses on these facilities.

Beginning in November 1954, target elements were treated by the Purex process to recover ^{239}Pu, ^{238}U, and ^{237}Np from irradiated ^{238}U in the F-Area canyon buildings. As part of the Purex process, target slugs were stripped of their aluminum cladding and dissolved in nitric acid. The ^{239}Pu, ^{238}U, and ^{237}Np were segregated into separate aqueous streams by solvent extraction and several stages of contactors and centrifugal separators. The plutonium, uranium, and neptunium were present in the process streams as nitrate salts in strong nitric acid solutions. During the precipitation and reduction processes carried out in the B-Line facilities, some particles containing metal oxides were formed. The primary routes for atmospheric releases were through the process ventilation vents and the aqueous stripping streams. All process vessels are vented through HEPA filters and large sand filters before being released through the main process 61-m stack. Air from other areas is directly vented to the sand filter only before discharge or sent through HEPA filters only before discharge. Offgas from the A-Line passes through bag filters before being discharged to the atmosphere via the 291-F stack (Evans et al. 1992 and Sanders 1977). In the early years of operation, exhaust gases from the B-Line facilities were vented only through HEPA filters before release through short stacks on the roof of the canyon buildings. However, the ventilation systems were modified to provide additional filtration following a fire at the Rocky Flats Plant (Carlton et al. 1993).

The Purex process was also used in the H-Area canyon building from July 1955 through 1959, after which the facility was modified to recover ^{235}U and ^{237}Np from enriched fuel elements. The H Modified (HM) process is different from the Purex process because it used mercuric nitrate as a catalyst to control dissolution of the uranium-aluminum alloy used to fabricate enriched fuel elements. The Frames process, which employs ion exchange instead of solvent extraction to separate and purify ^{238}Pu and ^{237}Np from waste products, was also used in the H-Area to recover ^{238}Pu from ^{237}Np targets (Carlton et al. 1993). Additionally, special campaigns for the production of radionuclides, such as ^{252}Cf and ^{244}Cm, have occasionally occurred. The atmospheric ventilation system in the H-Area facilities is essentially the same as that described for the F-Area facilities.

Other Sources of Alpha-Emitting Radionuclides

Much of the electric power distributed at the SRS was generated onsite at the power plants. These plants also provided steam for the Site. Historically, both electricity and steam at the SRS have been generated by burning coal, which in turn has resulted in the atmospheric release of signficant quantities of fly ash. Coal, and consequently the ash created by burning it, contains a

number of radionuclides, which originate from trace amounts of naturally occurring ^{238}U and ^{232}Th. Additionally, a number of other radionuclides result from decay of ^{238}U and ^{232}U, including a number of radioactive isotopes of radium, uranium, thorium, bismuth, polonium, and lead.

To make comparisons of releases associated with fly ash to releases associated with other Site operations, we can estimate the amounts of certain radionuclides that may have been released as a result of coal burning, based on the measured concentrations of radionuclides in fly ash and the amount of fly ash released (see Chapter 17 for additional details regarding coal burning and estimated airborne releases of fly ash). Eisenbud and Petrow (1964) reported concentrations of several radionuclides associated with coal burning, including ^{226}Ra, ^{228}Ra, ^{238}U, ^{228}Th, and ^{232}Th. Table 4.4-1 lists the concentrations of these radionuclides in ash reported by Eisenbud and Petrow (1964), along with estimated potential releases, based on the maximum estimated annual release of fly ash from the SRS (see Chapter 17).

Table 4.4-1. Radionuclide Concentrations in Fly Ash and Associated Maximum Airborne Emission Potential

Radionuclide	Concentration in Fly Ash[a] (pCi g^{-1})	Estimated Fly Ash Release[b] (g)	Calculated Radionuclide Release (Ci)
^{226}Ra	3.8	2.9×10^{10}	0.11
^{228}Ra	2.4	2.9×10^{10}	0.07
^{228}Th	2.6	2.9×10^{10}	0.08
^{232}Th	2.2	2.9×10^{10}	0.06
^{238}U	4.4	2.9×10^{10}	0.13

[a] Source: Eisenbud and Petrow (1964)
[b] Maximum annual fly ash release estimate (for 1988) from Chapter 17

These potential maximum release amounts suggest that coal burning may have released sufficient quantities of radionuclides to become important by comparison to F-Area and H-Area releases of plutonium and uranium, after approximately 1960, excluding 1969. If annual releases of plutonium and uranium on the order of 0.1 to 1 Ci are determined to be important contributors to potential risk to members of the public during future phases of this dose reconstruction, it may be necessary to further evaluate the potential importance of radionuclide releases associated with the burning of coal.

RELEASES AND RELEASE MONITORING

Administrative Area

Releases of alpha-emitting radionuclides have occurred at A-Area, but the releases are relatively small by comparison to the larger releases that of occurred at the Chemical Separation Area Facilities. The most notable releases occurred in the mid to late 1960s. Cummins et al. (1991) report 0.032 and 0.021 Ci releases of ^{244}Cm in 1964 and 1969, respectively. These releases are comparable to and generally less than total releases of alpha-emitting radionuclides associated

with fly ash generated by burning coal and are significantly less than the highest releases associated with the Chemical Separation Area Facilities.

M-Area Facilities

Isotopes of uranium were the only alpha-emitting radionuclides that had the potential to be released in significant quantities from M-Area facilities. From the first days of operation through 1974, activity release alarms were in place on building exhausts, but chronic atmospheric releases through stacks have only been measured since 1975. From 1975 through 1989, the highest recorded annual atmospheric release of uranium from M-Area facilities was 0.0001 Ci, or about 143 g (Evans et al. 1992), which is more than an order of magnitude less than the lowest recorded annual uranium release at the chemical separation facilities. Total reactor power levels during this time period (1975 through 1987) were approximately one-half the total power levels during peak production years (late 1950s and early 1960s). Fabrication of a larger number of fuel elements would have accompanied the higher power levels, so it is likely that atmospheric releases of uranium during peak production years would have been greater than releases measured since 1975. It is unlikely, however, that M-Area releases would have ever been great enough to appreciably add to chemical separation area releases.

Reactor Buildings

Because of the low potential for atmospheric radionuclide releases of any kind from the reactors, only gross beta and gamma activity was measured on air filters. Air filters were submitted for alpha analysis only if activity above background was detected. After the early 1970s, all filters were submitted for gross alpha analysis, but specific radiochemical analyses for plutonium or uranium were not made. Maximum annual alpha releases to the atmosphere, when they were quantified, were generally 2 or 3 orders of magnitude less than the lowest recorded annual plutonium and uranium releases from the chemical separation facilities. As with A-Area and M-Area releases, it is unlikely that reactor releases to the atmosphere would ever have been great enough to appreciably add to the chemical separation area releases.

Chemical Separation Area Facilities

Atmospheric releases of alpha-emitting radionuclides (including plutonium and uranium) from the separation facilities have been quantified since shortly after startup in November 1954. The primary release points at the F-Area and H-Area consisted of two tall ~61-m (200-ft) stacks, designated 291-F and 291-H, respectively. The first sampling period for plutonium and uranium in atmospheric effluents was December 14, 1954, to January 7, 1955 (Carlton et al. 1993). Continuous sampling was accomplished by passing a portion of stack effluent through a filter designed to trap particles and associated radionuclides. The filter was changed weekly and submitted for specific (including plutonium and uranium) radiochemical analysis. Carlton et al. (1993) indicated that plutonium emissions data for both F-Area and H-Area stacks were based on specific radiochemical analyses. Evans et al. (1992) indicated that uranium emissions data from F-Area were based on specific radiochemical analyses, and that emissions data from H-Area were based on gross alpha analyses. However, the original monthly reports through 1971 provide gross

alpha release estimates only, and there is no indication of routine specific isotopic analyses. It is possible that specific isotopic analyses were carried out only for quarterly or semi-annual filter composites and were therefore not reported on a monthly basis. However, the routine semi-annual and annual monitoring reports from 1955 through 1971 also reported only gross alpha release estimates.

The vast majority of alpha-emitting radionuclides released from the separation facilities likely consisted of plutonium and uranium. However, in 1967, approximately 28 mCi of ^{244}Cm were released between November 11 and 25, accounting for the majority of H-Area atmospheric releases during that month (Du Pont 1967k).

F-Area and H-Area Stacks

Total alpha emissions were reported monthly in 1955 and semiannually from 1956 through 1963 in the Health Physics Regional Monitoring report series and were reported semiannually from 1964 through 1966 and annually from 1967 through 1971 in the Environmental Monitoring at the Savannah River Plant report series. Annual plutonium (both ^{238}Pu and ^{239,240}Pu) and uranium emissions data were reported by Cummins et al. (1991). Carlton et al. (1993) and Evans et al. (1992) also provided emissions data for plutonium and uranium, respectively. However, a few of their release estimates were inconsistent with the data provided by Cummins et al. (1991), and the tabulated plutonium data reported by Carlton et al. (1993) appeared to represent only ^{239,240}Pu. All emissions data depicted in the following figures have been taken from the Health Physics Regional Monitoring report series (total alpha), the Environmental Monitoring at the Savannah River Plant report series (total alpha), and Cummins et al. (1991) (specific radionuclides).

Figures 4.4-1 and 4.4-2 (note the logarithmic scale) show the total alpha and plutonium plus uranium emissions data for 1955 through 1971 for F-Area and H-Area stacks, respectively. It appears that plutonium and uranium have comprised the majority of measured total alpha emissions from F-Area stacks during most years, particularly those years during which the most significant releases occurred (1955, 1956, and 1960). It also appears that plutonium and uranium have comprised the majority of measured total alpha emissions from H-Area stacks for the years during which the most significant releases occurred (1955 and 1969). For two years (1960 and 1967), however, total alpha releases from H-Area stacks were greater than the sum of plutonium and uranium releases. It is likely that other radionuclides were emitted during those years in sufficient quantities to account for a significant portion of the total alpha activity. For instance, as mentioned previously in this section, the majority of H-Area atmospheric releases during November 1967 consisted of ^{244}Cm, and other alpha-emitting radionuclides, such as ^{252}Cf, have occasionally been processed in the chemical separation areas.

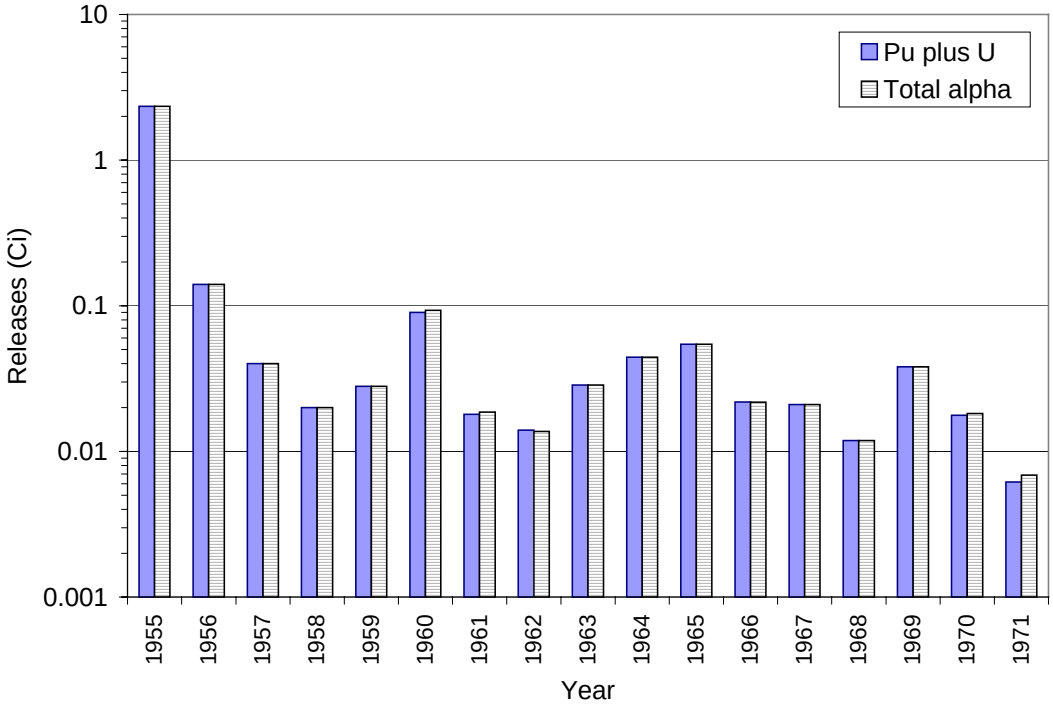

Figure 4.4-1. Total alpha and plutonium plus uranium emissions from F-Area stacks. Link to tabulated figure data.

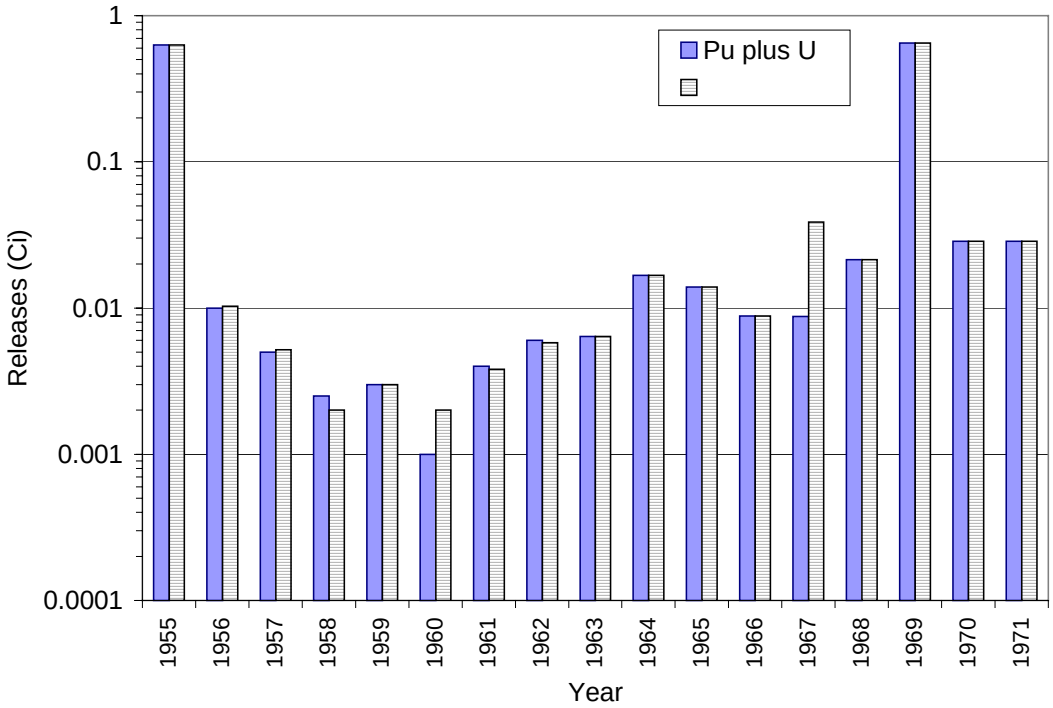

Figure 4.4-2. Total alpha and plutonium plus uranium emissions from H-Area stacks. Link to tabulated figure data.

Figure 4.4-3 shows the percent of total alpha releases by year from 1955 through 1971 for F-Area and H-Area stacks. It is clear that the majority (about 83%) of releases occurred during 1955 and 1969. More than 70% of the releases (predominantly ^{239,240}Pu) in 1955 occurred between August and December during a number of B-Line exhaust filter failures (Horton and Mealing 1956a). Backup filters were installed in both areas during December, reducing measured emissions significantly. In April 1969, breakage of supporting tile in the H-Area exhaust stack sand filter caused local depletion of sand and resulted in a significant alpha release, primarily ^{238}Pu (Ashley 1970a).

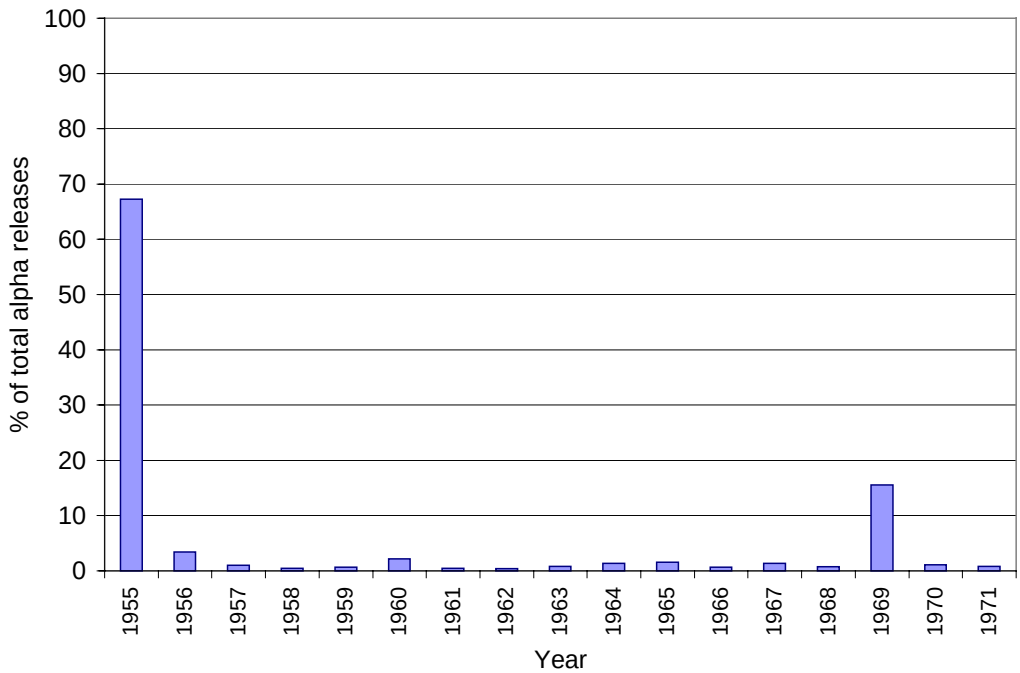

Figure 4.4-3. Percent of total alpha releases by year from F-Area and H-Area stacks from 1955 through 1971. Link to tabulated figure data.

Figures 4.4-4 (note the logarithmic scale) and 4.4-5 show the annual plutonium emissions and percent of total emissions by year, respectively, for F-Area and H-Area stacks (Cummins et al. 1991). Greater than 90% of the total plutonium emissions from separation area stacks between 1955 and 1989 occurred in 1955 and 1969 (specific incidents discussed previously).

Based on data provided by Cummins et al. (1991), distinctions were not made between ^{238}Pu and ^{239,240}Pu, and all plutonium emissions data were reported as ^{239,240}Pu before 1967. Beginning in 1967, ^{238}Pu emissions data were provided. Figure 4.4-6 shows the percent ^{239,240}Pu of total plutonium emissions (^{238}Pu plus ^{239,240}Pu) for F-Area and H-Area stacks from 1967 through 1989. It is clear that ^{238}Pu comprised a significant portion of total plutonium emissions during this time period, particularly from H-Area facilities, which were involved with ^{238}Pu recovery through the Frames process. In fact, reported plutonium emissions from H-Area stacks in 1977 and 1978 consisted entirely of ^{238}Pu. Totals of 2.46 and 1.26 Ci of plutonium were released from F-Area and H-Area stacks, respectively, between 1955 and 1989.

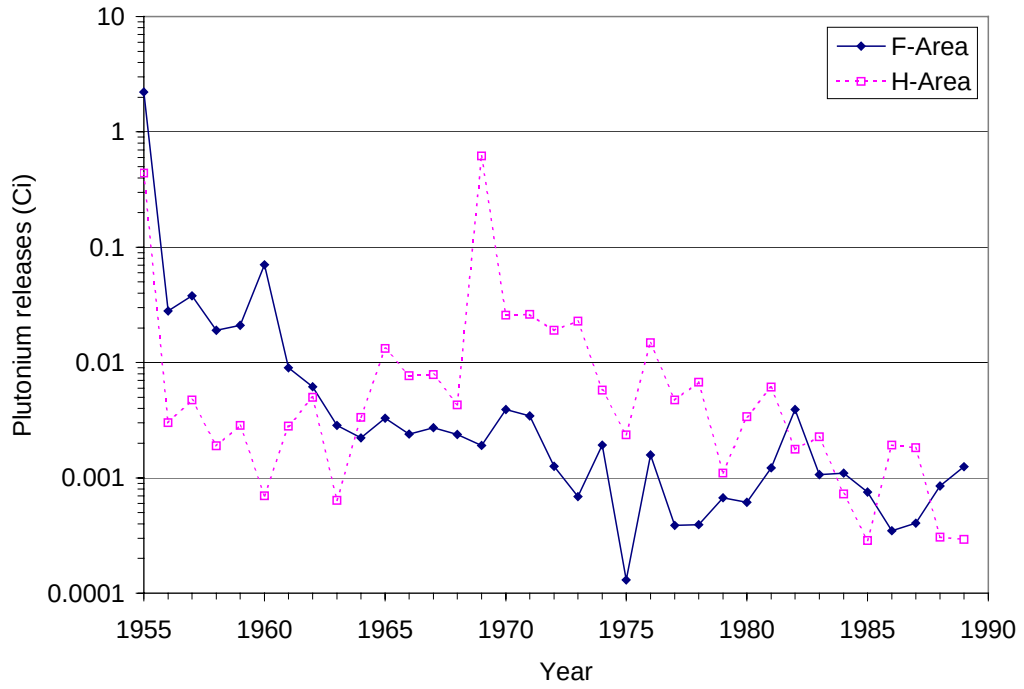

Figure 4.4-4. Annual plutonium releases from F-Area and H-Area stacks from 1955 through 1989. Link to tabulated figure data.

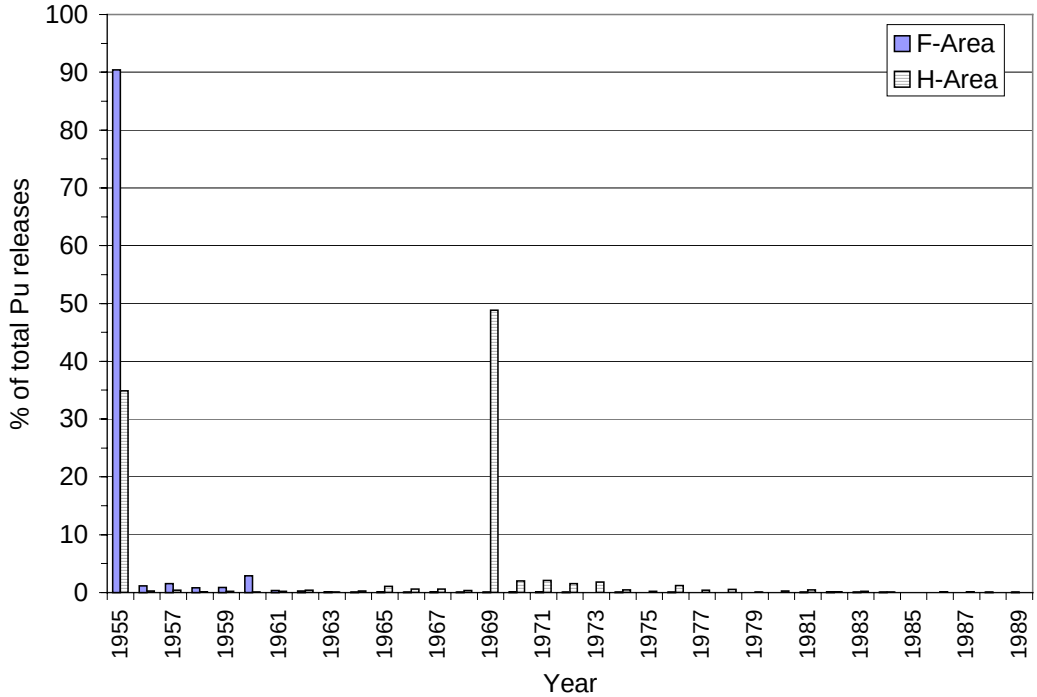

Figure 4.4-5. Percent of total plutonium released by year from F-Area and H-Area stacks from 1955 through 1989. Link to tabulated figure data.

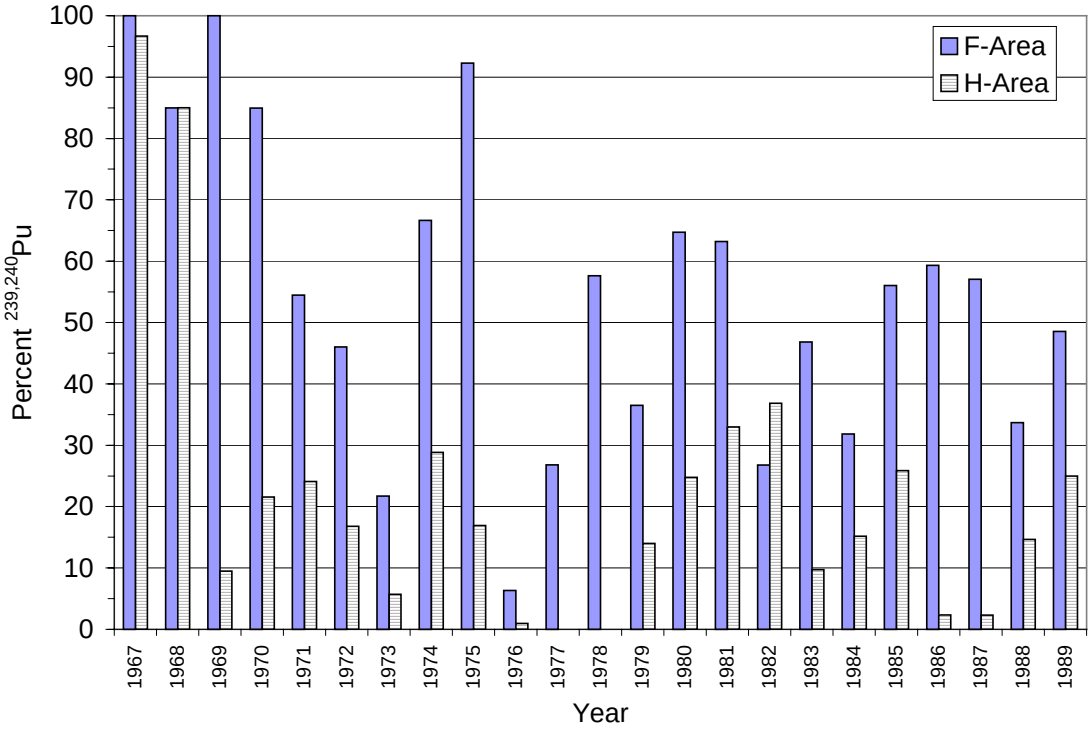

Figure 4.4-6. Percent of ^{239,240}Pu out of total plutonium emissions (^{238}Pu plus ^{239,240}Pu) for F-Area and H-Area stacks from 1967 through 1989. Link to tabulated figure data.

Figures 4.4-7 (note the logarithmic scale) and 4.4-8 show the annual uranium emissions and percent of total emissions by year, respectively, for F-Area and H-Area stacks (Cummins et al. 1991). The uranium emissions provided by Cummins et al. (1991) reportedly consisted of natural uranium, and include ^{238}U, ^{235}U, and ^{234}U. It is not clear why the emissions are indicated to consist only of natural uranium since both enriched and depleted uranium have been processed and recovered at SRS. If uranium emissions are determined to be important contributors to dose to members of the public, it may be necessary to more closely examine the specific isotopic composition of uranium emissions.

The majority of uranium emissions occurred during the first 2 years of operation (1955 and 1956) and during several years in the 1960s. Evans et al. (1992) indicated that greater emphasis was placed on reducing emissions since 1970; this is supported by both the plutonium and uranium emissions data. Totals of 0.57 and 0.29 Ci of uranium were released from F-Area and H-Area stacks, respectively, between 1955 and 1989.

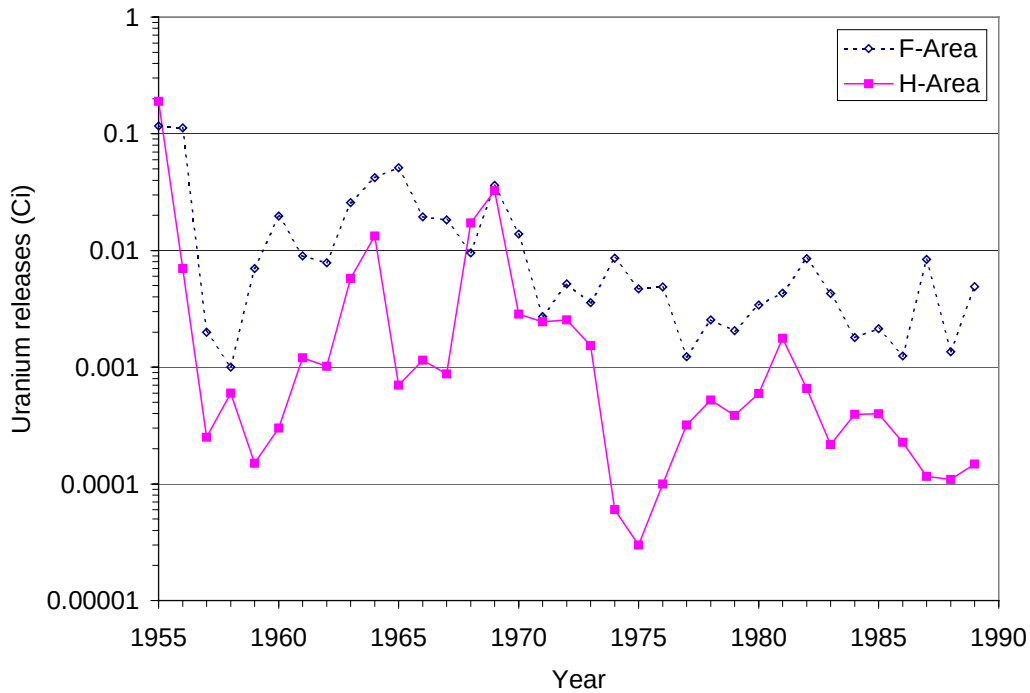

Figure 4.4-7. Annual uranium releases from F-Area and H-Area stacks from 1955 through 1989. Link to tabulated figure data.

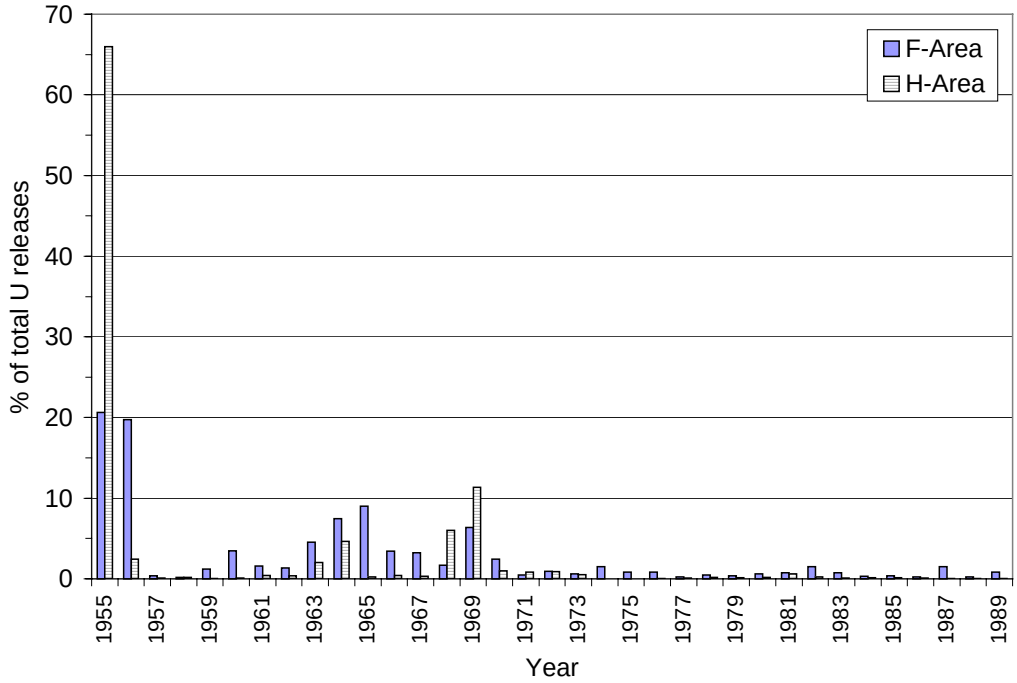

Figure 4.4-8. Percent of total uranium released by year from F-Area and H-Area stacks from 1955 through 1989. Link to tabulated figure data.

ACCOUNTING FOR SAMPLE LINE LOSSES

It is not apparent that Site-reported release quantities were modified to account for potential line loss or particle plateout along the inner walls of the sampling line, which would result in an underestimate of the amounts released. The ratio of the contaminant concentration leaving the sampling line to the concentration entering the line is often referred to as a transmission factor. We used a model developed by Texas A&M University to estimate transmission factors for alpha bearing particles sampled from the F-Area and H-Area effluent stacks (ATL 1993). These transmission factors can be used to modify reported releases to account for potential sample line losses.

A number of factors can influence the degree to which particles are transmitted through a sampling line. In general, larger and denser particles are most prone to deposition and loss along the length of the sampling line, particularly along horizontal sample line sections and by impaction in bends. Sample flow rate, effluent free stream velocity, and probe type also affect transmission through the sampling lines.

The effluent sampling lines for the reprocessing facilities at the SRS were quite long. Samples were extracted at the ~59-m (195-ft) level of the stack. The line extended to near ground level and then an additional 30.5 m (100 ft) horizontally to the point of sample collection. The total line length, including the in-stack portion, is estimated to be about 93 m (305 ft). This estimate is based on the following description: the sampling system consisted of a probe, a 0.15-m (6-in.) vertical tube, a 4.57-m (15-ft) horizontal tube, a 57.9-m (190-ft) vertical tube, and a 30.5-m (100-ft) horizontal tube. Each section of tubing was assumed to be connected by a 90-degree bend. It is possible that the actual sampling system was somewhat more complex and consisted of a greater number of bends and individual sections of tubing as described by Zippler (1979). However, the greatest amount of loss generally occurs in the probe and in the horizontal sections of tubing, the distances of which are known, so assuming an overly simple design does not significantly affect the calculation.

The flow rate through the sampling lines has varied somewhat and is assumed to be 2 cubic feet per minute (cfm) from December 1954 through October 1956, 3 cfm from November 1956 through September 1961, 2 cfm from October 1961 through September 1965, and 5 cfm after September 1965. See Chapter 4.2, Table 4.2-3 for more details regarding the sample flow rate during different time periods.

The free stream velocity refers to the rate or speed at which the effluent is moving through the stack and, therefore, the rate at which the sample enters the probe. Zippler (1979) measured the free stream velocity of the effluent in the 291-H stack and reported an average value of 2574 feet per minute (fpm) or approximately 13 m s^{-1}. This is consistent with the average flow for the 291 F stack (226,000 cfm) reported in Du Pont (1968a). Assuming a 10-ft stack diameter (Zippler 1979), this flow rate results in a free stream velocity of approximately 15 m s^{-1}. A flow rate of 13 m s^{-1} was assumed for the transmission factor calculations.

Several researchers have attempted to characterize the particle size distribution for the material released from the separations area stacks. Croley (1973) reported an average particle size of 1.5 microns (μm) (ranging from 0.5 to 4.5 μm) for samples collected during July 1973 from the 291 F and H stacks at the 195-ft level. Average particle sizes were determined by the light microscope technique, which does not account for the fact that alpha-emitting radionuclides are generally not uniformly distributed among different particle sizes.

Activity-based particle sizes are more appropriate for determining transmission factors. Such distributions are estimated by collecting effluent samples with a cascade impactor using several fractionation stages. The activity that is collected at each stage is determined, and an activity-based particle size distribution can be estimated.

Sanders (1978) collected samples from the 50-ft level of the 291-F stack in October 1975 and reported a geometric mean particle size of 5.43 µm with a geometric standard deviation of 2.69. Sanders (1977) described the procedures that were used to determine the particle sizes reported by Sanders (1978). Particles were selected for analysis based on the number of observed fission-fragment tracks, with those particles surrounded by many tracks being selected in favor of those surrounded by few tracks. This method of particle selection would appear to characterize the size distribution of particles carrying the majority of the plutonium.

Particle sizes as low as 0.4 µm were reported, but submicron particles accounted for only 7% of the total number of particles. The relatively large size of these particles was ascribed to coagulation of submicron particles by thermal and turbulent mechanisms to form larger particle agglomerates. The elemental composition of these larger particles showed them to be comprised primarily of crustal elements or dust in combination with very small amounts of plutonium. Sanders (1978) hypothesized that the larger dust particles did not pass through the HEPA filters, but instead they entered the exhaust system through leaks in the ducts, which would be undetected as long as the exhaust system remained under negative pressure compared to the atmosphere.

Carlson et al. (1983) reported activity-based particles sizes, or activity median aerodynamic diameters, for samples collected from the 50-ft level of the 291-F stack in 1982. Table 4.4-2 shows the average percentage of total activity measured on particles of several size intervals during four separate sampling tests. About two-thirds of the total activity is associated with submicron particles. This size distribution is consistent with distributions reported by Croley (1978) and by Gay and Watts (1981) for environmental samples collected from the vicinity of the 291-H stack in 1975, 1976, and 1977. On the other hand, this size distribution is not consistent with the data reported by Sanders (1978). The fact that the particle sizes determined by Sanders (1978) were not activity-based may help account for this inconsistency. It is also possible that leaks in the exhaust system had been repaired since the study by Sanders (1978), and less dust was entering the system in 1982, when Carlson et al. (1983) collected their samples.

Table 4.4-2. Percentage of Total Plutonium Activity Measured on Different Particle Sizes in Samples Collected from the 291 F Stack in 1982[a]

Particle diameter (microns)					
>8.2	8.2–3.5	3.5–2.1	2.1–1.0	1.0–0.5	<0.5
19.1%	6.6%	2.9%	4.2%	13.6%	53.4%

[a] Data from Carlson et al. (1983)

Selecting an appropriate particle size distribution is an important factor in establishing a credible transmission factor estimate. Based on the data *RAC* has been able to locate, the most appropriate particle size distribution is somewhat uncertain. The particle size distribution reported by Sanders (1978) is significantly larger than the distributions reported by Croley (1973) and Carlson et al. (1983). For this reason, we have calculated several transmission factors based on various particle size distributions and sample flow rates (Table 4.4-3). During the time period of

highest release amounts (i.e., before 1970), samples were collected using anisokinetic probes. For comparison, transmission factors for isokinetic probes, which collect samples with the same velocity in the sample line as in the effluent stream being sampled, are also shown. A particle density of 2.0 g cm^{-3} was assumed for the distributions reported by Sanders (1978) and Croley (1973), which is characteristic of the crustal elements that typically comprise dirt and dust. A particle density of 1.0 g cm^{-3} was assumed for the distribution reported by Carlson et al. (1983) because a unit density is inherent for an activity median aerodynamic diameter.

Table 4-4.3. Transmission Factors Calculated for Several Particle Size Distributions

Particle diameter	Sample flow rate (cfm)	Transmission factors (%) Anisokinetic	Transmission factors (%) Isokinetic
Sanders (1978)	2	23	26
GM[a] = 5.43 µm	3	26	30
GSD[b] = 2.69	5	29	37
Croley (1973)	2	71	76
GM = 1.5 µm	3	74	80
GSD = 1.5	5	78	86
Carlson et al. (1983)	2	69	71
(see Table 4.4-2 for	3	71	74
particle sizes)	5	73	77

[a] GM = geometric mean.
[b] GSD = geometric standard deviation.

Transmission factors calculated based on the particle sizes reported by Sanders (1978) are likely conservative estimates because the large particle size distribution results in a greater amount of deposition and particle loss as the effluent passes through the sampling line. Transmission factors calculated based on the particle sizes reported by Carlson et al. (1983) may be more accurate because the distributions were based on measured activity. Additionally, the particle sizes reported by Carlson et al. (1983) are generally consistent with the particle sizes reported by Croley (1973) and Gay and Watts (1981). However, the particle sizes reported by Sanders (1978) may be appropriate for estimating line losses for uranium-bearing particles. Voillequé et al. (1995) reported uranium to be associated with larger-sized particles, similar to the distribution reported by Sanders (1978). Additionally, assuming a larger particle size may be appropriate for estimating releases associated with filter breaks (such as those that occurred in 1955 and 1969), events which would have led to releases consisting of larger particles. Anonymous (date unknown) estimated a transmission factor of 65% using the PLATEOUT computer model and an assumed particle size of 1.5 µm, which is generally consistent with the transmission factors we calculated using the model developed by Texas A&M University and similar assumed particle sizes. In addition, Zippler (1979) states “Due to the complexity of our sample lines theoretically less than half of the particles with median diameters greater than two microns would ever reach the filter paper”, which is consistent with the transmission factors we calculated based on the larger particle size distribution reported by Sanders (1978).

Effluent samples from the 291-F and H stacks were extracted with anisokinetic probes through at least the 1970s, which may result in collecting samples that do not represent the stack effluent particle composition. Isokinetic probes, on the other hand, collect samples with the same

velocity in the sample line as in the effluent stream being sampled, and they are considered more appropriate for obtaining representative samples. Based on results obtained by running the model developed by Texas A&M University, anisokinetic probes result in a slightly lower total transmission factor than isokinetic probes as a result of lower transmission through the probe itself. Transmission through the sampling lines and bends is slightly higher, however, when an anisokinetic probe is assumed in the model, perhaps because of decreased turbulence.

The SRS became concerned about the possible difficulties associated with anisokinetic sampling and attempted to draw conclusions about the differences between anisokinetic and isokinetic sampling. Zippler (1979) compared the two sampling probe types and concluded that no significant advantage could be shown for the isokinetic sampling over the routine anisokinetic sampling. However, low levels (i.e., near the detection limit) of activity were collected for many of the samples, and higher measured concentrations of ^{103,106}Ru collected with the isokinetic probe indicated greater sampling by nearly a factor of 2 compared to the anisokinetic probe. For the purpose of estimating transmission factors, it is assumed that the Site collected samples using anisokinetic probes.

UNCERTAINTIES IN THE REPORTED RELEASES

Uncertainty analysis is important because it provides an estimate of the possible range of releases that is consistent with our knowledge of the individual parameters in the release estimate calculation. The uncertainty analysis combines the best scientific knowledge available about each parameter in a single calculation to provide a distribution of results that is realistic given what we know about each parameter.

At the chemical separation areas (F-Area and H-Area), the majority of alpha releases were confined to the 61-m (195-ft) stacks. Aliquots of effluent were taken from these stacks daily, analyzed by health physics area personnel for gross alpha activity measurements, and sent to the Environmental Monitoring Group for final analysis and determination of release quantities. Release quantities were calculated as described in Du Pont (1968a), Equation (4.4-1).

$$Q = \frac{(cpm)(CF)(F_{stack})}{(F_{sample})(E)(2.22x10^{12}\,dpm/Ci)}$$

where

Q	=	stack release (Ci)
cpm	=	counts per minute
CF	=	conversion factor to convert cpm to dpm (disintegrations per minute)
F_{stack}	=	stack effluent flow rate ($ft^3\ min^{-1}$)
F_{sample}	=	sampler flow rate ($ft^3\ min^{-1}$)
E	=	collection efficiency of particulate air filter.

The total uncertainty in the release quantities (Q) that were determined is dependent upon the uncertainty associated with each of these factors. Inherent in the measured counts per minute are additional factors including counter error and percent recovery of extracted radionuclide. Total uncertainty resulting from counter error is likely small (5% or less), particularly during periods of the highest releases, because the filters would have accumulated significant activity

during a continuously sampled 24-hour period, and counter error decreases as a function of increasing count rate. This is assumed to be a negligible source of uncertainty. The uncertainty in the various extraction processes that were used, including ether and ethyl acetate and tri-butyl phosphate extractions, is likely around 20% based on average recovery values provided by Geiger (1954). A triangular distribution is assumed.

Uncertainty in the conversion factor that was used to convert counts per minute to disintegrations per minute is dependent upon the efficiency of the detector. These conversion factors were based on an alpha standard with a ¼-in. diameter active area (Johnson 1977). There may be some error associated with the different geometries of the standard and sample planchets. However, the 2-in. diameter zinc-sulfide crystal used for alpha counting was slightly larger than the 47-mm filters that were counted, and the samples were placed less than 5 mm from the active surface of the crystal (Johnson 1998). It is expected that this would contribute a negligible source of error.

Stack flow rates for the F-Area and H-Area 61-m stacks were determined from monthly Power Department flow measurements of the sand filter discharge plus fan capacities of contributing streams that do not go through the sand filter (Zeigler 1986). The uncertainty for reactor stack flow rates was reported as ±20% (Du Pont 1965a). The uncertainty in F-Area and H-Area stack flow rates is assumed to be the same, and a triangular distribution is assumed for this source of error.

Uncertainty in the sampler flow rate is dependent upon the type of particulate filter that was used. Inherent uncertainty in the calibration of the moto-air sampler flow rate was reported as ±10% (Du Pont 1968b). However, from 1955 through at least February 1957, Whatman #41 particulate filters were used for effluent sampling. Flow rates for these filters were found to vary as much as 50% when operating at a fixed pressure differential (Hoy 1957). In 1957, a change was made to MSA 1106-B filter papers, which are more uniform and do not show significant flow rate variations. Therefore, from 1955 through 1957, the uncertainty in the sampler flow rate is assumed to be ±60%. In subsequent years, uncertainty is assumed to be ±10%. A triangular distribution is assumed.

Uncertainty in the collection efficiency of the filter paper is dependent upon the type of filter paper that is used. An assumed collection efficiency of 80% was used for all calculations (Du Pont 1968c). However, MSA 1106-B filter paper efficiency is essentially 100%, and assuming 80% efficiency would have overestimated releases. On the other hand, when Whatman #41 filters were used, the efficiency of collection varied with the face velocity across the filter, and the efficiency was found to vary as much as 25% when operating at a fixed pressure differential (Hoy 1957). Therefore, from 1955 through 1957, the uncertainty in the collection efficiency is assumed to be ±25%. In subsequent years, uncertainty is assumed to be a negligible source of error because the efficiency of MSA 1106-B filters appears to have been underestimated by assuming an efficiency of 80%. It is not entirely clear what types of filters may have been used after 1957. Therefore, we are not making an adjustment to correct for this potential underestimation. A triangular distribution is assumed for collection efficiency uncertainty between 1955 and 1957.

We used the Crystal Ball® uncertainty analysis software package Version 4.0c (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. The possible range of release estimates is then calculated using a Monte Carlo analysis, which involves multiple trial calculations, using randomly selected values from the possible range of parameter values. In a

single trial, each of the parameter distributions is sampled, and the selected values are used to compute an estimate of the release. This procedure is repeated many times in a Monte Carlo analysis, and all the release estimates are saved and displayed 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.

Many distributions of environmental data uncertainty are lognormal and are best represented by a geometric mean and a geometric standard deviation. The parameters calculated for the alpha release uncertainty are generic uncertainty parameters that can be applied to any release value during the appropriate time period. To determine the range of possible values for a given release value, the median value would be represented by the release estimate multiplied by the geometric mean. The range of values within 1 standard deviation of the mean is defined by multiplying and dividing the mean value by the geometric standard deviation. The 5th and 95th percentile values are obtained by dividing and multiplying the mean value by the square of the geometric standard deviation.

For alpha releases from 1955–1957, the geometric mean is 1.08 and the geometric standard deviation is 1.64. For alpha releases after 1957, the geometric mean is 0.98 and the geometric standard deviation is 1.21.

CONSISTENCY IN REPORTED DATA

Total plutonium and uranium emissions reported by Cummins et al. (1991), Carlton et al. (1993), and Evans et al. (1992) are consistent with total alpha emissions reported in the Health Physics Regional Monitoring report series (Alexander and Horton 1956; Horton and Mealing 1956a, 1956b; Mealing 1957; Mealing and Horton 1957; Mealing et al. 1958; Harvey et al. 1959a, 1959b; Du Pont 1959a, 1960a, 1960b, 1961a, 1962a, 1962b, 1963a, 1963b, 1964a) and the Environmental Monitoring at the Savannah River Plant report series (Ashley 1965b, 1966b, 1967b, 1968, 1969b, 1970a, 1971, 1972). Data provided by Cummins et al. (1991) were compiled from air monitoring results gathered from both routine and special monitoring at effluent sampling locations by the Environmental Monitoring Section of the Environmental Protection Department at the SRS. The reported alpha emissions are also consistent with data provided in annual SRP Waste Audit reports (Ashley 1960, 1962a, 1962b, 1963, 1965a, 1966a, 1967a, 1969a, and 1970b), and specific radionuclide emissions are consistent with data provided by Ashley et al. (1982). Total alpha release data compiled from original monthly monitoring reports from 1954 through 1956, 1959 through 1965, and 1967 through 1970 (Du Pont 1954a–l, 1955a–l, 1956a–l, 1959b–m, 1960c–n, 1961b–m, 1962c–n, 1963c–n, 1964b–m, 1965b–m, 1967a–l, 1968d–o, 1969a–l, 1970a–l) are also consistent with the data provided by the above sources. We have attempted to locate original hand-written release data compilations, but no original data have been located that could be used to further verify the reported emissions.

SUMMARY OF ALPHA RELEASES

Plutonium and uranium annual release values for F-Area and H-Area between 1955 and 1989 reported by Cummins et al. (1991) are compiled in a Microsoft Excel® workbook (Estimated_source_term.xls). Median values and 5th and 95th percentile values are calculated for

both F-Area and H-Area, based on the uncertainties discussed above. We have assumed that line-loss corrections were not made to reported release values and that only anisokinetic probes have been used. Therefore, we have also applied calculated transmission factors (Table 4.4-3) to all reported plutonium and uranium releases.

For plutonium releases occuring in 1955 (F-Area and H-Area) and 1969 (H-Area only) and all uranium releases, we have used the transmission factors calculated assuming the particle size distribution provided by Sanders (1978). This results in estimated releases approximately a factor of four greater than Site-reported releases values. Zippler (1979) states "Due to the complexity of our sample lines theoretically less than half of the particles with median diameters greater than two microns would ever reach the filter paper", which is generally consistent with the transmission factors we calculated based on the larger particle size distribution reported by Sanders (1978).

For plutonium releases during all years except 1955 and 1969 (H-Area only), we have used the transmission factors calculated assuming the particle size distribution provided by Carlson et al. (1983). This results in values approximately 35% to 45% greater than the Site-reported release values. Anonymous (date unknown) estimated a transmission factor of 65% using the PLATEOUT computer model and an assumed particle size of 1.5 µm, which is generally consistent with the transmission factors we calculated using the model developed by Texas A&M University and similar assumed particle sizes.

These estimates may exaggerate actual releases if the released material is not appropriately characterized by the assumed size distribution data provided by Sanders (1978) and Carlson et al. (1983). However, we feel it is important to conservatively estimate potential releases, particularly because filter breaks, which have resulted in the majority of alpha-emitting radionuclide releases, may have resulted in releases consisting of larger-sized particles. Additionally, Voillequé et al. (1995) reported uranium releases to be associated with larger-sized particles, similar to the distribution reported by Sanders (1978).

Figures 4.4-9 and 4.4-10 show the annual median release estimates we calculated for plutonium and uranium, respectively. The error bars represent the 5th and 95th percentile values, which were calculated based on the uncertainties and transmission factors discussed above.

The data indicate that the majority of plutonium emissions from both F-Area and H-Area stacks occurred during 1955 and 1969, primarily as a result of known filter break events. The data also indicate that the majority of uranium emissions from H-Area stacks occurred during 1955, 1968, and 1969, and that the majority of uranium emissions from F-Area stacks occurred during 1955, 1956, and throughout the 1960s. Evaluation of potential impacts resulting from alpha-emitting radionuclide releases should be focused on these years because the relative magnitude of total emissions during other years (including all years since 1970) appears to be quite small.

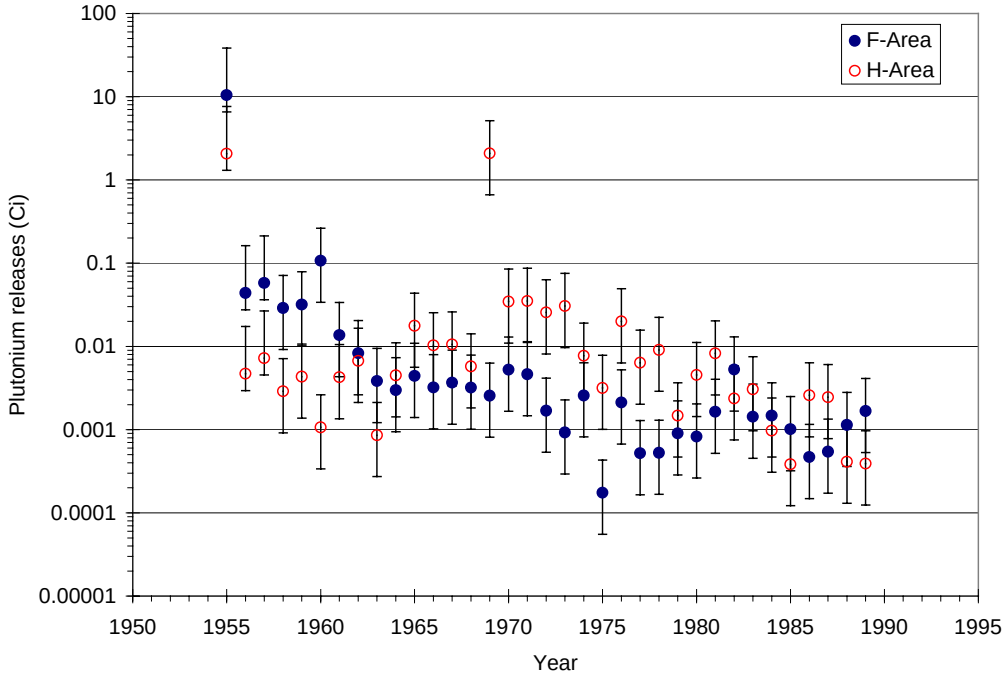

Figure 4.4-9. Annual median plutonium, including ^{238}Pu and ^{239,240}Pu, release estimates for F-Area and H-Area. The upper and lower error bars represent the 95th and 5th percentile values, respectively. Link to tabulated figure data.

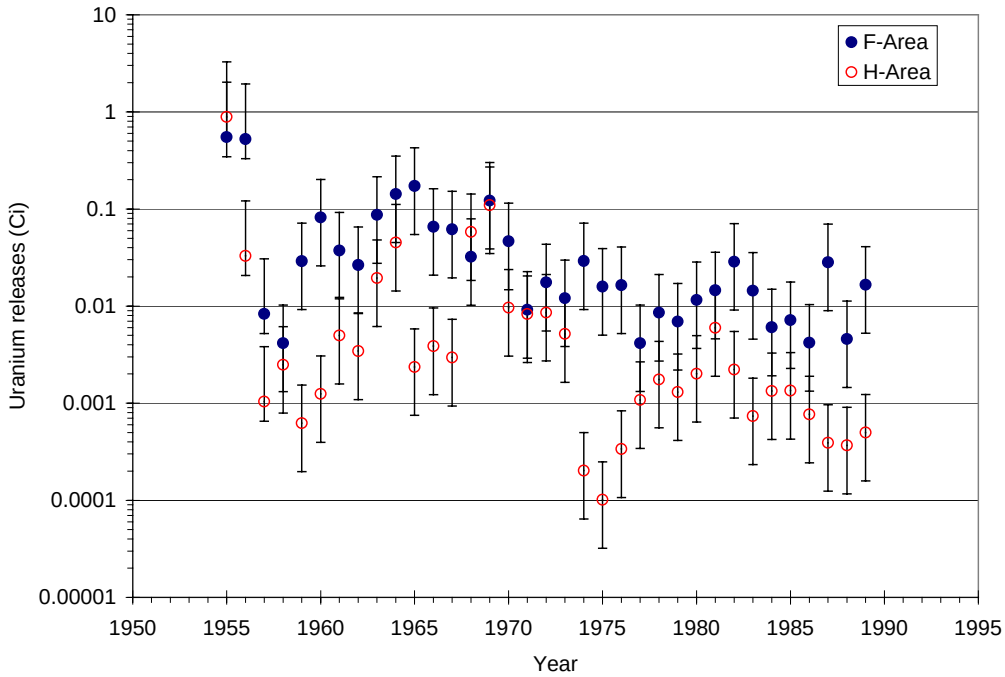

Figure 4.4-10. Annual median uranium release estimates for F-Area and H-Area. The upper and lower error bars represent the 95th and 5th percentile values, respectively. Link to tabulated figure data.

It should also be noted that emissions, including a number of alpha-emitting radionuclides, associated with the burning of coal may approach and even exceed the F-Area and H-Area plutonium and uranium emissions for many years, particularly after 1960 (see Table 4.4-1). Based on the relative importance of plutonium releases (to be determined during future phases of this project), particularly those occurring during 1955 and 1969, it may be necessary to more closely evaluate the potential health consequences associated with radionuclides present in airborne fly ash.

VALIDATION OF RELEASE ESTIMATES

A detailed validation of release estimates goes beyond the scope of work for this phase of the dose reconstruction project. However, because of several reviewer comments related to the appropriateness of sampling line deposition corrections made to Site-reported release estimates, we have included this discussion to further address the adequacy of our release estimates. Heffner (1999) offered lines of reasoning to support Site-reported release values by suggesting that modeled concentrations of Site-reported release estimates for 1955, the year of the highest plutonium release, show good correlation with measured gross alpha air concentrations during that year. It is asserted that either sampling line losses were insignificant or that some other process, such as large particle size, reduced the offsite impact of the release. It is not indicated what particle size was assumed for the modeling estimates made by the Site, but the larger particle size distribution we assumed for the transmission factor calculated for plutonium releases in 1955 certainly would result in increased deposition and, consequently, could similarly limit impact at the plant perimeter locations.

Based on soil (Chapter 12.2), air (Chapter 8), and vegetation (Chapter 9) monitoring data, it is not apparent that releases of alpha-emitting radionuclides have significantly impacted concentrations in media at plant perimeter or offsite locations. However, it is clear that these releases have impacted concentrations measured at onsite locations, near the F-Area and H-Area. We maintain that our estimated releases of plutonium and uranium, which take into account potential line losses, are conservative and appropriate, based on the events that led to the releases and on reported particle sizes. It is quite possible that modeling of our higher release estimate for 1955, taking into account the assumed particle sizes, will produce results that still correlate with measured air concentrations at the plant perimeter because of increased deposition of larger particles at onsite locations. At any rate, additional modeling of alpha-emitting radionuclide release estimates should necessarily include some amount of calibration to achieve reasonable correlation with gross alpha concentrations measured in air and vegetation samples.

ELECTRONICALLY COMPILED ALPHA RELEASE DATA

The data summarized in this section are electronically compiled in two Microsoft Excel® workbooks. One workbook (Ch4-4-Figure_data.xls) contains the figures provided in this chapter as well as the tabulated data that were used to produce the figures. In this workbook, there is a separate worksheet for each figure and one worksheet that contains the tabulated data for all of the figures.

The second workbook (Ch4-4-All_data.xls) contains the data that have been tabulated from various environmental monitoring reports and release summary documents. The workbook

contains several named worksheets, each containing a brief summary of the data compiled in the worksheet.

Table 4.4-4 summarizes the data that have been electronically compiled for alpha releases from the SRS. Additionally, the names of the individual spreadsheets in which these data are compiled (including a brief description of the data) are provided.

Table 4.4-4. Description of Data Electronically Compiled for Alpha Releases

Workbook name	Worksheet name	Brief description of data
Ch4-4-Figure_data.xls	Figures 4.4.1 through 4.4.10	Each worksheet contains a separate figure depicted in this chapter
	Data for figures	This worksheet contains the tabulated data for each of the figures
Ch4-4-All_data.xls	Total alpha	Total alpha releases reported in Health Physics Regional Monitoring report series
	Total alpha (2)	Comparison of total alpha releases to plutonium releases reported by Carlton et al. (1993) and uranium releases reported by Evans et al. (1992)
	SRP audits	Data compiled from annual Audit of SRP Radioactive Waste reports
	Plutonium releases	Plutonium release data compiled from Cummins et al. (1991) and Carlton et al. (1993)
	Uranium releases	Uranium release data compiled from Cummins et al. (1991) and Evans et al. (1992)
	Monthly reports	Total alpha monthly release data compiled from monthly monitoring reports
	Comparison of data	Comparison of data reported by different sources

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