5 RELEASE OF RADIONUCLIDES TO AIR

This chapter summarizes the Phase III effort to derive a detailed source term for radionuclides released into the air from Savannah River Site (SRS) facilities. These radionuclides and the annual quantities released are listed in Appendix B.

CDC previously sponsored Phase I and II of the SRS Dose Reconstruction Project. During Phase I, records necessary for the project were assembled, including facility release records, monthly and weekly reports, laboratory notes, research papers, and personnel interviews. During Phase II, radionuclide and chemical releases were estimated using the records obtained during Phase I. The Phase II study considered uncertainties in the SRS reports of historical radionuclide release. For some isotopes of concern, particularly ^{131}I, the Phase II study provided estimates of release that are notably larger than those historically reported by SRS (Phase II).

The Phase II study was used as the primary basis for the Phase III source term. The Phase II estimates were refined for use in Phase III to achieve greater source term resolution in time and space: i.e., releases by year and location. This greater resolution was mandated by geographically distributed scenarios and the concern with temporal changes expressed in the scenario specifications. Other refinements related to isotopic composition and chemical form provided for more precise modeling of health effects.

5.1 Development of Study Source Term

5.1.1 Approach to Development of Study Source Term

As discussed in Chapter 1, the scope of this Phase III dose reconstruction study was intended to move beyond a screening analysis to identify biologically significant radionuclides and dose pathways. In order to produce a dose assessment with the desired level of detail and confidence, a more refined source term than was available from Phase II was sought. This refined source term for air releases addressed specific requirements of the analysis, which included the following broad themes.

- To use the results of Phase II, including the identification of significant radionuclides by a screening analysis and modification of historical release data to account for instrumentation deficiencies.
- To provide enough spatial definition to the source term so that the geographical characteristics of the various scenarios (and their proximity to particular sources on site) would be adequately represented.
- To provide enough temporal definition to the source term so that the age-related characteristics of scenario members (as they interacted with the time-varying releases from the SRS) would be adequately represented.
- To provide enough detail about the chemical and isotopic characteristics of the releases so that the dose and risk modeling (treatment of health effects) would be more precise.
- To enhance the completeness and/or accuracy of the source term.

Starting with the Phase II results, a sequence of steps was followed to obtain as source term for Phase III with the desired qualities. Figure 5-1 outlines this sequence and briefly describes for each step the requirement or need addressed, the additional input information used, and the status of the data set representing the source term after each modification. Each step is described in more detail in the following sections.

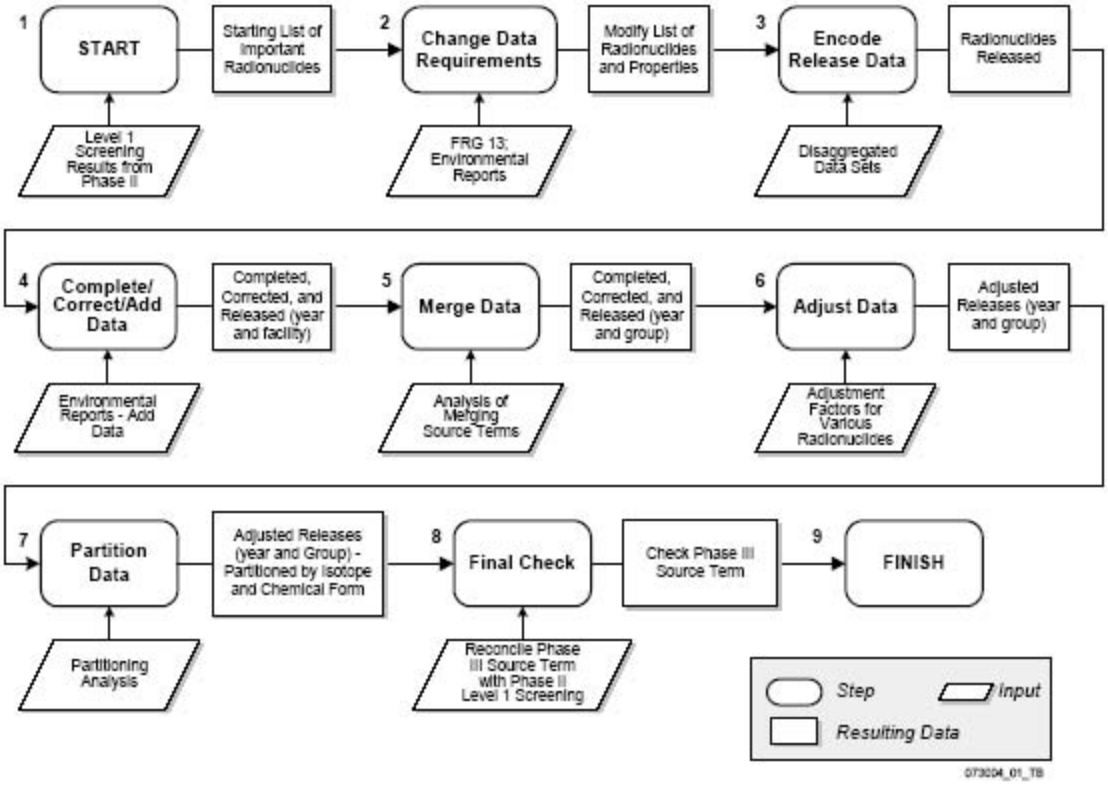

Figure 5-1 Development of Study Source Term

5.1.2 Identify Important Radionuclides based on Phase II

Phase II provided a number of results that became starting points for Phase III, including: 1) Two screening analyses that established a reduced set of radionuclides to consider in the dose reconstruction; and 2) Detailed source terms for these radionuclides, with varying degrees of spatial definition. These two aspects are summarized in the following sections.

5.1.2.1 Use Phase II Results (Step 1)

Phase II, Screening Level 1, identified twelve groups of key radionuclides released to air; Screening Level 2 further refined the list to include only six radionuclides. To avoid possibly overlooking an important contributor to the Phase III radiological assessment, all twelve groups of key radionuclides identified in the Level 1 screening were modeled in Phase III.

The Phase II screening assessment started with a master list of radionuclides that had been released into the air from SRS facilities. Preliminary estimates of their annual average release rates were also made. But because this list was too large for efficient analysis, a screening assessment was performed to identify a smaller list of key radionuclides that were the dominant contributors to radiation dose and risk (Phase II). This screening assessment was performed using a two-step method recommended by the National Council on Radiation Protection and Measurements (NCRP) for prospective evaluation of a new facility. The method featured 1) a screening considering the summation of six possible exposure pathways, and 2) a screening considering each of the exposure pathways plus the sum total of all six pathways (1), (2).

In both screening steps significant simplifications included:

- Releases were averaged over 36 years (from 1954 to 1989).
- Concentrations at the point of release were used, rather than concentrations plausibly at the locations of individuals.
- Standard human behavior patterns -- e.g., consumption rates of foods -- were assumed.
- Adult rather than age-dependent dose conversion factors were used (Phase II).

For these and other reasons, the screening values thus calculated were not plausible representations of doses to actual humans, even though the screening values have units of Sv/y. Rather, the screening values were highly conservative estimates of the average annual stylized dose to hypothetical receptors meant only to identify those radionuclides that should be considered in further analyses.[1]

For Level 1, a total screening value was calculated representing the sum of screening values calculated for each radionuclide. Then those radionuclides were identified that had screening values contributing at least 0.1% of the total screening value. Using a cut-off of 0.1% of the total estimated dose, the initial list of radionuclides was reduced to twelve. These twelve key radionuclides together accounted for more than 99% of the total screening value (Phase II). The radionuclides thus identified in the Level 1 of the screening assessments for air release are listed in Table 5-2. Many of the radionuclides are actually groups of aggregated isotopes such as combined ^{239}Pu and ^{240}Pu.

Table 5-2 Radionuclide Rank from Phase II Level 1 Screening

Radionuclide	Percent of Total Screening Value (%)
^{131}I	50.6
^{3}H	33.7
^{41}Ar	7.0
$^{239, 240}$Pu	2.6
^{129}I	2.3
^{103,106}Ru	1.1
^{238}Pu	0.7
^{14}C	0.6
^{137}Cs	0.6
Uranium*	0.3
^{89,90}Sr	0.2
^{241}Am	0.1
Total	99.8

*Screened in Phase II as combined ^{235}U and ^{238}U.

[1]A report describing the NCRP screening method notes, on page 4: "The assumptions and methods incorporated into all of the screening procedures presented in this Report are such that actual doses should not be underestimated by more than one order of magnitude. In most situations, the actual dose will be significantly less than the values calculated for screening" (2).

This is the starting list of important radionuclides for Phase III.

5.1.2.2 Phase II Estimates of Radionuclide Release

Phase II provided estimates as annual averages over 36 years of site operation for all twelve groups of key radionuclides identified in Level 1 of the screening analysis (see Table 5-3). These averages spanned a period from 1954 through 1989. Annual release estimates were provided for all twelve radionuclide groups except ^{14}C and ^{241}Am for which 36-year averages were provided. More detailed estimates were provided on an annual basis for tritium; $^{89,90}Sr$; ^{129}I; ^{131}I; ^{137}Cs; uranium; ^{238}Pu; and $^{239,240}Pu$. The level of detail varied depending on the isotope. In some cases, for example, releases from specific SRS facilities (such as specific nuclear reactors) were identified, and in other cases, releases from SRS Areas or groups of facilities (such as groups of seepage basins) were identified.

Table 5-3 Radionuclides Reported in Phase II Study by Facility and Year

Radionuclide Group	Annual Estimates Provided for a Specific Facility or a Group of Facilities?	Estimates Provided as a 36-Year Average Yearly Release from the Entire Site?
^{3}H	Annual	Yes
^{14}C	Not available	Yes
^{41}Ar	Annual	Yes
$^{89,90}Sr$	Annual	Yes
$^{103,106}Ru$	Annual	Yes
^{129}I	Annual	Yes
^{131}I	Annual	Yes
^{137}Cs	Annual	Yes
Uranium	Annual	Yes (reported as $^{235,238}U$)
^{238}Pu	Annual	Yes
^{239}Pu	Annual	Yes (reported as $^{239,240}Pu$)
^{241}Am	Not available	Yes

5.1.3 Change Data Requirements to Better Model Health Effects (Step 2)

The initial list of important radionuclides resulting from the Phase II Level 1 screening analysis was modified in several ways for use in Phase III:

1. Some groups of radionuclides were partitioned into separate isotopes. This was done to provide a more refined treatment of dose modeling. By treating isotopes individually, health effects coefficients specific to those isotopes could be used in the dose modeling; if isotopes were aggregated by element, as in the Phase II screening analyses, a single coefficient (related to the isotope with the greatest health effects) would characterize all the isotopes of the element, possibly greatly overestimating doses and risks. Table 5-4 shows 16 isotopes modeled.
2. Because the releases of Ruthenium were relatively small, all releases were modeled as ^{106}Ru.

3. Uranium isotope ^{234}U and ^{236}U were added to account for presence of these isotopes in the nuclear reactors.
4. One radionuclide, ^{240}Pu was dropped from consideration. The data upon which the Phase II estimates of releases were based did not list ^{240}Pu releases separately, but always combined with other isotopes. Environmental transport of these Pu isotopes is expected to be identical. Furthermore, the health effects coefficients for ^{239}Pu and ^{240}Pu are very close (ingestion and inhalation dose coefficients are the same; ground plane coefficients differ by a factor of about 2), so modeling ^{240}Pu as ^{239}Pu will have little effect on the dose estimates. Since there appeared to be no basis and no motivation for differentiating these two isotopes, separate consideration of ^{240}Pu was dropped.
5. It was considered appropriate to include as releases radioactivity measured as unidentified alpha or unidentified beta-gamma activity. Although the SRS had recorded releases of such material over the years of site operation, an explicit treatment in Phase II was not apparent. Therefore two additional categories were added for these types of releases. However, to provide bounding estimates of their health effects, the unidentified alpha activity was added to the releases for ^{239}Pu and the unidentified beta-gamma activity was added to the releases for ^{90}Sr. The treatment of unidentified beta-gamma emitters as ^{90}Sr is likely to be conservative for ingestion and inhalation doses, but may not be conservative for external doses. However, the addition of unidentified beta-gamma releases increased the ^{90}Sr total releases to water by a factor of about 2.5 over the Phase II screening analysis, leading to higher estimated doses. Because this is only an adjustment to one isotope in calculating the external doses, which are generally small fractions[2] of the total dose, this potential lack of conservatism is not considered significant.
6. At this point the need to refine the treatment of certain radionuclides by defining their chemical form was established. Two radionuclides: tritium and ^{131}I can have substantially different behavior in the environment depending on their chemical form. The methods used to partition these radionuclides into different chemical forms are described in Section 5.1.8.2. Methods used to partition some elements into different isotopes are described in Section 5.1.8.1

Table 5-4 Modified List of Radionuclides Considered for Phase III

^{241}Americium	^{129,131}Iodine
^{41}Argon	^{238,239}Plutonium†
^{14}Carbon	^{89,90}Strontium‡
^{137}Cesium	^{234,235,236,238}Uranium
Tritium (^{3}H)	^{106}Ruthenium

†Unidentified alpha-emitters were modeled as ^{239}Pu
‡Unidentified beta-gamma emitters were modeled as ^{90}Sr

Although some air releases were identified as containing ^{134}Cs and ^{137}Cs, they were modeled entirely as ^{137}Cs, since the screening analysis had already eliminated ^{134}Cs as an important isotope. Although ^{134}Cs has higher dose coefficients than ^{137}Cs, especially for external radiation, the fraction of the mixtures attributed to ^{134}Cs appears to be a few percent, so the overall effect on dose estimates is not expected to be significant.

[2] Except for the shoreline dose to children in the Near-River Family.

5.1.4 Encode Release Data from Cummins Report (Step 3)

The Phase III source term needed definition in space and time so the assessment of radiation doses and cancer risks would be able to reflect differences associated with the seven hypothetical exposure scenarios considered in Phase III. Enhanced spatial definition of the source term allowed the radiological assessment to model and to evaluate the importance of several facets of the Phase III SRS dose reconstruction, including:

1. Different mixtures of radionuclides dominated the releases from the major SRS discharge locations, and these locations are often separated by several miles.
2. Radiation doses at different exposure locations specified by the scenarios depended on: 1) the quantities of particular radionuclides released into the air at the various SRS location and 2) direction-related meteorological conditions, such as the frequency that the wind blew in the direction of a particular exposure location. Several exposure locations are close to the release points, relative to the distances between some release points.

A more refined definition of the spatial distribution of releases allows for a more precise estimate of doses compared to a source term with a single assumed release point to represent all SRS release points.

Enhanced temporal definition of the source term -- that is, specifying the amount of release for each radionuclide for each year -- allowed the radiological assessment to model and to evaluate the importance of several facets of the Phase III SRS dose reconstruction:

- The behavior of the children in each hypothetical scenario changed significantly during their childhood; this included breathing rates, food ingestion rates, time spent indoors and outdoor.
- The dose and risk factors for children were adjusted until they became adults.
- The annual release rates of many key radionuclides changed considerably over 39 years; of special note were the early years of large releases (e.g., 1956).

The source term, the behaviors of the hypothetical receptors, and their dose and risk characteristics all varied in time; by considering the annual variation of the source term, the interaction of all these time variations was considered. A source term that was expressed in terms of an average over 39 years would not have allowed these important variations to be adequately considered.

To provide the desired level of spatial and temporal definition in the source term, historical release data (3), (4) was keyed into an electronic data base. This data base described radionuclide releases by isotope, facility, and year. All the data were fully cross-checked and was subject to a thorough quality assurance program.

5.1.5 Complete and Correct Data Set (Step 4)

In the initial stages of the Phase III source term data compilation, it was observed that data were not available in the Phase II report for annual release estimates for some radionuclides for the years 1954 through 1989 and for most radionuclides for the years 1990 through 1992. For Phase III, refinements to the Phase II release estimates were made to obtain a source term for all radionuclides: 1) on an annual basis; 2) for all 39 years of the study; and 3) on the basis of major SRS facilities.

For the years 1954 through 1989, when annual release estimates were given in the Phase II report in sufficient detail, Phase II was used to determine annual facility-group release rates. Otherwise, data gathered from SRS references was used. For example, for ^{14}C and ^{241}Am, the Phase II report only provided estimates for the entire site as averaged over 36 years. For $^{103,106}Ru$, the electronic version of

Phase II provided a supplemental Excel file that listed annual release estimates for the F- and H-Separations Areas, but not for reactors. Phase II provided annual release data for ^{41}Ar, but the data were not proportioned among the five reactors that released this isotope.

To apportion release data among the different SRS facilities releasing radionuclides into the air, the primary reference was (3). In some cases, information in (3) and other references were given as combined values for a group of facilities (e.g., reactors). For these cases, SRS site release history was reviewed and the combined values were apportioned among facilities in accordance with the processes in the facilities that resulted in the airborne releases. See Section 5.1.8.1 and Appendix R for further information on how the release data were partitioned.

For the years 1990 through 1992, release estimates were not provided in Phase II for most radionuclides Therefore, SRS environmental reports and other environmental data (3), (4), (5), (6), (7), (8) were reviewed for these three years, and the missing source terms were added when information was available.

5.1.6 Merge Data from Fifteen Facilities into One of Four Groups (Step 5)

Section 5.1.4 describes the motivation for refining the spatial definition of the source term. At this point in the analysis, the source term was defined in a data base by each radionuclide, year, and facility. Fifteen separate facilities were considered to be independent sources of air releases. The types of radionuclide releases from each of these fifteen facilities are shown in Table 5-5.

Table 5-5 Main Sources of Air Releases of Radionuclides at SRS

Facility Type	Designation	Principal Radionuclides Released			
		Tritium	β-emitters	α-emitters	Activation Products
Reactors	C	X	X		X
	K	X	X		X
	L	X	X		X
	P	X	X		X
	R	X			X
Separations Facilities	F Canyon	X*	X	X	X
	H Canyon	X*	X	X	X
	H Tritium Stack	X*			
Waste Facilities	All Reactors Seepage Basin Evaporation†	X			
	CMX-TNX	X			
	D Area	X			
	F & H Seepage Basin Evaporation‡	X			
Other Facilities	A Area	X	X	X	X
	M Area	X		X	
	SRL	X§	X§	X§	

*Releases for these three facilities are combined in available literature for security purposes.
†The "All Reactors Seepage Basin Evaporation" is already a virtual source. Each reactor had its own seepage basin. The location of this virtual source was chosen to be close to the centroid of the locations of the reactors.
‡The "F & H Seepage Basin Evaporation" is already a virtual source, but the seepage basins for F- and H-Areas are

Facility Type	Designation	Principal Radionuclides Released			
		Tritium	β-emitters	α-emitters	Activation Products

only about one mile apart.
[§]Releases from the SRL may have been included in the A-Area releases for at least some years.

Although greater spatial definition was desired, these 15 major sources, as well as several minor sources were too numerous for efficient analysis considering the number of radionuclides, exposure scenarios, and exposure pathways to be addressed and the sensitivity and uncertainty analyses to be performed. Data storage capacity, software limitations, and post-processing time were major factors in deciding to reduce the number of air release locations. Ultimately dose data were recorded for each of 39 years, 30 radionuclides, 4 exposure routes, 10 transport pathways, 28 receptors, 10 exposure locations, and 4 air release locations. This produced a data set size in excess of 50 million, which was still challenging.

Although spatial simplification of the source term was desirable, any such simplification needed to preserve the inherent geographical variation of the air source term and a suitable level of precision in the dose calculations. Therefore, a two part analysis was performed:

1. Different combinations of individual sources into groups (virtual sources) were evaluated to determine a representative set of "virtual" sources. Different numbers of virtual sources were evaluated, ultimately settling on four groups of sources (as shown in Table 5-6).
2. A careful evaluation was performed to assure that representation of a group of sources by a single virtual source preserved a suitable precision in estimating concentrations; as discussed in more detail below, the representation of a group of sources by these virtual sources was deemed to be sufficiently precise.

This simplification of sources is summarized in Chapter 6 and addressed in detail in Appendix A. The source groupings developed as a result of this analysis are shown in Table 5-6 and Figure 5-2.

Table 5-6 Groupings for Four Virtual Sources

Groups	Facilities in Group
Group 1	A-Area, M-Area, SRL
Group 2	F-Canyon, H-Canyon, H Tritium Stack, F&H Seepage Basins
Group 3	C-Reactor K-Reactor, L-Reactor, Reactor Seepage Basin release attributable to C-, K-, and L-Reactors, D-Area, CMX-TNX
Group 4	P-Reactor. R-Reactor, Reactor Seepage Basin release attributable to P- and R-Reactors

The Phase III source term for releases to air, compiled up to this point for each facility by year and radionuclide, was aggregated based on these four virtual release points. The rectangular coordinates for the actual sources and the four virtual sources are given in Table A-4. Minor sources from seepage basins and waste areas were merged with the nearest reactor or canyon release. This reduced the number of actual sources considered from 15 to 11.

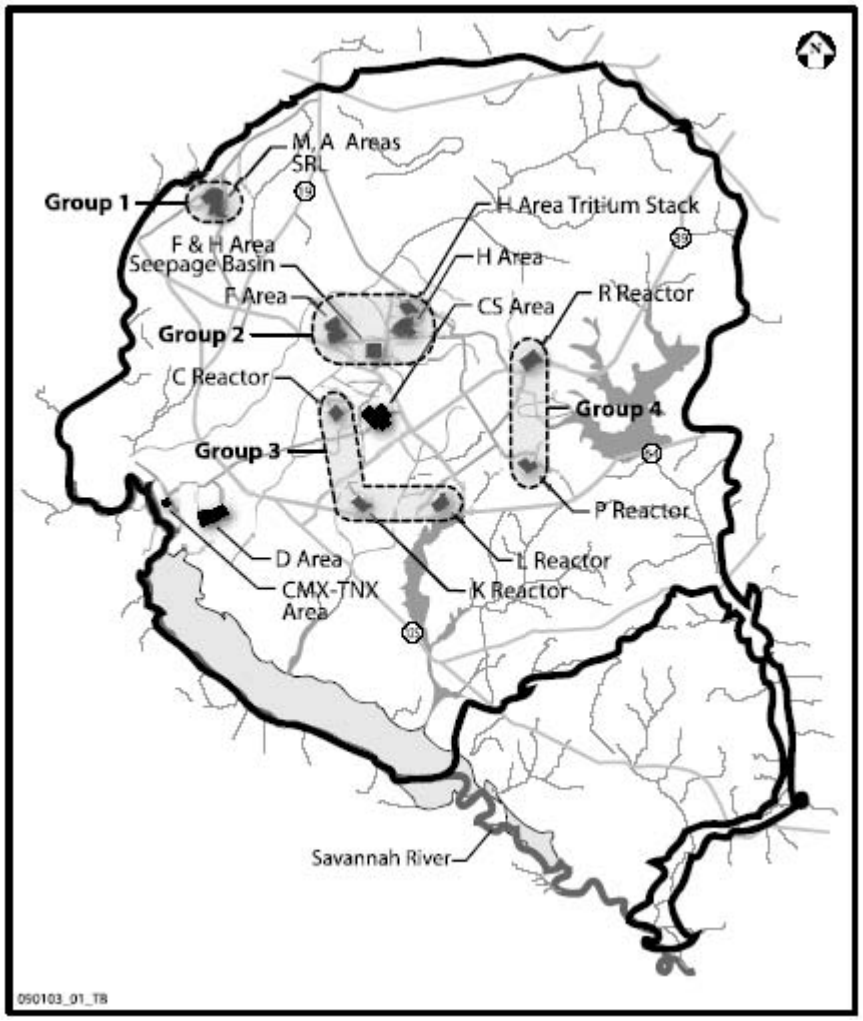

Figure 5-2 Virtual Source Locations

The use of virtual source locations, rather than the actual source locations, is an approximation. This approximation was evaluated by comparing concentrations calculated using the actual location to concentrations calculated using the virtual location. First, each real source location (e.g., Reactor L) was assumed to have a unit release rate (1 Ci/y). The GENII code was used to compute the resulting concentrations at the 9 offsite locations (e.g., Girard, GA). Then the virtual source location, corresponding to the real source location (e.g., for Reactor L the Group 3 virtual location was used), was assumed to have a unit release rate. Again the GENII code was used to compute the resulting concentrations at the 9 offsite locations. The exact concentrations using the real source location were compared to the approximate concentrations using the appropriate virtual source location. The process was then performed for each of the 11 real source locations in turn. Exact and approximate concentrations differed by 18% or less for all real source locations but one, for which concentrations differed by 27% or less. This is considered to be excellent agreement for a very stringent comparison, given that for most groups the individual sources did not operate alone. In most cases the agreement between the exact and approximate concentrations was even closer; for example, of the 99 comparisons (9 offsite locations x 11 actual sources) only 18 had differences of greater than 10%. Another comparison was performed in which all real source locations in a group were assumed to have the same release rate

and the virtual location was assumed to have a release rate equal to the sum of these release rates; e.g., for Group 1 each of the three real sources was assumed to have a release rate of 1/3 Ci/y, while the virtual source had a release rate of 1 Ci/y. For these conditions, exact and approximate concentrations differed by less than 11%. It should be noted that the "exact" and "approximate" concentrations discussed above are both analytical estimates of the average annual concentrations at various locations. These analytical estimates may differ from the actual average annual concentrations at the site because actual weather and release rates were time varying, while these analyses use annual release rates and meteorological conditions averaged over 20 years.

5.1.7 Adjust Data in Accordance With Modifications Made Phase II (Step 6)

Particularly for ^{131}I, uranium, plutonium, and tritium the Phase II estimates of radionuclide release were larger than those historically reported by SRS (3), (4). Phase II increased the released quantities to account for deficiencies in instrumentation and recording releases. Since historical data were used to provide a greater degree of spatial and temporal definition to the source term, these historical data needed to be modified for use in Phase III to account for these adjustments introduced in the Phase II report.

In Phase II, additional detailed analyses of historical estimates of release were performed for iodine, plutonium, uranium and tritium. These Phase II analyses compensated for several factors that may have caused the reported site releases to be smaller than the releases that actually occurred, including the use of different samplers over time, sampler collection efficiencies, errors associated with counting equipment, and the chemical forms of the released radionuclides. Based on a model of the transmission of effluents in sampling lines and Monte Carlo analyses, Phase II provided revised estimates for iodine, uranium, and plutonium that were generally larger from those reported over the years by SRS (Phase II). Of particular note was ^{131}I: The Phase II estimates were significantly larger than releases historically reported by SRS.[3] Phase II also observed that releases for the early years of operation (through about 1961) were more uncertain than later releases. Phase II developed probability distributions describing the releases for key radionuclides for each year modeled in Phase II. These probability distributions showed more spread (larger ratios of standard deviations to the mean value) in earlier years than in later years.

In order to incorporate the release values adjusted for uncertainties that were developed in Phase II, the adjusted values of release by year and by facility were imported from spreadsheets attached to the Phase II report (Phase 2, Directory titled Supporting_Files). Adjusted data were used for plutonium, uranium, and iodine releases. Adjusted data were not used for tritium releases in Phase III.

5.1.8 Partition Data to Account for the Mix of Isotope and Chemical Form (Step 7)

In some cases, the radionuclide releases studied in Phase II were compiled on a combined basis, e.g. all isotopes of uranium, all chemical forms of tritium. In order to enhance modeling of the health effects of air releases from the SRS in Phase III, some of these combined releases were partitioned further into specific isotopes or chemical forms. The following sections describe how certain combined releases were separated further into specific isotopes or specific chemical forms.

[3] SRS revised its iodine monitoring and sampling systems and procedures over time. According to the Phase II report, about 99% of the ^{131}I released into the air came from the separations areas. Until September 1961, only elemental ^{131}I was measured at the separations areas. SRS estimated organic iodide activity from elemental ^{131}I release, assuming that organic forms represented 70-90% of all iodine released from separations. The Phase II report addressed measurement uncertainties such as sample collection efficiency and measurement biases resulting from deposition of elemental iodine in sampling lines, and concluded that SRS underestimated the release of ^{131}I to air during early years (1950s and early 1960s). For example, for each year from 1955 through 1960, the Phase II report estimated that the annual total iodine released to the air was at least 20 times larger than was reported in Cummins (3). For the year 1961 the total iodine released to air was 12 times larger. SRS staff (9) have criticized these Phase II estimates. After 1961, however, the Phase II estimates of ^{131}I release more closely correspond to those reported by SRS (with few exceptions). Annual ^{131}I releases after 1961 were much smaller than in earlier years (Phase II).

5.1.8.1 Isotopic Partitioning

Several radioactive constituents that were reported in the Phase II study and in SRS environmental reports were apportioned from the combined activities stated by these sources into release estimates for individual isotopes. For example, the release estimates provided in the Phase II study for ^{89}Sr and ^{90}Sr combined, were partitioned into release estimates for each of these isotopes by considering the nuclear processes occurring at the facilities that generated these isotopes. These constituents included:

- ^{89}Sr, ^{90}Sr.
- ^{134}Cs, ^{137}Cs.
- Uranium.
- Plutonium.
- Unidentified alpha.
- Unidentified beta + gamma.

Table 5-7 summarizes the sixteen radionuclides that were determined from this partitioning process for release to air. Source terms for Phase III were developed for each of these sixteen radionuclides. Table 5-8 summarizes how the total activity of each constituent was partitioned into constituent isotopes for each SRS site area. Additional technical information and general assumptions for the partitioning process are in Appendix C.

Radionuclides identified as ^{89,90}Sr in the Phase II report were apportioned into ^{89}Sr and ^{90}Sr. For most facilities, the activity was assumed to be ^{90}Sr. For the separations area, the activity was apportioned into 75% ^{89}Sr and 25% ^{90}Sr based on Cummins, et al. (3).

Radionuclides released to air and identified as ^{134,137}Cs in the Phase II report were assumed to be entirely ^{137}Cs. Data regarding releases of ^{134}Cs and ^{137}Cs indicate that the fraction of ^{134}Cs was likely to be a few percent. (see Appendix C).

Table 5-7 Radionuclides Considered in Phase III Compared to Those Cited in Phase II

Radionuclides Identified in Step 1 of Phase II Screening	Radionuclides Considered in Phase III Air Release Assessments
^{3}H	^{3}H
^{14}C	^{14}C
^{41}Ar	^{41}Ar
$^{89, 90}$Sr	^{89}Sr & ^{90}Sr
^{103,106}Ru	^{106}Ru
^{129}I	^{129}I
^{131}I	^{131}I
^{134,137}Cs	^{137}Cs
Uranium	^{234}U, ^{235}U, ^{236}U, & ^{238}U
^{238}Pu	^{238}Pu
^{239,240}Pu	^{239}Pu
^{241}Am	^{241}Am

Table 5-8 Summary of Assumed Isotopic Distributions for Air Release

Constituent	SRS Area	Isotopic Distribution by Activity
$^{89, 90}$Sr	F&H Areas	75% ^{89}Sr; 25% ^{90}Sr
	A Area	100% ^{90}Sr
	D Area	100% ^{90}Sr
	Central Shops	100% ^{90}Sr
$^{134, 137}$Cs	D Area	100% ^{137}Cs
Uranium	Reactor Areas	91.73% ^{234}U; 1.79% ^{235}U; 6.45% ^{236}U; 0.03% ^{238}U
	F Area	1.27% ^{235}U; 98.73% ^{238}U
	H Area	91.73% ^{234}U; 1.79% ^{235}U; 6.45% ^{236}U; 0.03% ^{238}U
	M Area	49.49% ^{234}U; 2.25% ^{235}U; 48.26% ^{238}U
	A Area (SRL)	91.44% ^{234}U; 1.8% ^{235}U; 6.4% ^{236}U; 0.36% ^{238}U
	CMX/TNX	49.49% ^{234}U; 2.25% ^{235}U; 48.26% ^{238}U
	D Area	91.73% ^{234}U; 1.79% ^{235}U; 6.45% ^{236}U; 0.03% ^{238}U
Total Plutonium	All Areas	100% ^{239}Pu
Unidentified Alpha	All Areas	100% ^{239}Pu
Unidentified Beta-gamma	All Areas	100% ^{90}Sr

Radionuclides identified as ^{103,106}Ru in the Phase II report were assumed to all be ^{106}Ru. This assumption was consistent with the screening analysis used in the Phase II report to identify key radionuclides.[4] This assumption will conservatively estimate doses from ingestion and inhalation.

Radionuclides identified as uranium in Phase II were apportioned into four uranium isotopes: ^{234}U, ^{235}U, ^{236}U, and ^{238}U. The Phase II report only considered the uranium isotopes ^{235}U and ^{238}U when it performed its screening analysis. For Phase III, however, ^{234}U and ^{236}U were included. ^{234}U is present in natural and enriched uranium. ^{236}U is produced from neutron bombardment in nuclear reactors, and would have been present in uranium released from the separations areas, among other areas at SRS.

Activity attributed to ^{239,240}Pu in the Phase II report was assumed to be entirely ^{239}Pu. Although the partition between these two isotopes for this type of release was estimated (see Appendix C), the isotopes were not modeled separately because their environmental transport is the same and their health effects coefficients are very close. Radioactive releases at SRS were often reported as unidentified alpha activity, unidentified beta-gamma activity, or undifferentiated plutonium activity. In most cases, the unidentified constituents would have been a combination of several isotopes. The fractions of total activity that individual isotopes represented probably varied from one release event to another. For simplicity, unidentified alpha activity was conservatively assumed to be ^{239}Pu because it was one of the more prevalent isotopes created at SRS, and because it is more radiotoxic than uranium isotopes for many of the exposure pathways. Other transuranic isotopes that have been produced and used at the SRS (such as ^{237}Neptunium, ^{244}Curium and ^{252}Californium) may have contributed to unidentified alpha activity, but these isotopes were produced primarily during brief periods.

[4] In the supplemental files provided as part of Chapter 3 of Phase II, the following notation was made: "We combined all estimates of reported releases of ^{103}Ru (0.2 Ci), ^{106}Ru (0.5 Ci), and ^{103,106}Ru (~1 Ci), and then used the screening factor for ^{106}Ru to ensure a conservative approach" (Phase II).

Unidentified beta-gamma activity was modeled as ^{90}Sr, which was commonly reported in environmental release reports at SRS. ^{90}Strontium has a half-life that is sufficiently long (28 years) that it may persist in the environment for many years after release. On the other hand, the health effects of ^{90}Sr will exceed those of other radionuclides for important dose pathways (e.g., doses per unit of activity from inhalation and ingestion will be greater for ^{90}Sr than for ^{60}Co). Assuming unidentified beta-gamma activity is ^{90}Sr may underestimate doses from direct radiation, but since this is confined to this one radionuclide category and since direct radiation is generally not a major pathway, it is expected to have a small impact on overall dose estimates.

Undifferentiated plutonium (occasionally found in SRS reports) was assumed to be ^{239}Pu as this was the main plutonium isotope produced at the site.

5.1.8.2 Chemical Forms of Radionuclides

For the Phase III study two types of releases were partitioned according to their chemical form: tritium and iodine. Section 5.1.8.2.1 discusses Phase III assumptions about the chemical forms of tritium released to air. Section 5.1.8.2.2 discusses Phase III assumptions about chemical forms of iodine isotopes released to air.[5]

5.1.8.2.1 Tritium

Tritium was one of the major nuclear materials produced at SRS. The Phase II report identified two chemical forms of release: an elemental form (usually HT) and an oxide form (usually tritiated water – HTO). Tritiated water poses a much greater radiological risk than in the elemental form: 1) because water vapor is much more easily taken into the body than elemental tritium and 2) because tritiated water and water vapor readily enter into the food chain, while elemental tritium does not.

From the Phase II data, it was assumed for Phase III that all tritium released into the air from SRS was either in an elemental (HT) or oxide (HTO) form. Most tritium releases occurred from the reactor areas and from the tritium facilities located in the separation areas. Smaller quantities of tritium were released from other areas such as D-Area.

Reactor areas. In other SRS reports and studies, tritium losses from reactor areas were estimated to be 100% in the oxide form (10), (11), (12). Westinghouse (13) has indicated that releases from reactors were due to evaporation of tritiated water and virtually all evaporated waste was released into the air through the reactor facility stacks. Balancing this with other information,[6] it was assumed that all tritium released from the five reactors was in the oxide form.

Tritium was also released into the air from evaporation of water from the reactor seepage basins. Tritium released from the reactor seepage basins was assumed to be in the oxide form.

Separations facilities. In the SRS separations facilities, the chemical form of the released tritium depended on the nature of the work performed at the facilities. The chemical form of the released tritium was determined by using information from Murphy (14) and the inadvertent release and incident histories summarized in the Phase II report. Specific information on the tritium release form was often provided for acute and unplanned accidental releases, especially for incidents of tritium release exceeding 700 curies. Each year, the activity from all acute or unplanned releases having known chemical forms was

[5] Other radionuclides were not partitioned by chemical form. Although health effects may depend upon "the lung clearance class" for other radionuclides, only a single class was considered for each of these other radionuclides. In addition, the model for uptake of tritium by biota depends on the chemical form.

[6] For example, in 1992, 65.90 curies of tritium were released from K reactor in elemental form compared with 39,200 curies in oxide form. The elemental form release accounts for less than 0.2 % of the total tritium release

subtracted from the total tritium activity released for that year. The remaining activity was assumed to be split evenly between elemental and oxide forms, based on data from Murphy (14) suggesting that about 50% of the tritium release was in an oxide form. Then the activity whose chemical form was known from the acute or unplanned releases for that year was appropriately added to the activity in each of the above splits.[7]

Similar to the reactor area seepage basins, small quantities of tritium were released into the air from evaporation of the separation area seepage basins. The evaporated tritium was assumed to be in the oxide form.

Other SRS areas. Small tritium releases have been documented in Cummins, et al. from A Area, D Area, M Area, and CMX-TNX (3). These releases were assumed to have been all in oxide forms.

5.1.8.2.2 Iodine

The fission processes in nuclear reactors were the primary sources of radioiodine production at SRS. The chemistry of iodine in nuclear facilities and the environment is complicated. The most reactive form of iodine is elemental iodine (I_2). This form of iodine has a relatively high deposition rate. It is important for assessing exposures to the thyroid through the food chain. Under favorable situations (e.g. the existence of other reactive elements, and the presence of catalysts), elemental iodine can combine with other elements and form less reactive gases such as HI, HOI and IO_3, or react with organic compounds forming even less reactive organic iodides like methyl iodide (CH_3I). Iodine existing as an organic molecule contributes to exposure by inhalation but is relatively unimportant for food chain pathways. Radioiodine can also associate with particles that can lower its reactivity. The gaseous radioiodine compound can be absorbed by non-radioactive particulate matter as well.

Two isotopes of iodine were considered in Phase III: ^{131}I and ^{129}I. Annual ^{131}I releases were monitored for the H- and F-Area separations facilities, reactors, and other facilities, mainly SRL. Annual ^{129}I releases were monitored for only the separations areas.

The Phase II report addressed the chemical form of the released iodine. Phase II release estimates for ^{131}I from the F- and H-Separations areas were apportioned between elemental iodine (I_2) and organic iodine (Phase II). Phase II did not similarly partition release estimates for ^{129}I from the separations areas, although one might expect a similar distribution. For reactors, Phase II cited measurements of iodine released at commercial pressurized water nuclear reactors by Pelletier, et al. (15). From this reference, Phase II estimated that "about 30% of the total would be elemental iodine and that the remainder would be divided between less reactive inorganic forms (~40% of total) and organic iodides (~30% of total)."[8] For ^{131}I releases from SRL, it was suggested in Phase II (p. 4.2-27) that "most of the radioiodine released from the shielded cells was probably in an elemental form" (Phase II).

For Phase III, the released quantities of iodine into the air were split between elemental and organic iodine in a manner consistent with the information in Phase II and summarized in Appendix B. However, data indicate that the chemical forms of released iodine change as the iodine is transported through the air (16). Therefore, the computations in Phase III for transport, dispersion, and deposition of iodine were

[7] As an example, assume that the total tritium release from the separations facilities in a given year was 1000 Ci, and this release was in an unspecified elemental and oxide form combination, although there was reason to believe that about half was either form. That year, 200 Ci of tritium was known to have been inadvertently released in an elemental form and 300 Ci of tritium was released in an oxide form. The 1000 Ci of tritium released that year was assumed to consist of 200 + (1000 – 200 – 300)/2 = 450 Ci of tritium in an elemental form and 300 + (1000 – 200 – 300)/2 = 550 Ci in an oxide form.

[8] Essentially, the Pelletier data indicated that iodine was released in a highly reactive form, a moderately reactive form, and a less reactive form. For the sake of conservatism, the moderately reactive and highly reactive forms could be grouped rather than the moderately reactive and the less reactive forms, leading to a split of about 70% elemental and 30% organic.

performed assuming that the iodine was a mixture of chemical forms – i.e., 40% particulate, 30% reactive gas (elemental), and 30% nonreactive gas (organic).[9] These fractions were built into the GENII code by its creators based on their experience with modeling iodine releases (17). The manual for the RATCHET[10] computer code, which was used for the thyroid dose reconstruction work for the Hanford Reservation, observes the following regarding the transport and mid-air partitioning of elemental iodine (16):

> *In the time that it took the iodine to travel 3200 meters (2 miles), about two-thirds of the iodine had changed form. Approximately one-third of the iodine was in organic species and the remaining third was associated with particulate material. The partitioning of iodine at 3200 in Ludwick's experiments is consistent with the results of the measurement of iodine in plumes from the stacks at the Hanford Site…with the partitioning of iodine in the plume following the Chernobyl reactor accident.*

5.1.9 Compare Final Source Term to Phase II Level 1 (Step 8)

To assure consistency between the Phase II release estimates and the Phase III source term, the total release of each radionuclide over 39 years, as determined in Phase III, was compared with the 36-year total release estimate used in Phase II for the screening assessment. This comparison is shown in Table 5-9. The information herein cited for Phase II was obtained from the Excel spreadsheets included with the Phase II report. As discussed in greater detail in Appendix C.4, the source term used for Phase III is generally consistent with Phase II. Some groups of isotopes, however, deserve comment:

^{241}Am: The citation of zero for Phase II in Table 5-9 reflects the fact that a 36-year screening total was not provided in the Phase II Excel spreadsheets for ^{241}Am. However, other entries in this table suggest that the value used in Phase II may well have been 5.60×10^{-03} Ci over 36 years, which is only slightly smaller than the 39-year total for Phase III of 5.88×10^{-03} Ci.

Plutonium isotopes: The Phase III source term is slightly larger than the Phase II release estimates because unidentified alpha activity was assumed to be ^{239}Pu. Because the unidentified alpha activity only summed to 0.08 curies, this adjustment to ^{239}Pu was not a significant change to the Phase II release estimate.

^{89,90}Sr: The Phase III source term for ^{89,90}Sr increased by 45 curies over the Phase II estimates because unidentified beta-gamma activity was assumed to be ^{90}Sr.

Uranium: The 39-year total uranium value used for Phase III is larger than the 36-year Phase II screening value. After performing the screening assessment, the Phase II study increased the estimated releases of uranium from SRS over that reported by SRS to account for sampling and measurement inefficiencies (see Chapter 4.4 of the Phase II report). The Phase III source term reflects these increased Phase II estimates as well as releases during the years 1990 through 1992.

[9] Data files containing annual releases of elemental and organic forms of 131I were input into the pre-processor and added. The total 131I was then split between chemical forms in the ratios of 40% particulate, 30% elemental, and 30% organic. The revised files from the preprocessor were then input to GENII for the air transport, deposition, and radiation exposure computations. See Appendix G for details.

[10] RATCHET: Regional Atmospheric Transport Code for Hanford Emission Tracking (16).

Table 5-9 Comparison of 39-Year Phase III Total Releases (Ci) to 36-Year Phase II Releases Used for Phase II Screening Assessment (Ci)

Radio-nuclide	Phase III Total from All virtual Sources	Phase II Total	Difference (Phase III – Phase II)	Percent Difference (Phase III –Phase II) x 100/Phase III	Note
^{241}Am	5.88×10^{-3}	0	5.88×10^{-3}	+ 100	X
^{41}Ar	$6.36 \times 10^{+6}$	$6.36 \times 10^{+6}$	$- 1.00 \times 10^{+3}$	- 0.02	
^{14}C	$2.97 \times 10^{+3}$	$3.00 \times 10^{+3}$	- 30.0	- 1.01	
^{137}Cs	3.51	3.51	3.72×10^{-3}	0.11	
^{3}H	$2.51 \times 10^{+7}$	$2.69 \times 10^{+7}$	$-1.80 \times 10^{+6}$	- 7.17	
^{129}I	5.67	5.65	2.20×10^{-2}	0.39	
^{131}I	$4.91 \times 10^{+4}$	$4.90 \times 10^{+4}$	$5.42 \times 10^{+1}$	0.11	
^{238}Pu	2.08	2.08	4.8×10^{-3}	0.23	X
^{239}Pu	13.2	13.14	6.03×10^{-2}	0.46	X
^{103,106}Ru*	$1.58 \times 10^{+2}$	$1.58 \times 10^{+2}$	-0.31	- 0.20	
^{89,90}Sr*	47.0	1.54	45.5	96.73	X
Uranium*	3.51	1.00	3.29×10^{-2}	0.94	X

*In the Phase II study, Ru-103,106 was screened assuming that all activity was Ru-106, Sr-89,90 activity was screened assuming that all activity was Sr-90, and uranium activity was screened assuming that all activity was U-235. For Phase III, activity reported as uranium was apportioned among the uranium isotopes U-234, U-235, U-236, and U-238.

5.2 Phase III Source Term by Groups of Radionuclides

This section contains summaries of the assumptions and procedures used to develop the source term for each radionuclide released into the air and listed in Table 5-9.

Tables of the annual quantities of these radionuclides released from each of the four virtual sources identified in Section 5.1.6 are provided in Appendix B. Uncertainties associated with these release estimates are addressed in Chapter 12.

5.2.1 Tritium

The main sources of tritium release to the air from SRS were the reactors and the separations areas. Phase II provided tritium release estimates for reactors and the separations areas for almost the entire period of interest. There is good agreement between the Phase II release estimates and SRS data for the reactor and the separations areas (Phase II.). For Phase III, however, sources were added that were not addressed in detail in Phase II. These sources included tritium that evaporated annually from the reactor and separations areas seepage basins, as well as annual releases from the A-, M- and D-Areas, SRL, CMX-TNX. The information on annual release of tritium from these additional areas, facilities, and seepage basins was obtained from Cummins, et al. (3).

Seepage basin evaporation data in Cummins, et al. were presented as totals for all reactor and separations areas seepage basins (3). Annual quantities of tritium evaporated from the reactor seepage basins had to be apportioned into two virtual groups of reactor facilities consistent with Table 5-6. It was assumed for

Phase III that evaporation from the seepage basins was linked to the tritium inventories that were discharged to the reactor seepage basins in liquid form. Since tritium losses by evaporation are expected to be proportional to the concentration of tritium in the seepage basin, assuming that these releases to air are proportional to the tritium inventory discharged to the basin, conservatively neglects changes in concentration from seepage of tritium into the ground; nevertheless, this seepage of tritium is then accounted for in the liquid source term. For each year, the total tritium inventory that was released into all reactor seepage basins was determined. The fractions of this total tritium inventory that were released into each of two groups of reactor seepage basins was also determined (C-, K-, and L-seepage basins in one group and P- and R-seepage basins in another). Then these fractions were assumed for the annual release of evaporated tritium into the air from each group of reactor seepage basins. These two fractions were multiplied by the total annual amount of tritium evaporated from reactor seepage basins to arrive at annual releases from both virtual groups of reactor facilities.

In addition, for Phase III, environmental report data for 1990 through 1992 was used rather than the reactor estimates for these years from Phase II. The combined totals from these environmental reports are somewhat larger than those listed in Phase II. This may be due to limited data in early studies. For these years, tritium release data were reported in the SRS environmental reports as combined releases from all reactors (4), (5), (6). Information from Phase II was used to group the tritium released from all reactors into two virtual groups of reactor facilities.

5.2.2 ^{131}I

As noted in Section 5.1.8.2.2 annual ^{131}I releases were determined in this Phase III study for the H- and F-Area separations facilities, reactors, and for SRL, and were grouped into the four virtual sources summarized in Table 5-6. Annual release information for the years 1954 through 1989 was obtained for these facilities from Phase II. For the separations areas and reactors, the median values from the Excel spreadsheets published with the electronic version of Phase II were used. Annual release information for the years 1990 through 1992 was obtained from environmental reports (4), (5), (6). Information from (3) was used to partition the ^{131}I estimates from Phase II, which are given for all reactors, into two virtual sources consistent with the reactor groupings in Table 5-6.

5.2.3 ^{41}Ar

^{41}Argon is an activation product that was created in large quantities during reactor operations. It is a noble gas having a short (1.83 hours) half life.

For Phase III, as in Phase II, all ^{41}Ar released into the air was assumed to come from the SRS reactors. Information from Phase II, from Cummins, et al. (3), and from Savannah River environmental reports (4), (5), (6) for the years 1990 through 1992 was used to partition ^{41}Ar release from the five reactors into the two reactor virtual source groupings listed in Table 5-6.

5.2.4 ^{241}Americium

For Phase III, annual ^{241}Am release data were obtained for the years 1977 through 1989 from Cummins, et al. (3). This reference identified ^{241}Am only for the F- and H-Separations Areas. For the years 1999 through 1992, additional data were available from SRS environmental reports (4), (5), (6) and from (18).

5.2.5 ^{14}C

For Phase III, estimates of ^{14}C released into the air were made for the separations areas and for the reactors. For the separations areas, data were available from Cummins, et al. for the years 1955 to 1989 (3). Data from SRS environmental reports were used for 1990 through 1992 (4), (5), (7).

For the reactors, data were available from Cummins, et al. for the following time frames (3):[11]

- C-Reactor: 1955 through 1985.
- K-Reactor: 1955 through 1988.
- L-Reactor: 1955 through 1968, 1987, and 1988.
- P-Reactor: 1955 through 1988.
- R-Reactor: 1955 through 1964.

After 1988, information on ^{14}C release from nuclear reactors was available in the SRS Environmental Report for 1992 as summed over all reactors (6). For 1992, the ^{14}carbon released from the reactors was apportioned the C-, K-, L-, and P-Reactors in equal fractions.[12]

5.2.6 ^{137}Cs

For Phase III, annual releases of ^{137}Cs were estimated for all years from the separations areas and for a few years from the reactors.

Most of the SRS atmospheric release of ^{137}Cs occurred from the F- and H-separations Areas (Phase II, 3). To develop source terms for ^{137}Cs release from the separations areas, information from these references was used for the years 1954 through 1989, and data from SRS environmental reports was used for the years 1990 through 1992 (4), (5), (6).

For reactors, ^{137}Cs release was not reported for the years 1954 through 1989 (3). ^{137}Cs releases from reactors were reported, however, in SRS environmental reports for the years 1990 and 1992 (4), (6). These releases were small (on the order of 0.00002 Ci in 1990 and 0.00003 Ci in 1992), but were added to the source term for completeness. For 1990, the released activity was apportioned equally among C-, K-, L-, and P-Reactors. For 1992, the activity was all released from L- and K-Reactors. This was done because for 1992, data were available that apportioned the total released ^{137}Cs activity (3.05×10^{-5} Ci) between these two reactors (7).

5.2.7 ^{129}I

For Phase III, consistent with the information in the Phase II study and in Cummins, et al., it was assumed that all release of ^{129}I to the air came from the separations areas (Phase II, 3). For Phase III, the annual ^{129}I release estimates from the Phase II study were compared with the smaller annual release estimates provided in Cummins, et al. (3). For the Phase III study, the Phase II estimates were used rather than the Cummins, et al. estimates. As discussed in Chapter 4 of the Phase II report, the historical SRS reports may have underestimated the true release because of sampling line loss, sampler collection efficiency, and counting errors (Phase II).

For 1990 through 1992, annual release estimates from SRS environmental reports were used for the separations areas (4), (5), (6).

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[11] Recall that R-Reactor shut down in 1964, C-Reactor shut down in 1985, P-Reactors shut down in 1988, and that L-Reactor was shut down in 1968, but was restarted in 1985 and shut down again in 1988. K-Reactor was shut down in 1988 but briefly operated in 1992 (Phase II).

[12] Release from R-Reactor was assumed to have been much smaller for the years 1990-1992, compared with other reactors, because it had permanently shut down much earlier (at least 21 years) than did the other reactors. Because of uncertainty about the fraction of the total release that should have been allotted to each of the other four reactors, the 1992 ^{14}C release (0.183 Ci) was apportioned equally among the four reactors. Hence, 75% of the ^{14}C was released from the C-, K-, and L-Reactor group, and 25% was released from the P- and R-Reactor group.

5.2.8 $^{103, 106}$Ruthenium

For Phase III, annual release estimates for ^{106}Ru were made for the F- and H-Separations Areas. For the years 1955 through 1989, release data were obtained from an Excel worksheet linked to the electronic version of Phase II (Phase II). For the years 1955 through 1968, data were provided as combined ^{103,106}Ru for both the F- and H-Areas. For the years 1969 through 1989, data were separately provided for ^{103}Ru and ^{106}Ru (Phase II).

For Phase III, releases documented in Phase II for the years 1955 through 1989 of ^{103}Ru, ^{106}Ru, and combined ^{103,106}Ru were assumed to be entirely ^{106}Ru. This assumption was consistent with that used in the Phase II screening analysis (Phase II) and provides conservative estimates of dose by inhalation and ingestion. Releases of ^{106}Ru for the years 1990 through 1992 from the separations areas were obtained from SRS environmental reports (4), (5), (6).

The Phase III source term also includes ^{106}Ru release from the reactor areas for the year 1990. This release was documented in the SRS Environmental Report for 1990 (4). Although the release was small (about 0.00003 Ci), it was included for completeness. This release was apportioned among the reactors.

5.2.9 ^{89}Strontium and ^{90}Strontium with Unidentified Beta-Gamma

For the Phase III study, ^{89}Sr and ^{90}Sr were anticipated to have been released into the air from the separations areas. ^{90}Sr was anticipated to have been released into the air from the reactors and from the A-Area.

Separations areas. Annual release data for combined $^{89, 90}$Sr from the F- and H-Separations Areas were provided in the Phase II report for 1954 – 1989 (Phase II). In Phase III, the combined ^{89,90}Sr activity was apportioned into 75% ^{89}Sr and 25% ^{90}Sr (see Section 5.1.8.1). To this activity was added the activity that was reported through 1989 in Cummins, et al. as unidentified beta-gamma activity (3). All this unidentified activity was assumed to be ^{90}Sr (see Section 5.1.8.1).

^{89}Sr and ^{90}Sr activity was assigned to the years 1990 and 1991 using information from SRS environmental reports (3), (5). For these two years, data were reported in these two references as combined ^{89,90}Sr, including unidentified beta-gamma. This combined activity was assumed to be all ^{90}Sr. For 1992, Arnett, et al. reported 1.62×10^{-3} Ci of combined $^{89, 90}$Sr with unidentified beta-gamma activity (6). A second reference reported 2.75×10^{-4} Ci of combined $^{89, 90}$Sr activity without referencing unidentified beta-gamma activity (6). The difference was 1.35×10^{-3} Ci. The activity (1.62×10^{-3} Ci) reported by (6) was partitioned between ^{89}Sr and ^{90}Sr in a 75%-25% ratio. The difference (1.35×10^{-3} Ci) was assumed to be ^{90}Sr.

Reactors. For reactors, ^{90}Sr release data were not provided in the Phase II report (Phase II). However, for the years 1954 through 1989, annual unidentified beta-gamma activity was released from reactors and reported in Cummins, et al. (3). This activity was assumed for Phase III to be ^{90}Sr. Reactor-specific annual ^{90}Sr estimates were apportioned using Cummins (3) data into two virtual source groups consistent with Table 5-6. For the years 1990 through 1992, the release data obtained from environmental reports for the reactor areas were equally apportioned into C-, K-, L- and P-Reactors.

A-Area. For the years 1954 through 1989, annual releases from A-Area that were identified in Cummins, et al. as unidentified beta-gamma activity (3) were assumed to be ^{90}Sr. For the years 1990 through 1992, SRS environmental reports were used to develop ^{90}Sr release estimates from A-Area (4), (5), (6).

5.2.10 Uranium and Plutonium with Unidentified Alpha

For Phase III, annual estimates of uranium released into the air were made for the F- and H-Separations Areas and for M-Area. Annual estimates of plutonium released into the air were made for the separations areas, the reactors, and for A-Area.

Separations Areas. The Phase II release estimates were based on information from Cummins, et al. (3). However, the Phase II estimates were increased in earlier years, compared with that reported by Cummins, et al. (3), because of concerns about sampling uncertainties. For Phase III, the Phase II estimates of release of uranium and plutonium from the separations areas were enhanced using additional data from environmental reports.

For Phase III, uranium activity was apportioned into four uranium isotopes based on the isotopic partition assumptions summarized in Section 5.1.8.1 and Table 5-8. Note that uranium isotopic composition varied in the F- and H- Separations Areas, because the processes conducted in each area differed. Uranium data for Phase III were available for 36 years from Phase II (Phase II) and for three years from SRS environmental reports (4), (5), (6).

For the years 1954 through 1989, uranium released from the F-Separations Area was reported separately in Phase II from uranium released from the H-Separations Area. This information was sufficient to partition the uranium released from both the separations areas. But for 1990 through 1992, information about uranium released from the separations areas was provided in SRS environmental reports (4), (5), (6) only as combined releases from both areas. Isotopic partitioning of uranium was performed for these years using an SRS report for the year 1992 that provided uranium release data separately for F- and H-Areas (6). This reference indicated that 95% of the uranium in 1992 was released from the F-Area and 5% from the H-Area (6). It was assumed that this 95%-5% uranium split between F- and H-Areas was also applicable to 1990 and 1991. The total uranium released from each separations area for these years was apportioned into isotopic distributions consistent with Table 5-8. Then the total release of each uranium isotope from both separations areas was determined for purposes of the virtual source grouping. Finally, release data for each uranium isotope in F- and H-Area were increased for Phase III using the same method described in Chapter 4 of Phase II to compensate for sampling line loss.

For plutonium, Phase II increased its estimates of plutonium released into the air, relative to that reported by SRS, to compensate for sampling line loss. The source term adjustment in Phase II for plutonium air release was done for ^{238}Pu and for ^{239,240}Pu together. Because sampling line loss should have been the same for any plutonium isotope, for Phase III the procedure cited in Phase II to compensate for sampling loss was applied to both ^{238}Pu and ^{239}Pu. The total plutonium activity reported in Phase II before adjustment matched the calculated plutonium term in Cummins, et al. (3). Using the isotopic information provided in (3), plutonium reported as total plutonium in Phase II was apportioned for Phase III into ^{238}Pu and ^{239}Pu.

Cummins, et al. reported unidentified alpha activity released from the F- and H-Separations Areas (3). For Phase III, this unidentified activity was included as ^{239}Pu. The activity was increased in Phase III, relative to that reported in Cummins (3), to compensate for sampling line loss. As discussed earlier, the inclusion of this unidentified activity as ^{239}Pu did not significantly change the plutonium source term.

Reactors. Annual estimates of ^{239}Pu released from the nuclear reactors were made for Phase III using information from Cummins, et al. for the years 1973 through 1989 (3). In this case, starting in 1973, Cummins (3) provided air release data for unidentified alpha activity for the reactor areas. For Phase III it was assumed that this alpha activity was ^{239}Pu. For 1990 through 1992, information from SRS

environmental reports was used (4), (5), (6). No estimates were made for uranium release from reactors, because such releases were not identified in Phase II, Cummins (3), or other documents.

M-Area. Annual estimates of uranium isotope releases from M-Area were made in Phase III using information from (3). This reference listed annual uranium releases (as U-Nat – i.e., natural uranium) from 1975 through 1989, except that no releases were listed for 1981 and 1984. This activity was apportioned among the isotopes ^{234}U, ^{235}U, and ^{238}U. Releases for the years 1990 through 1992 were estimated using information from SRS environmental reports (4), (5), (6).

A-Area. Cummins, et al. reported annual release of unidentified alpha activity from A-Area for most of the years from 1961 through 1992 (3). No release information was provided for 1964-1967, 1972, 1979, 1981, 1982, 1984, and 1985. This unidentified alpha activity was assumed to be ^{239}Pu for Phase III. Information from environmental reports was used to complete the estimates for 1990 through 1992 (4), (5), (6).

5.3 Annual Release of Tritium, ^{131}Iodine, and ^{41}Argon

As will be seen in Chapter 11, radiation doses calculated from the air pathway are dominated by tritium, ^{131}I, and ^{41}Ar. To help shed light on these calculated doses, the source term assumed in Phase III for these radionuclides is graphically depicted in the Figures below. The annual release of tritium to air from the four virtual sources used in Phase III is shown in Figure 5-3. The bulk of the tritium released over the 39 years of nuclear materials production came from the F-and H-Separations Areas (Virtual Source 2) rather than from the reactor areas. Annual releases of ^{131}iodine from all four virtual sources are shown in Figure 5-4. Note that the bulk of the ^{131}I was released from the separations areas (Virtual Source 2), and that the largest ^{131}I release from this virtual source occurred in 1956. (Also note that the y-axis of this graph (Curies of ^{131}I released) is in a logarithmic scale.) Annual releases of ^{41}Ar from the four virtual sources are shown in Figure 5-5. Although ^{41}Ar releases rise and fall over the years, one can see a succession of peaks that gradually diminish in height. The largest peak occurred over roughly a five-year period between 1958 and 1963. Successively smaller peaks occurred in the years 1967 and 1968, in 1973, and in 1986.

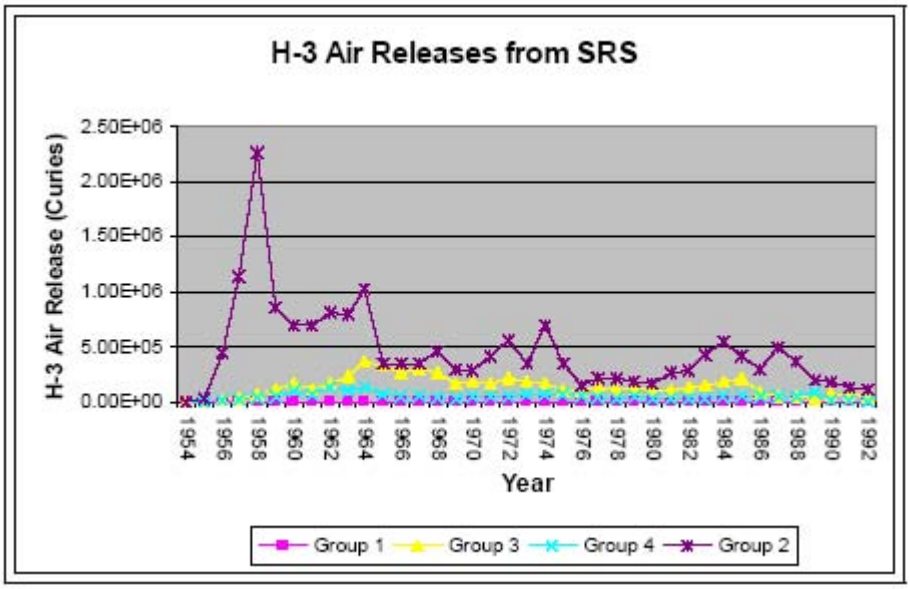

Figure 5-3 Tritium Release to Air from SRS

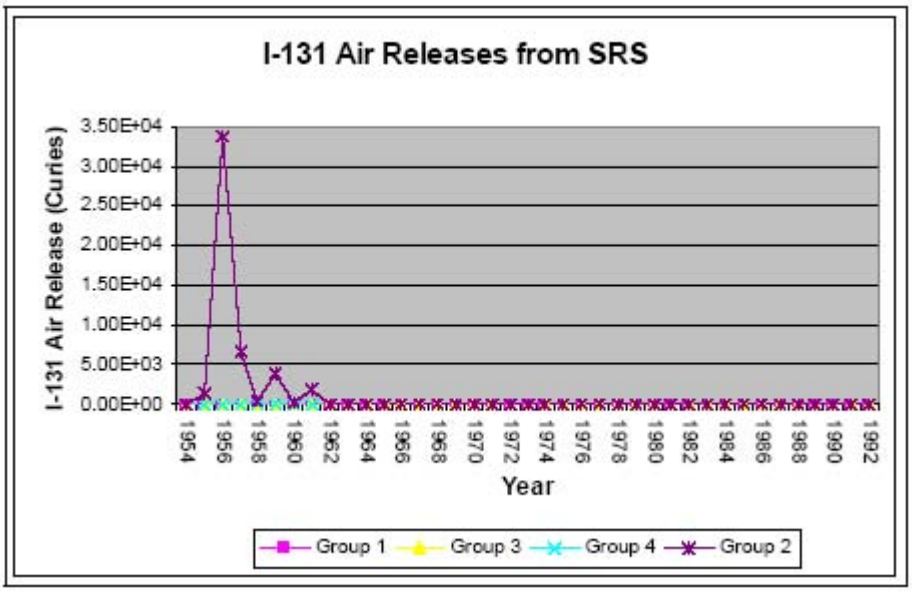

Figure 5-4 Release of ^{131}I to Air from SRS

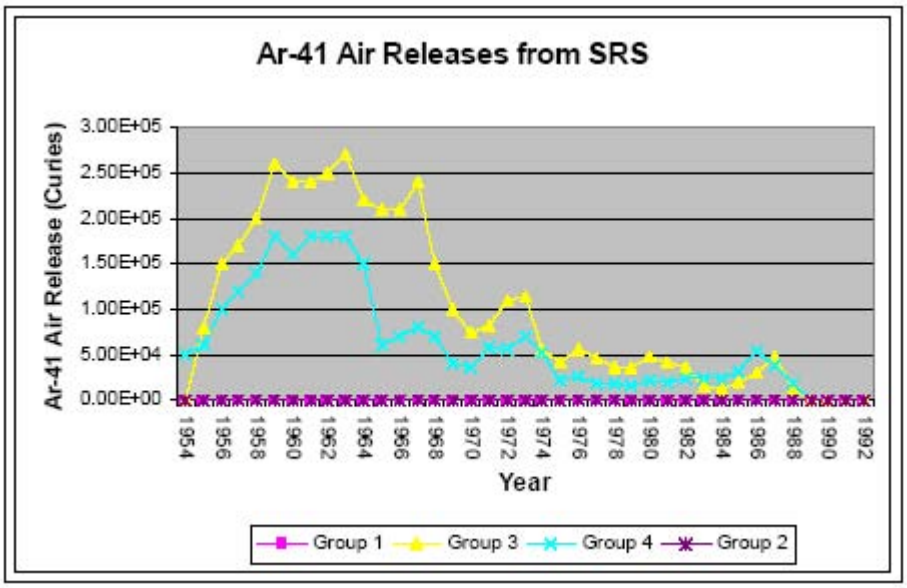

Figure 5-5 Release of ^{41}Ar to Air from SRS Reactors

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