CHAPTER 4.3

PRODUCTION AND ATMOSPHERIC RELEASE OF ACTIVATION PRODUCTS

ABSTRACT

The primary activation product of interest in terms of airborne release and potential offsite dose is ^{41}Ar. Even though it is a short-lived radionuclide, ^{41}Ar is a noble gas readily released from the reactor stacks, and most has not decayed by the time it moves offsite with normal wind speeds. SRS reactor operations produced and released relatively large quantities of ^{41}Ar, and its production rate in the air blanket surrounding a reactor should have been roughly proportional to the reactor power level. Cummins et al. (1991) provides an SRS-developed estimate of ^{41}Ar releases, which we compare to other measurements, check against reactor power levels, and accept as a generally reasonable estimate of SRS ^{41}Ar releases.

While these values represent the best available estimates for ^{41}Ar releases from the SRS reactors, the values presented for the later years (1974–1988) are quite low when compared to reactor power levels and overall average ^{41}Ar production levels. We also observe that the ^{41}Ar release values presented for certain of the early years (1955–1967) are quite high when compared to reactor power levels for the same period. The reason for these apparent discrepancies is not clear, and adds to the uncertainty in our estimates of ^{41}Ar releases.

INTRODUCTION

Most of the radioactivity produced by the five SRS production reactors involved fission products, created when ^{235}U, ^{239}Pu, or ^{233}U split into two or more smaller atoms. In addition, neutrons captured by some materials inside the reactor created radioactive isotopes called activation products. Some activation products were created outside the fuel or target protective cladding, and thus were more readily released to the outside environment than fission products produced inside intact fuel elements. Of these, ^{41}Ar was quite readily released to the environment, and must be evaluated in a dose reconstruction for its potential to expose people living nearby.

The Health Physics and Radiological Health Handbook lists some 300 target/product pairs in its neutron activation database (Shleien 1992). It is neither feasible nor necessary to consider all possible activation products in this study; we limit this research to consideration of those materials with any significant potential for dose to individuals living offsite. *RAC*'s approach to identifying these key radionuclides involves screening for such characteristics as quantity. Chapter 3 of this report provides details concerning our screening of radioactive materials encountered at the SRS, to identify those of greatest potential significance. We reproduce some of that screening logic below, to demonstrate the approach used to focus our activation product research.

THE SCREENING PROCESS

We began the screening process during Phase I of the study, and published a report in June 1995 identifying key radionuclides in the context of dose reconstruction (Meyer et al. 1995).

For air, the more important contributors emerging from the Phase I screening were ^{241}Am, ^{14}C, ^{137}Cs, tritium, ^{129}I, ^{131}I, ^{238}Pu, ^{239,240}Pu, ^{103}Ru, ^{106}Ru, ^{89}Sr, ^{90}Sr, uranium, and the noble gases including ^{41}Ar. For surface water, the more important dose contributors were ^{241}Am, ^{137}Cs, ^{60}Co, tritium, ^{131}I, ^{238}Pu, ^{239,240}Pu, ^{89}Sr, ^{90}Sr, ^{35}S, ^{99}Tc, ^{65}Zn, and Zr,^{95}Nb.

During Phase II of the project, we performed a more detailed assessment of release estimates to provide a more realistic ranking of the radionuclides that were identified in the Phase I, cautious screening methodology. The assessment was based on a methodology recently published by the National Council on Radiation Protection and Measurements (NCRP 1996). We looked at the relative importance of each radionuclide with respect to pathway of exposure. Chapter 3 of this report summarizes the results of that assessment.

For surface water pathways, ^{137}Cs dominates all pathways except the ingestion of drinking water, where tritium is most important. Strontium-90 is also relatively important as a contributor to dose via the surface water pathways when considering the ingestion of vegetables and meat as the major exposure pathways. Phosphorus-32 is important with respect to the ingestion of fish, while ^{131}I should be considered when the ingestion of drinking water and meat are evaluated.

For releases of radionuclides to air from the SRS, Chapter 3 shows that tritium and iodine are the major contributors to radiation dose from all airborne pathways except plume immersion. Argon-41 contributes almost 100% of the dose to a person who may be standing in the plume when it is released from the facility. Plutonium-238 and uranium are also relatively important contributors to dose via the inhalation pathway, contributing about 20% and 5% of the dose, respectively. Plutonium-239,240, a minor contributor to dose, is responsible for less than 5% of the dose from eating vegetables. Nevertheless, because it does meet our criteria of contributing greater than 1% to the dose, a more detailed source term reconstruction was undertaken.

We therefore focus our attention on activation products with potential for significant offsite human exposure: ^{32}P, ^{35}S, ^{41}Ar, ^{51}Cr, ^{60}Co and ^{65}Zn. We exclude radionuclides with an estimated total dose contribution of less than 1% via either the water or air pathway.

ACTIVATION PRODUCT CHARACTERISTICS AND RELEASE POTENTIAL

The radionuclides considered here were created in the SRS reactors by neutron capture. Their half-lives range from 1.8 hours (^{41}Ar) to 5.3 years (^{60}Co). Table 4.3-1 lists characteristics of these radionuclides. The radionuclide ^{41}Ar is a noble (nonreactive) gas produced in the air surrounding the reactor. Because this air volume communicated with the air in the reactor building proper, much of the ^{41}Ar produced in a reactor was released with ventilation air exiting the reactor stack.

Potential for Activation Product Releases at the Production Reactors

The largest releases of activation products occurred during the 1960s, and consisted primarily of ^{41}Ar from the reactors. The five reactors primarily responsible for ^{41}Ar gaseous releases were located on a rough circle, about halfway between the Site center and the boundary. As discussed in Chapter 2 of this report, the primary role of the production reactors was to produce plutonium and tritium for weapons development. Neutrons produced during fission interacted with fuel, targets and structural materials, and with chemicals in the moderator, creating activation products but releasing relatively small quantities of most to the environment.

This was because the products were largely contained within the materials in which they were created. Such activation products, if contained in solid materials including the fuel or metal reactor structure, did have the potential for fractional release later when fuel rods and targets were dissolved in the reprocessing canyons. Activation products produced within the liquid moderator chemicals, or within fuel elements which subsequently developed leaks, had a higher release potential, especially during the early years when the reactor fuel cooling basins were routinely purged as a maintenance procedure. Chapter 5 of this report considers activation product releases to surface waters, and the reader is referred to that chapter for a detailed discussion of such releases.

Table 4.3-1. Characteristics of Activation Products

Radionuclide	Primarily produced from	Half-life	Beta energy (MeV) max	Gamma energy (MeV)
^{32}P	^{31}P	14.3 d	1.7	None
^{35}S	^{35}Cl	87.3 d	0.17 (100%)	None
^{41}Ar	^{40}Ar	1.83 h	1.2 (99%)	1.29 (99%)
^{51}Cr	^{50}Cr	27.7 d	Electron capture	0.32 (9%)
^{60}Co	^{59}Co	5.27 y	0.31	1.17 (100%) 1.33 (100%) 0.51 (3.4%)
^{65}Zn	^{64}Zn	244 d	Electron capture	1.12 (49%)

[a] Source: Health Physics Handbook (Shleien 1992).

At least one activation product was produced in such a way that it was rapidly released to the outside environment. Neutrons interacting with the natural noble gas ^{40}Ar in the air around the reactor vessel created ^{41}Ar, which quickly mixed with reactor building air and was released via the airflow through the stack. Once out the stack, reactor gases moved with local air flows offsite, and individuals living nearby were potentially exposed to beta and gamma radiation emitted during ^{41}Ar decay. Because the half-life of ^{41}Ar is relatively short, radiation exposures calculated for this radionuclide were highest near the site boundary, and decreased more rapidly than for most other radionuclides with distance from the site. Argon-41 is the principal activation product of interest in terms of atmospheric releases, and when considering offsite human health risk.

The reactors began operating in the years 1953–1954, and were all operating by 1955. They were routinely shut down to replace fuel and targets and occasionally when unplanned events occurred. Test reactors, spontaneous fission, and some materials delivered from outside the SRS also contributed small quantities of activation products to the SRS inventory.

Potential for Release of Activation Products in Areas Other Than the Reactors

As mentioned above, under ideal conditions, most activation products produced within reactor fuel and targets would remain contained within the cladding until reprocessing occurred. Some leakage inevitably occurred, however, allowing activation products to move into the moderator circulating around the fuel, or to leak into the fuel cooling and disassembly basins. In addition, some materials circulating in the moderator were activated during neutron exposure. Under normal conditions, a significant fraction of the activation products in the moderator adhered to the fuel and targets; these materials were transported to the disassembly basin when the reactor fuel was unloaded. During cooling time in the basin, dissolution and corrosion allowed a fraction of the activation products to escape into the basin water. Air and water at the reactors were monitored for such possible releases.

Starting in the 1960s, basin water was passed through ion exchange resins to remove most radionuclides. Resins were either reworked to capture and concentrate the radionuclides for disposal or buried in the Burial Grounds (renamed the Solid Waste Disposal Facility in 1990). Chapter 5 considers these processes in more detail. During cooling in the basins, short-lived radionuclides decayed away.

After cooling, fuel and targets were treated in the chemical separations areas (the canyons). During these chemical separations, small quantities of some activation products volatilized during the evaporation of aqueous wastes; some material was discharged to seepage basins. In addition, small quantities of activation products moved through H-Area process cooling water because of heating and cooling coil leakage. Most of this activity was collected in retention basins and sent to seepage basins, but some leakage did occur into Four Mile Branch (Four Mile Creek) (Carlton and Denham 1996).

Chemical separations process aqueous wastes were evaporated and sent to the large waste storage tanks in F-Area and H-Area. Condensate from the evaporators was sent to seepage basins until 1988, when the Effluent Treatment Facility began to be used to remove radionuclides and chemicals from such wastes. Treated wastes from the new facility were discharged into Upper Three Runs Creek. High-level liquid wastes were stored for decay in temporary tanks, then shipped to F-Area for processing through the waste system. Solid wastes were buried in the Solid Waste Disposal Facility.

According to Carlton and Denham (1996), “Most of the atmospheric and aqueous effluents in the chemical separations and SRTC have been monitored for possible activation product releases...Activation product activity released through stacks was small enough that offsite transport through the atmosphere has not been detected.”

Activation Products Release Data

The SRS has in recent years published a number of documents summarizing radioactive releases for the primary operating history of the Site (1954–1989). The Carlton and Denham (1996) report discussed above presents annual data by release point (reactors, reprocessing areas, and certain other areas onsite) for the activation products ^{32}P, ^{51}Cr, ^{60}Co and ^{65}Zn. While these summaries are of interest in the examination of releases by location onsite, an earlier report prepared, Cummins et al. (1991), summarizes releases for the same period by pathway and year and for all six activation products of interest here. For the purposes of dose reconstruction, we choose to extract annual release data from Cummins et al. (1991).

Table 4.3-2 and Figure 4.3-1 present annual data for the six activation products we considered. To display the data more effectively, the ^{41}Ar values were divided by 100 before presentation in Figure 4.3-1.

Table 4.3-2. Activation Products Released from the SRS Per Year, All Sources, by Pathway (Ci)[a]

Year	^{32}P – Seepage basins	^{32}P – Streams	^{35}S – Seepage basins	^{35}S – Streams	^{41}Ar – air[b]	^{51}Cr – Seepage basins	^{51}Cr – Streams	^{60}Co – Seepage basins	^{60}Co – Streams	^{60}Co – Air	^{65}Zn – Seepage basins	^{65}Zn – Streams
55					5.0×10^{4}				6.0×10^{-2}			
55					1.4×10^{5}				2.2×10^{-1}			
56					2.5×10^{5}				6.1×10^{-1}			
57					2.9×10^{5}			2.4	1.6			
58					3.4×10^{5}			9.1×10^{-1}	1.1			
59					4.4×10^{5}			4.7×10^{-1}	2.7			
60					4.0×10^{5}		1.1×10^{1}	2.7×10^{-1}	7.8			4.3
61				6.6×10^{1}	4.2×10^{5}		6.2×10^{1}	1.6×10^{-1}	6.3			2.1×10^{1}
62				5.1×10^{2}	4.3×10^{5}		2.1×10^{2}	8.1×10^{-2}	1.3×10^{1}			3.2
63				1.6×10^{2}	4.5×10^{5}		1.3×10^{3}	7.5×10^{-1}	5.1			3.3×10^{1}
64		3.1		9.1×10^{1}	3.7×10^{5}	6.1×10^{1}	1.1×10^{3}	2.7	3.1		4.0×10^{-2}	2.0×10^{1}
65	3.7×10^{1}	1.4×10^{1}		8.6×10^{1}	2.7×10^{5}	1.0×10^{2}	7.8×10^{2}	1.6	1.1×10^{1}		3.3×10^{-1}	8.9
66	1.1×10^{2}	1.0×10^{1}		1.2×10^{2}	2.8×10^{5}	7.6	6.3×10^{2}	2.7×10^{-1}	5.1		1.0×10^{-2}	7.5
67	9.0	4.1		1.5×10^{2}	3.2×10^{5}	2.5	2.4×10^{2}	2.5×10^{-1}	1.7		1.2×10^{-1}	7.3
68		2.4	1.9×10^{2}	1.7×10^{2}	2.2×10^{5}	1.2×10^{2}	8.6×10^{1}	1.9	1.8	3.9×10^{-2}	3.8	4.5
69		7.3×10^{-1}	7.7×10^{1}	6.4×10^{1}	1.4×10^{5}	3.0×10^{2}	6.9×10^{1}	1.4	2.1×10^{-1}	1.1×10^{-2}	5.6×10^{-1}	1.5
70		7.7×10^{-1}	1.8×10^{1}	2.6×10^{1}	1.1×10^{5}	8.6	1.6×10^{1}	4.9×10^{-1}	1.7×10^{-1}	2.8×10^{-3}		5.1×10^{-1}
71		9.2×10^{-1}	3.1	1.1×10^{1}	1.4×10^{5}	2.5×10^{1}	7.3	4.7×10^{-1}	4.6	2.2×10^{-2}	3.0×10^{-2}	1.8
72		3.0×10^{-1}	9.7×10^{-1}	1.5	1.7×10^{5}	3.1×10^{1}	1.4	4.0×10^{-2}	1.8×10^{-1}	7.2×10^{-3}		
73	1.8×10^{-2}	1.3×10^{-1}	1.7×10^{-1}	1.7	1.8×10^{5}	1.4×10^{1}	4.5×10^{-1}	4.3×10^{-2}	2.7×10^{-2}	1.8×10^{-3}		
74	2.2×10^{-2}	1.4×10^{-2}	7.3×10^{-1}	5.0	1.1×10^{5}	7.9	6.0×10^{-1}	4.9×10^{-1}	1.0×10^{-3}	3.9×10^{-3}	7.6×10^{-1}	1.4×10^{-1}
75	8.0×10^{-3}	2.0×10^{-3}	5.1×10^{-2}	2.9×10^{-1}	6.5×10^{4}	4.6	1.2×10^{-1}	4.2×10^{-1}	9.0×10^{-3}	1.1×10^{-3}	5.7×10^{-1}	2.0×10^{-3}
76	9.0×10^{-3}	1.7×10^{-2}	3.1×10^{-1}	3.4×10^{-1}	8.3×10^{4}	6.4	3.6×10^{-1}	4.7×10^{-1}	2.0×10^{-3}	1.3×10^{-4}	4.4×10^{-1}	
77	2.2×10^{-2}	6.4×10^{-3}	5.1×10^{-2}	6.1×10^{-2}	6.5×10^{4}	6.1	6.6×10^{-1}	4.0×10^{-1}	1.0×10^{-1}	3.8×10^{-4}	5.3×10^{-1}	
78	3.6×10^{-3}	2.7×10^{-5}	1.3×10^{-2}	1.5×10^{-2}	5.3×10^{4}	1.5	4.0×10^{-2}	1.7×10^{-1}	3.9×10^{-4}	3.8×10^{-4}	5.4×10^{-2}	
79	2.0×10^{-3}	4.0×10^{-3}	1.9×10^{-2}	1.1	5.3×10^{4}	1.3	5.4×10^{-1}	1.0×10^{-1}	4.1×10^{-1}	4.0×10^{-3}	8.0×10^{-2}	
80	1.9×10^{-3}		1.1×10^{-2}		7.0×10^{4}	1.5		2.2×10^{-1}	1.6×10^{-3}	6.2×10^{-4}	1.4×10^{-1}	
81	3.7×10^{-3}		7.4×10^{-2}		6.2×10^{4}	2.4		1.6×10^{-1}	2.5×10^{-4}	8.9×10^{-5}	7.4×10^{-2}	
82	6.9×10^{-3}		1.8×10^{-1}		6.0×10^{4}	2.9		2.6×10^{-1}	1.1×10^{-4}	4.4×10^{-5}	9.2×10^{-2}	
83	1.6×10^{-2}		1.0×10^{-1}		4.1×10^{4}	1.2		2.2×10^{-1}	1.9×10^{-3}	1.7×10^{-4}	9.5×10^{-2}	
84	3.0×10^{-3}		1.5×10^{-2}		3.6×10^{4}	5.9		2.7×10^{-1}	3.1×10^{-4}	5.4×10^{-5}	1.0×10^{-2}	
85	6.1×10^{-4}		4.3×10^{-2}		5.2×10^{4}	3.8		2.9×10^{-1}			3.0×10^{-3}	
86	8.5×10^{-5}		2.7×10^{-2}		8.3×10^{4}	5.5×10^{-1}		1.3×10^{-1}		8.0×10^{-6}	5.2×10^{-2}	
87	1.3×10^{-5}		2.3×10^{-2}		8.8×10^{4}	9.8×10^{-2}		1.5×10^{-2}			2.1×10^{-2}	
88	9.8×10^{-6}		2.4×10^{-3}		3.0×10^{4}	2.7×10^{-3}		8.1×10^{-4}		3.0×10^{-6}	4.7×10^{-3}	
89	2.1×10^{-4}		2.1×10^{-4}									

[a] Source: Cummins et al. (1991).

[b] ^{41}Ar values before 1971 are estimates per Cummins et al (1991).

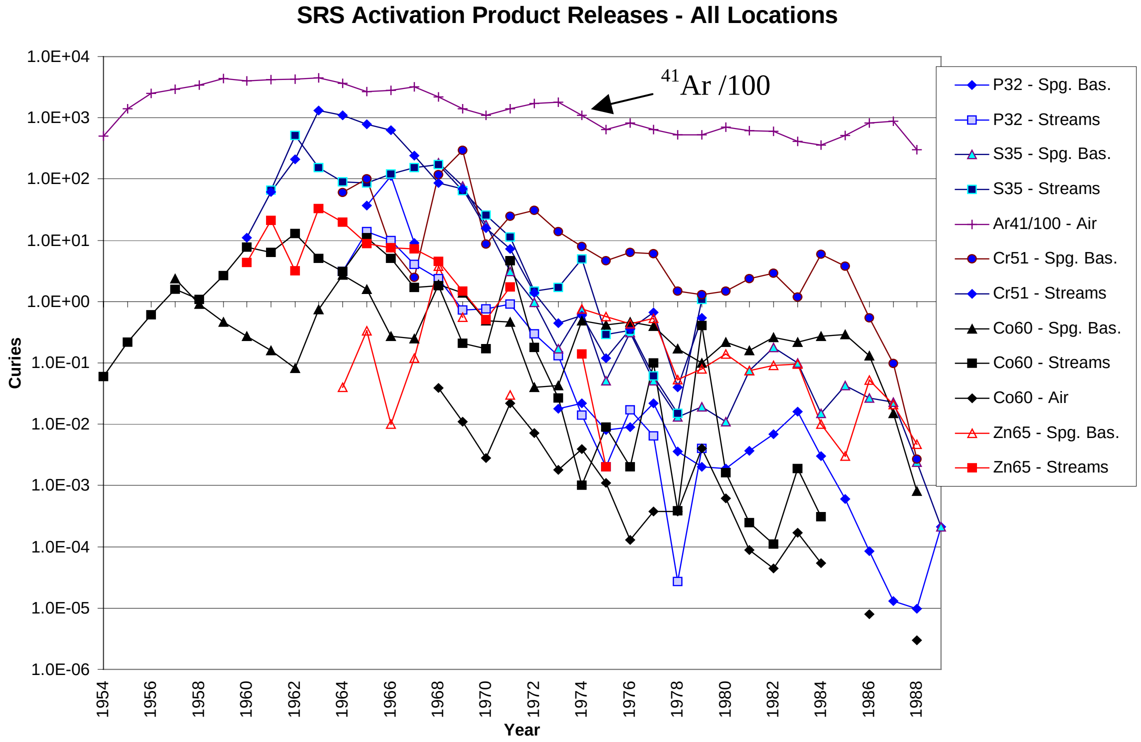

Figure 4.3-1. Activation products released from the SRS per year, all sources, by pathway (Cummins et al. 1991).

SUMMARIES OF OTHER AVAILABLE DATA

We searched the *Radiological Assessments Corporation* (*RAC)* SRS Dose Reconstruction Phase I document database for records with information related to the activation products. Most of the documents recovered are brief reports of specific events and do not lend themselves to developing a clear pattern related to activation product releases over the entire history of the Site. They primarily record offnormal incidents at the SRS, when specific activation products were noticed or anticipated in an effluent and a study was undertaken to determine the radionuclide's source or actual release rate. Because ^{32}P, ^{35}S, and ^{51}Cr have relatively short half-lives, there are few observations of these nuclides in effluents. The nuclides ^{65}Zn and ^{60}Co, with longer half-lives, were more often measured in effluent and in offsite media. Although ^{41}Ar has a very short half-life, it was created in relatively large quantities routinely during reactor operation. As a noble (nonreactive) gas, it was released much more readily to the environment with the stack air stream. Data and estimates discovered during the Phase I document review and summarized here are more complete than for the other activation products.

The Phase I records concerning these nuclides make up a rather sparse set and do not support detailed, comprehensive estimates of SRS releases or dose calculations. However, they do focus on important events associated with activation product releases dating back to early 1956. These events coincide with peak releases seen in recent radionuclide release summaries published by the SRS (Cummins et al. 1991; Carlton and Denham 1996) and, thus, may be used to corroborate key releases and release periods. We reviewed these records and summarized them below, listed by primary document date (some of the records are actually record groups, containing memos written on a specific topic over several years). Where data are presented, the summaries below contain excerpted tables of those data and graphics where useful. These tables represent the primary data sets for these data fragments in this report -- no spreadsheets were prepared to support them. Similarly, the data graphs presented below are the primary data sets for these data fragments -- no spreadsheet analyses were performed or are available to support them.

Early Discussions Concerning Potential Increases in ^{41}Ar Releases

A note, Hall and Coombs (1956), states that the stack discharges of “A^{41}” were on the order of 1500 Ci per month per reactor area. The monthly stack discharge ranged from 570 Ci in C-Area to 1840 Ci in R-Area. The note states that, “Assuming the present stack activity to be the average activity, the stack discharge ranges from 860 to 2300 Ci per month, with the exception of L area which is shut down.” Hall and Coombs note that the ^{41}Ar activity of the stack effluent is almost insignificant while the reactor is shut down and increases greatly during and following startup. The note references an attached table summarizing the results, but the table is missing from the copy recovered by *RAC*.

A memo, Hoy (1962), notes that design modifications being considered for the reactors may cause increased ^{41}Ar releases to the atmosphere. The Health Physics group estimated that 1500 Ci d^{-1} released per reactor would cause dose rates as high as 2.4 mrem h^{-1} at 0.8 mi (well inside the Site boundary). Maximum offsite dose, with overlapping plumes from all five reactors, would not exceed 0.0022 mrem h^{-1}. Hoy (1962) noted that the stack Kanne chambers, low-level beta-gamma counters, and area background detectors might be affected. The memo noted that “the normal gamma background level in the vicinity of SRP is 0.35 mr/24 hr . . ..”

A note to file, Coombs (1964), indicates that an anticipated 50-fold increase in the ^{41}Ar release rate, due to ventilation of the K-Reactor with dry air vs. CO_2 beginning in June 1964 did not occur. Kanne chamber readings indicated an ^{41}Ar release of 85 Ci d^{-1}, three times higher than the previous K-Area discharge rate. Exposure rates at the K-Area fence (using a Geiger-Mueller tube and scaler) ranged from 0.34 to 0.49 mrem d^{-1}, “within the normal background range” but highest at the predicted ^{41}Ar maximum concentration. Coombs (1964) speculates that a high percent of air may have already been in the reactor cavities from leakage. Calculations, based on the 50-fold predicted ^{41}Ar increase, had indicated a potential for 2.4 mrem h^{-1} at 0.8 mi downwind. An increase in ^{14}C emission was also predicted. Coombs (1964) notes an estimated discharge of ^{14}C of 0.84 Ci y^{-1} at 30–65 ft^3 min^{-1} purge of K-Area reactor cavities at 10% reactor down time.

Stream Releases

A two-page document fragment, titled “Reactor Area Release Summary” (Anonymous ca. 1964), summarizes amounts of radioactive material released to streams from 1954 through 1964. From 1954 through 1960, radionuclides were characterized as “primarily short-lived materials,” associated with the discharge from the reactors of failed fuel elements. The fragment notes “Discontinued routine purge of deionized water,” and “75% Np,” in association with the 1959 data. The data related to (or likely related to) activation products are summarized below in Table

4.3-3. Table 4.3-4 provides an indication as to the specific radionuclides probably making up the "primarily short-lived materials" source term in the first table. No further information was available to support this speculation.

Table 4.3-3. Releases to Streams

Year	Releases to streams (Ci)
1954	30
1955	112
1956	303
1957	875
1958	537
1959	1323
1960	836

For 1960, Anonymous ca. 1964 notes that "Specific radioanalysis (was) initiated last half of year," but it does not provide the results of the analyses. For 1961–1964, five to six radionuclides are considered individually in author (year). Only activation products are listed in Table 4.3-3.

Table 4.3-4. Activation Products Released to SRS Streams

Year	Total curies to streams	Specific radionuclides analyzed	Curies to streams for the specific radionuclides analyzed
1961	360	^{51}Cr	62
1962	1142	^{51}Cr	199
		^{35}S	512
1963	2594	^{51}Cr	1280
		^{35}S	156
1964	1725	^{51}Cr	1085
		^{35}S	91

The document contains a remarks section, noting changes in processes leading to changes in release rates. Better chemistry control and less corrosion, plus discontinued purging of the thermal shield system, reduced releases in 1960.

Johnson (1962) states that ^{35}S, the main contributor of the long-lived materials, was likely due to degradation of the cation (sulfonated) resin as a result of radiation damage. Johnson (1962) also states that "Cation resin beds were replaced with mixed beds in the moderator purification system to facilitate better Pd control; ^{35}S releases decreased as a result." A 1964 note states that, "The failure to sorb the ^{35}S was attributed to saturation of the anion resin." Also noted was a six-fold increase in the release of ^{51}Cr at that time because of a change in chemistry increasing the retention of chromium by fuel assemblies.

A table titled, "Savannah River Plant. Radioactivity in Reactor Effluent Waters–CY 1968," (Anonymous 1969) tabulates combined reactor effluents for 17 radionuclides including the activation products listed in Table 4.3-5.

Table 4.3-5. Radioactivity in SRS Effluent Waters, All Reactors Combined

Activation product	Combined reactor effluents (Ci released)	Contribution in Savannah River downstream at Highway 301 bridge (pCi L^{-1})
^{51}Cr	85.8	<8
^{32}P	2.4	NA
^{65}Zn	4.5	<1.1
^{60}Co	1.8	<1.4
^{35}S	172	26

Other Data

A memo (Patterson 1969) that discusses ^{60}Co releases notes that a ^{60}Co release was "first" reported in September 1967 and that, "To date only a gaseous form of ^{60}Co has been released from the High Level Caves short stacks, from the 'B' stack, and from the 776-A short stack." Particulate cobalt activity had not penetrated the high efficiency filters. The High Level Caves Off Gas Exhaust system was the source of ^{60}Co in "B" stack." The memo notes particulate ^{60}Co problems in ducts and filters; with the ducts of Cell 12 radiating 1 to 2 R h^{-1} due to internally deposited ^{60}Co. High-efficiency filters in the Cell 12 system had shown dose rates up to 10 R h^{-1} through the filter housing after ^{60}Co manipulations in the cell. Table 4.3-6, and Figure 4.3-2 summarize onsite and offsite ^{60}Co measurements in air for a 7-month period in 1968 and 1969 when activities involving cutting materials containing ^{60}Co were occurring. A partial summary of these same data is also presented in a memo, (Taylor 1969a).

According to Patterson (1969), since September 1967 there had been three periods of significant ^{60}Co release:

1. September–November 1967: 8.8 mCi
2. February 8–March 15 1968: 33 mCi
3. January 9–March 20, 1969: 3.2 mCi.

Patterson (1969) notes that the Environmental Monitoring group collected vegetation samples in the Technical Division area after the first two release periods, with very low concentrations detected. The report notes that ^{60}Co was released from the 776-A short stack when cutting solution was discharged to the high-level drain after ^{60}Co specimens were cut in the metallurgy cell. Several tables, including several handwritten pages, are attached to the report. *RAC* has compared the handwritten (Patterson 1969) and summarized values (above) for the three periods noted and found them to be essentially identical. A note indicates that the SRS ultralow level counting laboratory had just been completed, enabling the detection via coincidence counting of very low levels of ^{60}Co (to 0.5 disintegrations min^{-1}, which equals 227 fCi or 227×10^{-15} Ci).

Table 4.3-6. Onsite and Offsite SRS Monitoring for ^{60}Co (Ci/m^3 × 10^{-15})[a]

Month	Hi Vol.[b]	Uncrt.[b]	735-S[b]	Uncrt.	200F&H	Uncrt.	OP[b]	Uncrt.	25 mile[b]	Uncrt.	100 mile[b]	Uncrt.
Dec-68			0.29	0.25	0.11	0.06	0.04	0.02	0.071	0.01	0.062	.010
Jan-69	0.09	0.008	1.34	0.65	0.11	0.05	0.1	0.02	0.05	0.01	0.048	.010
Feb-69	0.038	0.01	9.53	1.46	0.34	0.06	0.116	0.01	0.052	0.01	0.166	.020
Mar-69			7.88	1.45	0.19	0.06	0.133	0.02	0.08	0.01	0.042	.010
Apr-69			1.93	0.52	0.11	0.03	0.178	0.02	0.075	0.01	0.064	.020
May-69			4.81	0.87	0.144	0.1	0.095	0.02	0.135	0.03	0.049	.020
Jun-69			13.06	0.51	0.512		0.529	0.04				

[a] Source: Patterson (1969).

[b] HiVol - high volume air sampler. Uncrt. - Uncertainty. Building 735-S. Buildings 200-F and H. OP - Outer SRS Perimeter. 25 Mile radius from SRS. 100 Mile radius from SRS.

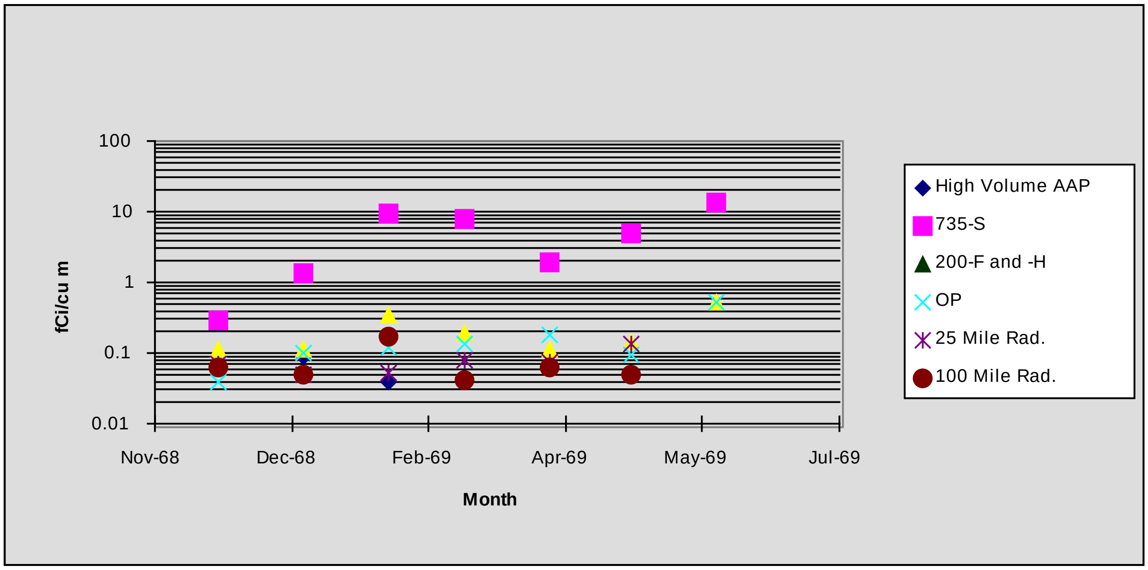

Figure 4.3-2. Onsite and offsite SRS monitoring for ^{60}Co (Patterson 1969).

The following data were compiled from two memos (Taylor 1969a, 1969b) attached to Patterson (1969) after *RAC* evaluation and correction of an apparent error in Taylor (1969b). The correction was based on comparisons to contemporary SRS data, data overlap between the two memos, and a note in Taylor (1969b) indicating a detection limit, using ^{60}Co coincidence counting that was a factor of 1000 lower than the lowest ^{60}Co values presented in the subject memo. The data in Table 4.3-7 and Figure 4.3-3 reflect the factor-of-1000 correction where indicated.

Table 4.3-7. Cobalt-60 in Air—Aiken Airport and Onsite

Date	Sample (m^3)	^{60}Co Ci m^{-3} [× 10^{-15}])	+/–
1/30/68	36,833	.017[b]	.001
10/17/68	178,715	.035[b]	.003
1/3/69	106,040	.038[b]	.004
1/9/69	36,589	.065[b]	.009
1/14/69	35,768	.140[b]	.012
1/15/69	35,149	.103[b]	.010
1/17/69	106,030	<.003[b]	
1/24/69	104,309	.097[b]	.006
1/31/69	109,679	.189[b]	.008
2/7/69	106,073	.073[b]	.005
2/11/69		.034	.010
2/13/69		.065	.010
2/15/69		.030	.010
2/18/69		.042	.020
2/22/69		.012	.003
2/25/69		<.010	
Onsite:			
1/1–31/69[c]	45,851	.590[b]	.021

[a] Source: Extracted from Taylor (1969a and 1969b).

[b] Data corrected for apparent error in the SRS source memo; these values have been reduced by a factor of 1000.

[c] Onsite air sampler, Administration Area (SRS 700 Area).

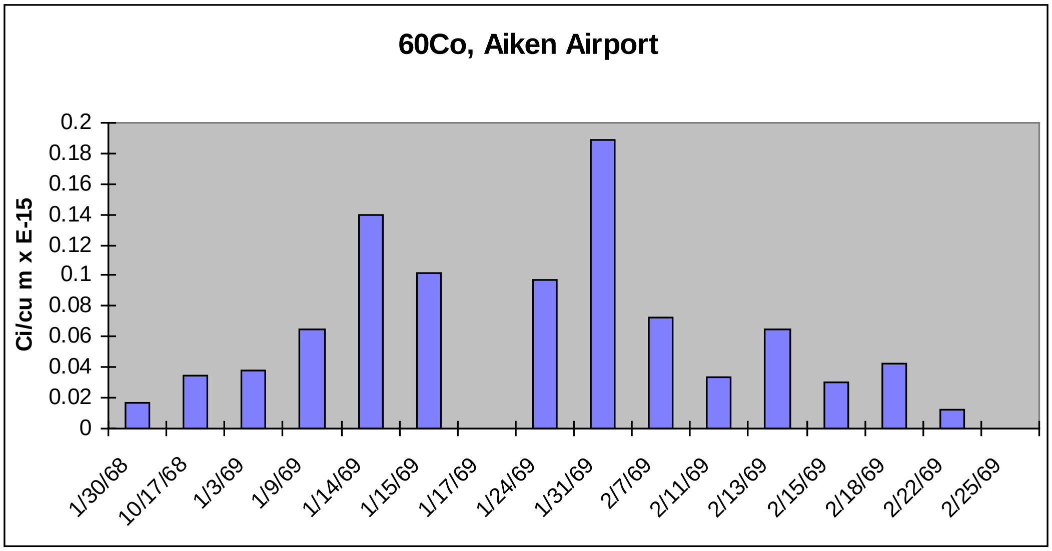

Figure 4.3-3. Cobalt-60 in air—Aiken Airport (extracted from Taylor 1969a, 1969b).

Jacobsen and Marter (1972) provides an interesting perspective on anticipated reactor releases. L-Reactor shut down in February 1968 and was restarted in October 1985. The reactor ran through December 1987, but with two extended shutdowns during the period. While this 1972 report is not of value in estimating L-Reactor activation product releases after the actual 1985 restart, it does base many of its conclusions on the functioning 1972-era reactors, stating that ". . . all facility improvements made in the three operating production reactors at SRP for minimizing environmental effects of operation will be incorporated (in L-Reactor)." The most significant of these modifications was to be construction of a closed-loop heat exchanger-filter-deionizer for cooling, clarifying and decontaminating disassembly basin water. The disassembly basin had been the primary source of release of radioactive materials to liquid effluent during prior operation of L-Reactor. The new system was said to be "comparable to those now used in other operating reactors and will greatly reduce the potential for release of radioactive materials." A program of seal welding was also proposed to reduce leakage from other areas in the reactor system, including the heavy water recirculation system.

Jacobsen and Marter (1972) considers the potential for release of all types of radionuclides; this section extracts data related to the activation products. Jacobsen and Marter anticipated that ^{41}Ar releases would be relatively constant from year to year, with reactor power ranging from about 2000–2300 MW(t). Actual maximum average power after the 1985 L-Reactor restart was reached in April 1986 at about 1900 MW(t), but average power over the 25-month period was just under 700 MW(t), including 8 months of extended shutdowns, or 1040 MW(t) not including the extended shutdowns. This latter value was about one-half the average value developed by the other reactors over their full power lives.

Jacobsen and Marter (1972) notes that some 55,000 gal of heavy water, 99.7% isotopic purity, would be used as moderator. The moderator would initially contain very little tritium, which would build in over the years after startup. Table 4.3-8, extracted from the startup report, shows the anticipated offsite effect of the tritium buildup and consequent increase in routine tritium leakage from the reactor. It is reproduced here to indicate the baseline dose predicted by the SRS for the release of other radionuclides, including the activation products.

Table 4.3-8. Predicted Population Dose from Operation of L-Reactor, for a Potential Mid-1970s Startup[a]

Operation (years following L-Reactor restart)	Population dose (man-rem), from:		
	Atmospheric releases	Aqueous releases	Total
1	26	1.3	27.3
2	28.8	3.9	32.7
3	31.5	5.9	37.4
4	33.7	7.8	41.5
5	36.3	9.8	46.1

[a] Source: Jacobsen and Marter (1972).

More specifically, the annular cavity (the space between the reactor tank and the circumferential shield systems) was to be filled with dehumidified air, with an assumed exchange rate to the reactor building of 100 cfm. This space had been designed to be filled with CO_2, but leakage had made this approach impractical. Natural (1%) ^{40}Ar in the annular cavity air would be converted to ^{41}Ar by neutron activation and would then be released to the environment via the main stack. Jacobsen and Marter calculate a production rate, decay period, and equilibrium concentration for ^{41}Ar in the operating reactor, but they also compare the value to the then-current value for P- and K-Reactors with similar neutron fluxes and reactor system designs and annual releases of about 50,000 Ci ^{41}Ar. They discuss the possible design of a delay tank to further reduce ^{41}Ar concentration prior to stack exhaust, but they indicate that no such design had been developed for L-Reactor or the other reactors as of 1972. Leakage of air through the moderator system was anticipated to result in another 1000 to 2000 Ci ^{41}Ar released up the stack per year. Fission product gases, concentrating in the blanket gas helium over the moderator, were predicted to be approximately 1000 Ci y^{-1} of ^{85m}Kr, 300 Ci y^{-1} of ^{88}Kr, 800 Ci y^{-1} of ^{135}Xe and 4,000 Ci y^{-1} of ^{133}Xe.

Based on past history in the L-Reactor fuel and target storage basin, estimated first year aqueous releases of activation products to Steel Creek were calculated in Jacobsen and Marter (1972) and are presented in Table 4.3-9.

Table 4.3-9. Estimated First Year Aqueous Releases, Based on 1971 K-Area Data[a]

Activation products	Release (Ci)
^{51}Cr	0.6
^{32}P	<0.1
^{65}Zn	<0.1
^{60}Co	<0.1
^{35}S	3.1

[a] Source: Jacobsen and Marter (1972).

The Jacobsen and Marter report projects the L-Reactor exhaust air rate from the 200-ft stack to be 120,000 ft^3 min^{-1} (3400 m^3 min^{-1}). On the basis of the expected ^{41}Ar release rate, the exhaust stack concentration of ^{41}Ar during L-Reactor operation (presumably at 2000–2300 MW[t]) was estimated to be $2.8 \times 10^{-5} \times$ Ci cc^{-1}, or $28 \times$ Ci m^{-3}. This is equivalent to 95,000 × Ci min^{-1}, or 50,000 Ci y^{-1} ^{41}Ar released , as previously noted in the report.

The Jacobsen and Marter report also estimates annual atmospheric SRS releases associated with L-Reactor operations to be 7×10^{-3} Ci y^{-1} for ^{60}Co. The report estimates maximum atmospheric plant perimeter concentrations associated with L-Reactor operations for ^{41}Ar as $9.9 \times 10^{-12} \times$ Ci cc^{-1}, and for ^{60}Co as $5.6 \times 10^{-19} \times$ Ci cc^{-1}.

Wood and Randolph (1974) describes the development, calibration, and in-stack testing of a continuously operating monitor for the noble gases at SRS. No release data of value are presented, however.

A memo, Mirshak (1983), notes the results of a comparison between SRS estimated ^{3}H and ^{41}Ar concentrations using the WIND system and concentrations measured by the U.S. Environmental Protection Agency. The average ^{41}Ar emission rate for P-Reactor was 1×10^{-3} Ci s^{-1} (a rate of 32,000 Ci y^{-1}) on December 15. A memo from Garrett to Corey on March 28, 1983 notes the same study. *RAC*'s reactor power study (see Chapter 2 of this report) indicates that P-Reactor was in stable operation at the time, at a power level of approximately 1700 MW(t).

A memo, (Anonymous 1986) tabulates SRS annual releases of ^{41}Ar as shown in Table 4.3-10.

Table 4.3-10. SRS Annual Releases of ^{41}Ar (all reactors)[a]

Year	^{41}Ar released (Ci)
1982	60,000
1983	41,000
1984	36,000
1985 (preliminary)	52,000

[a] Source: Anonymous (1986).

Anonymous (1986) estimates the offsite maximum individual dose as 1.54×10^{-6} mrem Ci^{-1} released. *RAC* notes that during this period P-, K-, and C-Reactors were all operating at high average power levels (except that C-Reactor shutdown in mid-1985 and L-Reactor restarted in late 1985).

Merz and Marter (1986) states that the annual guide of 30,000 Ci for ^{41}Ar released to the atmosphere from P-Area was exceeded during November 1985. A release of 2250 Ci in November increased the 1985 total to 30,460 Ci. It is noted that 90% of the ^{41}Ar released originates in the annular cavity, leaks into the process room, is mixed with air and released out through the stack. Because the K-Area and P-Area reactor systems are similar with respect to the conditions that produce ^{41}Ar, one or both of the monitors was believed to be inaccurate. Releases of ^{41}Ar for both areas are tabulated for 1985 (Table 4.3-11). Reactor power levels were similar over this period, although power data for March and April were not recovered during Phase II of this SRS study. To date, *RAC* has been unable to resolve the apparent discrepancy in ^{41}Ar release levels.

Table 4.3-11. Argon-41 Releases from SRS P- and K-Reactors, January–November 1985[a]

1985 Month	P-Reactor release (Ci ^{41}Ar)	K-Reactor release (Ci ^{41}Ar)
January	4200	758
February	5340	950
March	5340	600
April	-	1070
May	1360	1210
June	3100	1142
July	3100	1173
August	3080	1050
September	1750	19
October	940	940
November	2250	826

[a] Source: Merz and Marter (1986).

Sigg and Huang (1986) describes the development, calibration, testing, and validation of a mobile ^{41}Ar plume monitor, the TRAC. The device is sensitive to normal ^{41}Ar releases and is said to be capable of detecting a plume at 70 km downwind from a reactor, under typical transport conditions. The document provides no ^{41}Ar release data, although it does note, that since ^{41}Ar is formed by neutron activation of stable argon in air, its formation rate should be proportional to reactor power for a given reactor. Differences in reactor fluxes and annular cavities may give different source terms from different reactors at the same power level. Although SRP reactor power levels would be a better gauge of source terms, for simplicity in this report it is assumed the average of TRAC's observations represent average operating conditions.

SUMMARY: BEST ESTIMATES OF ^{41}AR RELEASES DURING SRS OPERATIONS, 1954–1988

Figure 4.3-1 earlier in this chapter presents airborne release rate data for the activation products in general. Of these, the primary radionuclide of interest in terms of airborne release and potential offsite dose is ^{41}Ar. SRS reactor operations produced and released relatively large quantities of ^{41}Ar, and its production rate in the air blanket surrounding a reactor should have been roughly proportional to the reactor power level. Cummins (1991) provides an SRS-developed estimate of ^{41}Ar releases. The data were presented in Table 4.3-2, and are summarized in Table 4.3-12 and Figure 4.3-4, below.

Table 4.3-12. Best Estimate of ^{41}Ar Releases to Air, All Reactors

Year	^{41}Ar to air (Ci)	Year	^{41}Ar to air (Ci)	Year	^{41}Ar to air (Ci)
55	5.0×10^{4}[a]	66	2.8×10^{5}[a]	78	5.3×10^{4}
55	1.4×10^{5}[a]	67	3.2×10^{5}[a]	79	5.3×10^{4}
56	2.5×10^{5}[a]	68	2.2×10^{5}[a]	80	7.0×10^{4}
57	2.9×10^{5}[a]	69	1.4×10^{5}[a]	81	6.2×10^{4}
58	3.4×10^{5}[a]	70	1.1×10^{5}[a]	82	6.0×10^{4}
59	4.4×10^{5}[a]	71	1.4×10^{5}	83	4.1×10^{4}
60	4.0×10^{5}[a]	72	1.7×10^{5}	84	3.6×10^{4}
61	4.2×10^{5}[a]	73	1.8×10^{5}	85	5.2×10^{4}
62	4.3×10^{5}[a]	74	1.1×10^{5}	86	8.3×10^{4}
63	4.5×10^{5}[a]	75	6.5×10^{4}	87	8.8×10^{4}
64	3.7×10^{5}[a]	76	8.3×10^{4}	88	3.0×10^{4}
65	2.7×10^{5}[a]	77	6.5×10^{4}	89	

[a] Values from Cummins (1991). Values before 1971 were estimated; data after 1970 were based on measurements.

While these values represent the best available estimates for ^{41}Ar releases from the SRS reactors, we note that the values presented for the later years (1974–1988) are quite low when compared to reactor power levels and overall average ^{41}Ar production levels. We also observe that the ^{41}Ar release values presented for certain of the early years (1955–1967) are quite high when compared to reactor power levels for the same period. The reason for these apparent discrepancies, demonstrated in the following graphs, is not clear.

The following graph (Figure 4.3-4) plots the data from Cummins (1991) data on a linear scale, for comparison to reactor power levels as seen in Figure 4.3-5.

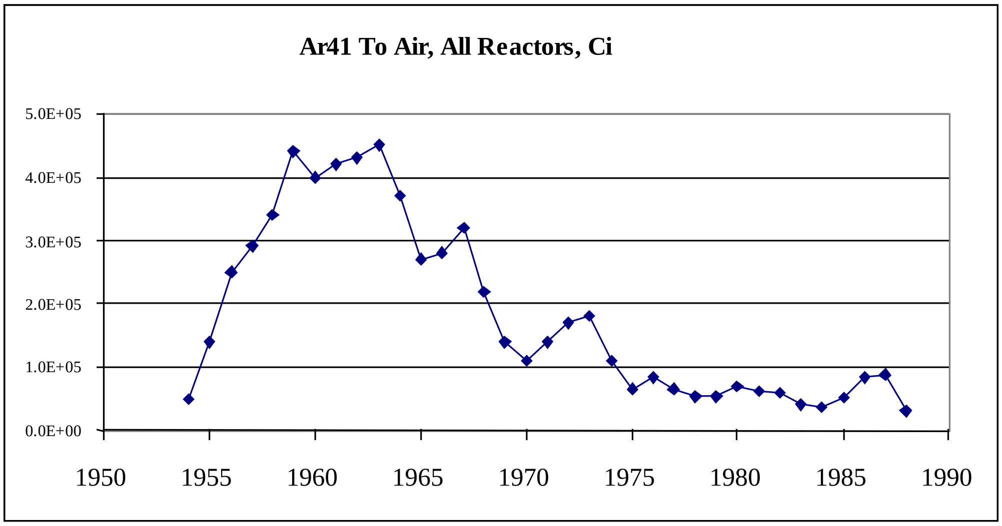

Figure 4.3-4. SRS Estimate of ^{41}Ar releases, 1954–1989 (from Cummins et al. 1991).

Chapter 2 of this report presents data extracted by *RAC* concerning SRS reactor power levels for the period 1955–1988. Those data are summarized in Figure 4.3-5.

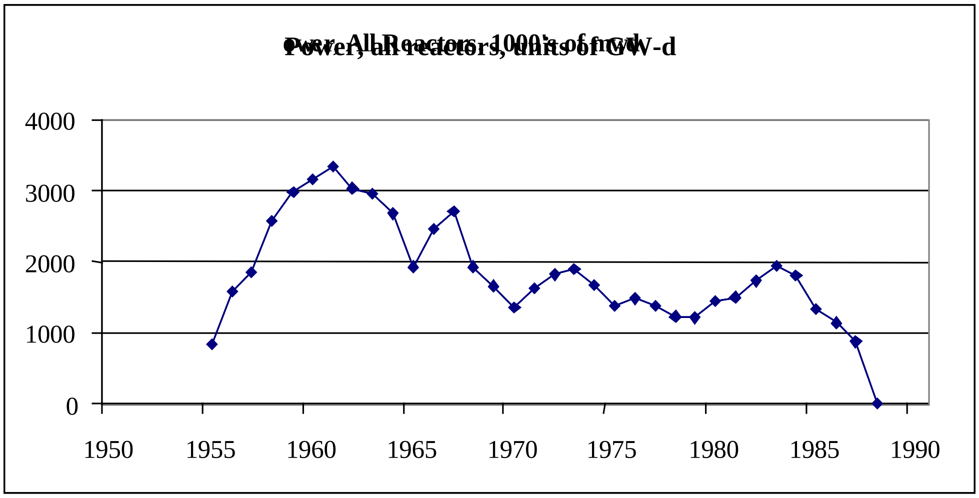

Figure 4.3-5. Combined SRS reactor power output, 1955–1988.

Figure 4.3-6 allows examination of annual reactor power level data by individual reactor.

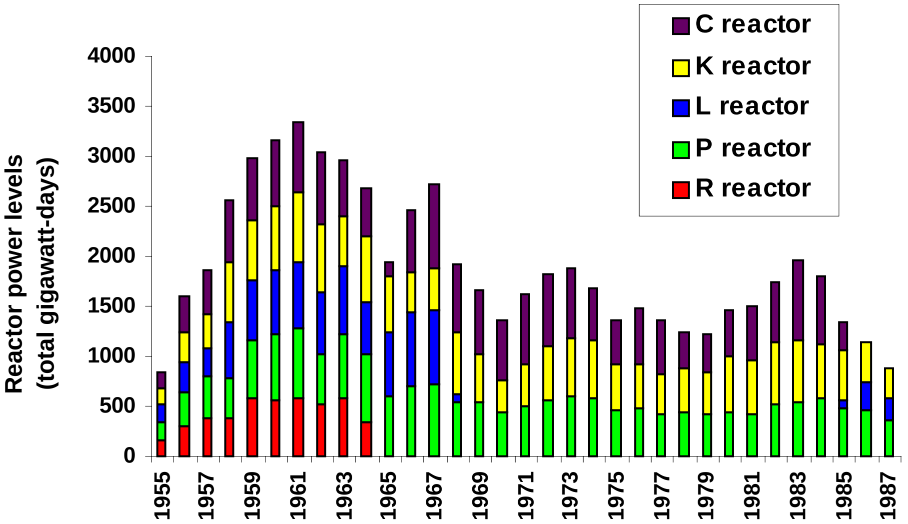

Figure 4.3-6. SRS reactor power output, 1955–1988. (Note: for black and white copies: the vertical order within the data bars is the same as the order of reactors in the key.) From Chapter 2 of this report.

Figure 4.3-7 presents a comparison of the ^{41}Ar and reactor power data sets, with the ^{41}Ar values having been divided by 100 to allow direct comparison on the same scale. Here, the discrepancy between the two sets of information becomes apparent, with relatively high estimated ^{41}Ar release values apparent in the early years, and relatively low values apparent during the later years.

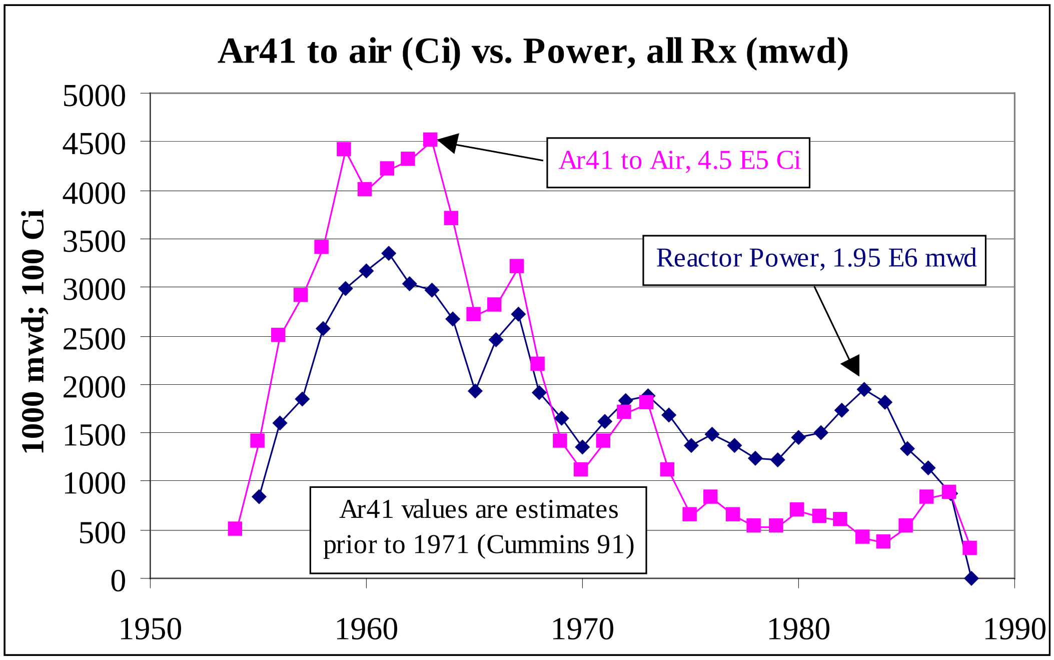

Figure 4.3-7. Comparison of SRS ^{41}Ar release estimates versus reactor power levels.

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