CHAPTER 4.2

RELEASES OF RADIOIODINES AND BETA-GAMMA-EMITTING PARTICLES TO THE ATMOSPHERE

ABSTRACT

This chapter first discusses the releases of the most important radioiodine isotopes (^{131}I and ^{129}I) to the atmosphere from facilities at the Savannah River Site (SRS). The historic measurements at the facilities are interpreted in the context of current knowledge of radioiodine chemistry and the effectiveness of various sampling techniques. The analysis leads to revised estimates of radioiodine releases. These revised estimates, or radioiodine source terms, can be used to estimate thyroid doses to persons residing near the SRS. The fuel processing facilities in F-Area and H-Area were sources of the highest releases of radioiodines, which were discharged through tall stacks. The largest estimated releases occurred in 1956. Releases during the years 1955–1961 were significantly higher than the releases that occurred between 1962 and 1971. Those latter releases were, in turn, markedly greater than releases in the later 1970s and 1980s.

The latter part of the chapter addresses atmospheric releases of particles that contain beta-gamma-emitting radionuclides. These non-volatile fission and activation isotopes are generally associated with solid particles, in contrast to the radioiodines that are most likely to be released in gaseous forms. The principal radionuclides released were isotopes of ruthenium, cerium, cesium, strontium, zirconium, and niobium. As was the case for radioiodines, the most important release points were the stacks at the fuel processing facilities. Overall, releases of ^{103}Ru plus ^{106}Ru were largest. (Separate analyses were not performed for many years.) However, in some years, the pair of radionuclides, ^{95}Zr plus ^{95}Nb, was discharged in greater quantities. In one year, the highest release was of ^{137}Cs. Releases of ^{141}Ce plus ^{144}Ce and ^{89}Sr plus^{90}Sr were generally lower than those of ^{103}Ru plus ^{106}Ru and ^{95}Zr plus ^{95}Nb.

INTRODUCTION

Radioactive isotopes of iodine and other beta-gamma-emitting radionuclides are produced by fission of uranium and plutonium in reactor fuels and by neutron activation of both radionuclides and stable isotopes in the fuel. As is discussed in the following sections, many such radioisotopes are produced, but only a small number are important for environmental dose assessment. Because most of the releases of radioiodines and beta-gamma-emitting particles occurred when the fuel was processed (usually many days after removal from the reactor), the radionuclides of greatest interest in this context are those that are produced in relatively large quantities and have half-lives that exceed 1 day. This is illustrated in the discussion of the radioiodine releases.

RELEASES OF RADIOIODINES

As a general term, “radioiodines” refers to the entire set of radioactive isotopes of the element iodine. Many such isotopes, with mass numbers between 128 and 142, are produced in nuclear reactor fuel by fission and activation. However, in this report we focus primarily upon those isotopes whose half-lives are long enough that significant amounts can be present at the time of fuel processing. The first of these is ^{131}I; its half-life is 8.04 days and it is produced in large quantities (tens of millions of curies) in a load of reactor fuel. Other radioiodine isotopes, with half-lives ranging from about 1 hour to 1 day (^{132}I, ^{133}I, ^{134}I, and ^{135}I), are produced in similarly large amounts during reactor operation. However, they decay in the fuel quickly following reactor shutdown and much smaller quantities are available for release when the fuel is processed. A number of radioiodines have even shorter half-lives and their decay in the fuel is correspondingly more rapid. The second isotope of interest here is ^{129}I, which has a half-life of about 16 million years. Although only a small quantity (less than 1 curie) of ^{129}I is produced in the normal life of a reactor fuel core, the amount produced has not been diminished by decay since the start of SRS operations. The ^{129}I that was released from SRS facilities remains as part of the pool of environmental ^{129}I, but the ^{131}I that was released decayed to low levels within about two months.

The following section contains a brief introduction to some aspects of the chemistry of radioiodine. This information is important to understanding the importance of radioiodine releases and their assessment. Then, the methods and problems of sampling radioiodine releases are discussed. Historic estimates of radioiodine releases are presented and the rationale for revising those estimates is described. Finally, revised estimates of the amounts released are presented; these are the so-called source terms for radioiodines.

Sources of Radioiodine at the Savannah River Site

Radioiodines are produced by nuclear fission and activation processes in nuclear reactor fuel. Thus, the several reactors at the Savannah River Site (SRS) were sources of radioiodine releases. Because reactor fuel rods are surrounded by a cladding designed to contain the fission and activation products and the rods are stored in cooling water, atmospheric releases from those facilities were limited. The reprocessing facilities, where the cladding and the reactor fuel itself were dissolved, were more important sources of gaseous fission product releases. Research facilities of the Savannah River Laboratory (SRL) were a third source of radioiodine releases. These facilities were used for pilot studies and research; some work involved irradiated fuel or target elements containing radioiodines.

All these facilities are shown in Chapter 2, Figure 2-3. The reprocessing facilities are located in the 200 Areas (F-Area and H-Area) near the center of the SRS. The P, L, K, R, and C Reactors are also fairly centrally located; the areas were named 100-P, 100-L, etc. The SRL facilities are located in the Administrative Area (A-Area) near the northwestern boundary of the SRS.

This chapter focuses on the reprocessing facilities, which were the sources of the highest releases of radioiodines identified by Kantelo et al. (1993). The release point at each of these plants was a tall ~61-m (200-ft) stack, designated 291-F and 291-H, respectively. Gases that were carried from the dissolver vessels were treated to reduce the radioiodine concentrations; however, radioiodines were evolved from liquids in other parts of the facilities as well, and those gases were also carried to the tall stacks. Sand filters used to reduce releases of particulate radionuclides did not have a large effect on the quantities of ^{131}I and ^{129}I released. For the reactors, the main atmospheric discharge points were also tall stacks. Several stacks of differing heights served SRL facilities.

Radioiodine Chemistry

The chemistry of iodine in nuclear facilities and in the environment is complex and not fully understood. We discuss here only basic features that are important to estimating radioiodine releases from nuclear facilities and to understanding the principal human exposure pathway. Iodine is one of the halogens, a series of reactive elements that also includes fluorine, chlorine, and bromine. Iodine is an essential element in human nutrition and is primarily located in the thyroid gland. Radioactive iodine, such as ^{131}I, is chemically the same as stable iodine (^{127}I) and, when ingested or inhaled, also is concentrated in the thyroid.

Relationship between Chemical Form and Deposition on Surfaces

The reactive nature of elemental iodine gas, which is composed of two iodine atoms and is symbolized by I_2, makes it likely to deposit on surfaces. Examples are (a) walls of vessels in a nuclear facility and (b) vegetation in the environment. Deposition on vegetation is the first step in the pathway that leads from iodine in air to iodine in cows' milk and subsequently to irradiation of the thyroid glands of consumers of contaminated milk. That sequence of transfers is often referred to as the air-grass-cow-milk exposure pathway. (For the small fraction of the population that consumes goats' milk, the pathway is basically the same, but the milk source differs.) Deposition of elemental iodine on the interior wall of a sampling line can lead to underestimating the amount of radioiodine that was released from the stack being sampled. Loss of the elemental iodine from the sample air stream means that the concentration measured in the sample will be lower that the concentration that was present in the stack exhaust air.

When iodine combines with other elements, the resultant compounds are generally less reactive than elemental iodine. Hydrogen iodide (HI) is the most reactive of such compounds. Iodine may be a component of a radioactive particle (firmly bound within it) or sorbed on the surface of a non-radioactive particle. Iodine may combine with other gases, such as oxygen, nitrogen, and organic compounds, to form less reactive gases. Least reactive of the variety of iodine compounds are the organic iodides. Methyl iodide, symbolized by CH_3I, is the most common example of this group. These compounds are formed in nuclear facilities and comprise part of the radioiodine releases from them. Organic iodides are much less likely to deposit on surfaces in the environment; as a result, they contribute much less to human thyroid doses from the air-grass-cow-milk transport pathway. Their lack of reactivity also means that techniques that use chemical reactions to trap elemental iodine in an air sample will not successfully collect organic iodides in the same sample.

There are forms of iodine whose reactivity is intermediate between those of elemental iodine and organic iodides. Iodine associated with particles is one; others are gaseous compounds. A number of inorganic iodine compounds, including HOI, have been postulated; however, firm definition of the intermediate reactivity fraction has proved elusive. Knowledge of these compounds is limited by sample collection methods that have been devised to isolate them and by a lack of understanding of the reactions that produce them. Voillequé (1979) provided a summary and discussion of the sampling techniques that had been used for radioiodines and for stable iodine and the relationship between iodine species and environmental transport. More recently, the question of radioiodine species was revisited as part of a workshop conducted for the Centers for Disease Control and Prevention to address issues related to results of the dose reconstruction for Hanford releases (Grogan 1998).

The relationship between chemical form and deposition velocity, a measure of the air to surface transfer process, is summarized in Table 4.2-1. Because deposition of airborne iodine onto pasture grass is the first step in the air-grass-cow-milk pathway, the relative importance of a species for that exposure pathway is indicated by the magnitude of its deposition velocity. For that reason, the table is a useful guide to the relative doses received by the most highly exposed groups in a population. It should be noted that the table contains only representative values to show the relative ranking and that not all are based upon the same amount of experimental data. For example, elemental iodine has been studied more than any of the other compounds (e.g., Chamberlain and Chadwick 1953, 1966; Chamberlain 1960; Heinemann and Vogt 1980). The estimate for HOI is based upon a single laboratory experiment (Voillequé and Keller 1981). Deposition of radioiodine onto pasture grass in any particular circumstance depends upon several other variables in addition to the chemical form of the radioiodine.

Table 4.2-1. Approximate Relationships Between Chemical Forms and Deposition Velocities for Radioiodine

Chemical form	Deposition velocity for pasture grass (cm s^{-1})
Elemental iodine (I_2)	1
Other inorganic forms (such as HOI)	< 0.1
Iodine associated with particles	0.1–0.2
Organic iodides	0.0001–0.005

Although doses are generally much lower than those due to milk consumption, humans are also exposed to radioiodine by inhalation of contaminated air. For inhalation exposures, there are no great differences in the dose contributions from the various species. Doses from external exposure are also much lower than those following consumption of contaminated milk. Because external exposure depends in part upon deposition of radioiodine, elemental iodine makes the most important contribution to that pathway.

In summary, elemental iodine is the most important chemical form from the point of view of the air-grass-cow-milk (or air-grass-goat-milk) exposure pathway that leads to the largest thyroid doses in human populations. Because of the large differences in human exposure associated with the release of different radioiodine species, it is important to distinguish among them if possible. Information that can be used to make inferences about iodine species released at the SRS is discussed in the next section and in the later discussion of early sampling techniques at the SRS.

Measurements of the Forms of Radioiodine in Facility Effluents

The chemical forms of radioiodine in facility effluents were not and are not measured routinely at the SRS or at other Atomic Energy Commission (AEC)/U.S. Department of Energy (DOE) or commercial facilities. However, special studies at various locations have provided data on the chemical forms of radioiodine in releases from operating power reactors and reprocessing facilities.

In an early evaluation of a caustic and thiosulfate scrubber for radioiodine monitoring, Sill and Flygare (1960) found that 70% of the ^{131}I in effluents from the Idaho Chemical Processing Plant (ICPP) was not trapped by their system. Because the scrubber had been shown to be efficient for I_2, it was concluded that the effluent must contain chemical forms other than elemental iodine. Much later, Hetzer et al. (1980) found that the principal gaseous component of the ^{129}I discharge from the ICPP was organic iodides, which accounted for 60–80% of the total. The elemental iodine fraction ranged from 2–20% of the total, and a species of intermediate reactivity (identified as HOI by their species sampler) consistently accounted for about 20% of the radioiodine activity.

Measurements at two European reprocessing plants have been reported (IAEA 1980). At the Dounreay reprocessing plant in the United Kingdom, measurements indicated that about 70% of the radioiodine release was in organic form. At the Karlsruhe reprocessing facility in Germany, the organic iodide fraction of the ^{129}I discharge was found to be about 25%. The reasons why effluents from that facility had a smaller organic iodide fraction are not known, but they could relate to differing processing schemes or other factors.

Special investigations in similar facilities at the SRS and Hanford identified several organic iodides in atmospheric discharges using gas chromatography (Smith and West 1967; Haller and Perkins 1967). Methyl iodide, ethyl iodide, propyl iodide, and butyl iodide were identified, and it is possible that other longer chain compounds were also present. These studies did not quantify the fractions of various species in the total ^{131}I releases. Other investigations by Perkins (1964) at Hanford showed that the fraction not present as I_2 or HI (both of which were effectively captured by their scrubber) ranged from 50–70% of the total. Subsequent studies at the SRS found that the organic iodide fraction at the 200-H facility comprised 80–90% of the total ^{131}I. The predominant airborne organic forms were found to be methyl iodide and butyl iodide (Kantelo et al. 1993).

A series of investigations of the sources of ^{131}I in commercial power plant effluents yielded the bulk of the available information on radioiodine species for reactors. Data from nine boiling water reactors (BWRs) and ten pressurized water reactors (PWRs) were examined to develop composite species distributions for these two reactor types. The PWR species distribution is expected to be most applicable to the SRS production reactors, but there is not a great difference between those results and the composite distribution for BWRs (Keller et al. 1982). A small fraction (2%) of the released ^{131}I was associated with particles, 27% was elemental iodine, 31% was in organic form, and 40% was present as inorganic forms of intermediate reactivity (identified as HOI in the studies).

Sampling of Airborne Radioiodine

There are several aspects of successful sampling for contaminants in airborne effluents. An enumeration of these principles was published as an American National Standards Institute (ANSI) guide (ANSI 1969). This section briefly discusses issues that are relevant to understanding the effectiveness of the sampling for airborne radioiodines at the SRS.

A primary requirement of sampling is that a representative sample be withdrawn from the air being discharged from the stack. That is, the sample should faithfully show “the quality and characteristics of the entire volume” of the exhaust. If the discharge is not well mixed, this is difficult to accomplish. Because the airborne radioiodine is primarily in gaseous form, difficulties that affect extraction of samples of particles from the exhaust air stream are not highly relevant.

Following its extraction from the air being discharged, the sample is transported to the point of radioiodine collection from the sample stream. Ideally, the distance between the point of sample withdrawal and the point of collection will be short to minimize any losses due to deposition of radioiodine in the sampling line. The next steps are the capture of the radioiodine in the sampled air and the measurement of the collected radioiodine activity. These important issues are discussed in reverse order in the following sections.

Collection and Measurement of Radioiodine Samples

The earlier discussion of radioiodine species suggests that different chemical forms require particular attention when attempting to collect a sample of all airborne radioiodines or of the various forms individually. These sampling problems were not widely understood during the 1940s and 1950s and were not solved until the 1960s.

Based upon the Hanford experience, releases of radioiodine from the reprocessing facilities at the SRS were expected to be important and a stack monitoring system for ^{131}I was designed (Anonymous 1953a, 1953b; Gould 1953; Moison 1953; Carmichael, and Karraker 1955). The system employed a caustic scrubber to collect the ^{131}I in the sampled air. Its success depended upon a chemical reaction between the airborne radioiodine, presumed to be present as I_2, and the NaOH solution through which the air was bubbled. This sampler would not collect organic iodides; its efficiency for collection of inorganic forms other than I_2 and HI is uncertain. The NaOH solution was transported through a coiled line, which was monitored by a scintillation detector and analyzer focused on the main ^{131}I gamma ray. The output of the system, proportional to the ^{131}I release rate, was recorded in the control room. This real-time monitor could detect a release rate of about 1 Ci of $^{131}I_2$ per day. The caustic scrubber solution was also collected over a 24-hour period. Sampling and laboratory analysis of the solution provided a more sensitive, but not real-time, measure of ^{131}I releases. A release rate of about 2 mCi of $^{131}I_2$ per day could be detected in that way (Carmichael and Karraker 1955; Marter 1962).

Beginning in 1956, a second sampling system that employed particulate filters impregnated with silver nitrate ($AgNO_3$) was operated in parallel with the caustic scrubber stack monitor. These filters were replaced daily and counted in the laboratory to determine the ^{131}I content and obtain an estimate of the daily discharge of ^{131}I. The filter was tested in the laboratory using I_2; measured collection efficiencies routinely ranged from 85–95%, but they were as high as 100% under carefully controlled experimental conditions. Field tests in 1956 yielded filter collection efficiencies of 76–100% relative to a caustic scrubber (Du Pont 1956i, 1956l). Because collection of ^{131}I by these $AgNO_3$-impregnated filters also relied on a chemical reaction and binding of the ^{131}I to the silver in the filter, these filters also would not trap organic iodides. The collection efficiency of these filters for other inorganic forms is not known.

The effluent monitoring systems were thoroughly reviewed following the first widely discussed ^{131}I release from the SRS reprocessing facilities in May and June 1961 (Reinig 1961; Marter 1963). The investigators concluded that neither the caustic scrubber and monitor nor the $AgNO_3$-impregnated filters used to collect ^{131}I had performed well. Initial indications from the monitor that there was a high ^{131}I release rate were not considered credible because of previous errant indications by that system. However, later analysis of the amount of ^{131}I collected in the scrubber solution showed that the indications had been correct. Analysis of the data led to the conclusion that the collection efficiency of the $AgNO_3$-impregnated filters, relative to the caustic scrubber, had dropped to 32% and was well below the nominal value of 80% that had been assumed to provide cautious estimates of the releases (Reinig 1961; Marter 1963).

The releases at the end of May and in early June led to readily detectable contamination in milk in the environs of the SRS. See Chapter 10 of this report for a more detailed discussion regarding radioiodine concentrations in milk. There was a formal investigation of the event, which was reported to AEC Headquarters (Blair 1961).

A detailed investigation of the sampling and monitoring systems that were in use at the reprocessing facilities was conducted. As part of this effort, plant personnel evaluated alternative collection media for ^{131}I in the 200-F and 200-H exhaust stack sampling and monitoring systems. As a result of comparative testing, it was decided that charcoal cartridges would be used to collect ^{131}I in reprocessing facility effluent samples. Although not considered entirely satisfactory because of the heating produced by reactions of undefined oxides of nitrogen (NO_x) with the charcoal, this practice was started in the fall of 1961 (Marter 1962).

The investigations into effluent sampling produced a number of significant findings, some of which are summarized below. Others are discussed in the following two sections.

- Laboratory testing of the NaOH scrubber indicated an efficiency of 92% for I_2; however, daily comparisons against charcoal sampling of 200-H plant effluents yielded a 20-day average scrubber efficiency of only 20%. Because charcoal is a good collector for all iodine species, this comparison indicates that much (nearly 80%) of the ^{131}I in the effluent was not I_2.
- Two laboratory tests indicated a collection efficiency for I_2 of 85% for the $AgNO_3$-impregnated filters (compared with a caustic scrubber) that was not affected by a change in sampling flow rate from 472 to 944 $cm^3\ s^{-1}$ (1 to 2 $ft^3\ min^{-1}$). Field testing, also compared with a caustic scrubber, showed efficiencies of between 34 and 64%. In those tests, the collection efficiency was found to be inversely proportional to the sampling flow rate. Plant staff concluded that an efficiency of 32% was appropriate for the sampling flow rate during period of abnormal release in May–June 1961.
- Further field testing of $AgNO_3$-impregnated filters, compared against charcoal, showed filter collection efficiencies for ^{131}I that were much lower. For 28 days of sampling the 200-H exhaust and 25 days of sampling the 200-F exhaust, collection efficiencies averaged 1.3 and 4.9%, respectively. These results are again consistent with an effluent that is primarily composed of organic iodides and contains relatively little I_2.

Follow-up studies and measurement were reported by Smith and Jolley (1963). At the inlet to the stack, the $AgNO_3$-impregnated filters were only 8 percent efficient when compared against charcoal collectors. This finding supports the previous conclusion that most (~90% in this case) of the ^{131}I was not in elemental form. It is consistent with the earlier field efficiency measurements that employed those filters and with later measurements, at the SRS and elsewhere, that showed a high organic iodide fraction in reprocessing plant effluents. Although the indicated organic fractions for the SRS reprocessing facilities are higher than seen elsewhere, this might be explained by the fact that at SRS the effluent passed through a sand filter. That filter provided a large surface area for deposition of I_2 and subsequent chemical conversion to less reactive forms.

Jacobsen and Jolly (1963) reported efficiencies of 24 and 30 percent for two $AgNO_3$-impregnated filter papers exposed to process ^{131}I, but the sampling times were only 15 minutes. Efficiencies of two NaOH scrubber solutions for the same test conditions were 56 and 59 percent. For short-term samples the efficiency of activated charcoal exceeded 99 percent, but for 24-h samples in the presence of NO_2, or a combination of NO_2, NH_3, and NH_4NO_3, the average efficiency was about 88 percent (Jacobsen and Jolly 1963).

Sampling Line Losses for Radioiodines

The effluent sampling lines for the reprocessing facilities at the SRS were quite long. Samples were extracted at the ~59-m (195-ft) level of the stack. The line extended to near ground level and then an additional 30.5 m (100 ft) horizontally to the point of sample collection. The total line length, including the in-stack portion, is estimated to be about 93 m (~305 ft).

Table 4.2-1 also serves as a guide to the relative probability of losses due to deposition of gaseous iodine species in sampling lines. Although the numerical values of deposition velocity differ, the general relationships are the same. Thus, the large organic iodide fraction of the ^{131}I in the sample would be carried through the sampling line effectively, with little loss due to deposition on the walls of the line. The ratio of the organic iodide concentration leaving the line to that entering it (called the transmission factor for organic iodide) would be very close to the desired value of 1. However, based upon current knowledge, we expect that there would be significant deposition of the reactive iodine fraction (I_2) during passage through the line. A model of the behavior of radioiodine in sampling lines is discussed below.

Measurements at the SRS. Sampling line losses for I_2 were investigated by SRS staff following the elevated releases of ^{131}I in reprocessing facility effluents in May–June 1961. Testing was conducted in both the laboratory and in the actual sampling line in the field. Because the laboratory test employed only a very short (~1-m) piece of tubing, the field test is most relevant. Elemental iodine was released into the horizontal 30.5-m (100-ft) section of the sampling line and measured at the normal sample collection point. About 32% of the I_2 was initially lost because of deposition in the line. However, during the first day after the injection period, 7% of the deposited I_2 was resuspended from the walls of the line and collected at the sampling point. This amounted to an additional 2% of the amount injected, resulting in a net loss of ~30% in that section of line (Marter 1962).

The observed transmission factor for the test (0.7) can be extrapolated to the full length of the sampling line. The entire line is nominally three times longer than the section used for the test. The transmission factor for samples of I_2 carried through the entire line is estimated to be approximately 0.7^3 or ~0.34 for conditions comparable to those used for testing.

Two later tests involving injection of 1 and 6 mCi of $^{131}I_2$ into the stack did not yield detectable amounts of ^{131}I at the sample collection point. These unusual results do not seem to have been investigated further. It may be, as was speculated, that a combination of deposition in the stack and in the sampling line reduced the concentrations of ^{131}I below the level that was detectable.

In a later presentation, Jacobsen and Jolly (1963) reported rather different results for line loss. They performed experiments using a separate line running from the ~15-m (50-foot) level of the stack to the sample collection point in the fan building. Simultaneous measurements were made at both end of the line, which was about half as long as the normal sampling line, to estimate deposition losses. They found that NO_2 in the air stream greatly decreased the deposition loss, but that losses were higher when ammonia was also present to react with the NO_2. On the basis of their results, they estimated that typical losses would average only 10–15 percent.

Unfortunately, the document prepared for the oral presentation contains few details about the experiments. Information about the flow rate through the line, the concentrations of ^{131}I, NO_2, NH_3, and NH_4NO_3 measured during the tests, and the relationship of the levels of the non-radioactive constituents to routine operational conditions would be very useful, but has not been found. The results provided are difficult to interpret without the supporting data. They imply a transmission factor for the main sampling line in the range 0.7–0.8, which is quite different from the estimate of ~0.34 based on a test using a section of that line (discussed above).

Model of Radioiodine Transmission Sampling Lines. This section describes a model that has been developed over many years and is believed to reflect the principal processes that affect transmission of radioiodine through sampling lines. An early version of the model considered only deposition and resuspension and was consistent with experimental data that showed there was a delay in the movement of radioiodine through lines and a change in the activity balance from reactive species (I_2) to nonreactive forms like organic iodides (Pelletier et al. 1978a). Figure 4.2-1 shows a diagram of one segment of the current model. It includes a third process, fixation of some of the deposited iodine on the surface.

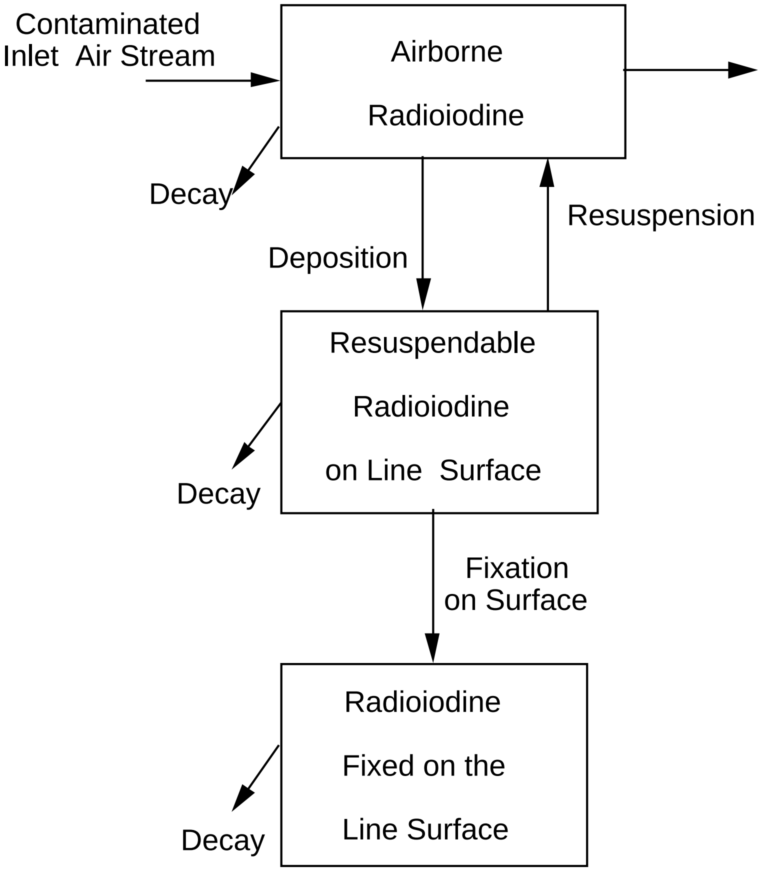

Figure 4.2-1. Model of radioiodine transport in a sampling line.

Typically, a sampling line is modeled as a sequence of 20 segments like the one shown in Figure 4.2-1. The airborne activity leaving one line segment becomes the input for the next segment of the model. Radioactive decay is shown as a removal process for all three compartments; however, it is important only for the radioiodine on surfaces.

Laboratory measurements of radioiodine deposition and resuspension in mock-ups of steel sampling lines (Unrein et al. 1985; Widner et al. 1985; Edson et al. 1987; Glissmeyer and Sehmel 1991) are the source of estimates of the deposition and resuspension rate constants. Kabat (1983) provides information for other surfaces. In addition to the SRS measurement described above, experimental studies of installed sampling lines were also conducted and documented in Ström and Hesböl (1978) and Curtis and Guest (1986). It was analysis of experimental measurements that led to the inclusion of the fixation process in the revised model, which greatly reduced the variability of the resuspension rate constants computed from the data. Voillequé (2001) describes the model and derivation of parameter values in more detail.

Three differential equations describe the transport of radioiodine activity in a line segment as functions of time (*t*, s). Each equation addresses the amount of radioiodine in one of the three compartments shown in Figure 4.2-1. The three equations are

$$\frac{dq_a}{dt} = I + rq_s - \left(\lambda_v + \Delta + \lambda\right)q_a \qquad (4.2\text{-}1)$$

$$\frac{dq_s}{dt} = \Delta q_a - \left(r + \lambda + \phi\right)q_s \qquad (4.2\text{-}2)$$

$$\frac{dq_f}{dt} = \phi q_s - \lambda q_f \qquad (4.2\text{-}3)$$

where
q_a = activity (Bq or µCi) of the airborne radioiodine in the segment
I = rate (Bq s^{-1} or µCi s^{-1}) at which activity enters the line segment
q_s = activity (Bq or µCi) on line surfaces that is available for resuspension
r = resuspension rate constant (s^{-1})
λ_v = air removal rate constant (s^{-1}); $\lambda_v = Q\, V_s^{-1}$
Q = sampling line flow rate ($cm^3\ s^{-1}$)
V_s = volume of the line segment (cm^3)
Δ = deposition rate constant (s^{-1}); $\Delta = V_d\, A_s\, Vs^{-1}$
V_d = radioiodine deposition velocity (cm s^{-1})
A_s = surface area (cm^2) of the line segment
λ = radiological decay rate constant (s^{-1}) for the radioiodine isotope
ϕ = rate constant (s^{-1}) for the surface fixation process
q_f = activity (Bq of µCi) fixed on the surface.

The three equations are solved as functions of time for each of the line segments, usually 20, used to model the sampling line. The transient behavior following either an increase or decrease of radioiodine input to the line can be computed. Because the focus of this discussion is on long term average releases, the behavior at equilibrium is addressed here.

During a period of relatively constant input of elemental iodine into the sampling line, the surface activity in each segment will gradually increase to an equilibrium value. The equilibrium is reached when deposition of I_2 onto the surface of the segment is balanced by the removal due to resuspension from the surface and fixation to the surface. The air concentration in each segment also reaches an equilibrium value as this balance is achieved.

The equilibrium transmission factor for elemental iodine (TF_{ee}) is defined as the following ratio:

$$TF_{ee} = \frac{\lambda_v q_{ae20}}{I} \tag{4.2-4}$$

The numerator, which is the product of the equilibrium ^{131}I activity in the last (20th) line segment and the exhaust rate constant, is the rate (Bq s^{-1} or µCi s^{-1}) at which activity leaves the line under equilibrium conditions. The denominator is the rate (Bq s^{-1} or µCi s^{-1}) at which $^{131}I_2$ activity enters the sampling line. The transmission factor depends upon the properties of the sampling line as well as the behavior of elemental iodine.

In general, transmission factors for various iodine species depend upon the properties of the sampling line (length, diameter, flow rate) and the properties of the particular chemical form. While the dimensions of the sampling line are usually known and the equations for the model can be solved, knowledge of the parameters needed to use them is not exact. Uncertainty is associated with the estimates of the deposition, resuspension, and fixation rate constants. Monte Carlo techniques were used to estimate the transmission factors for elemental iodine in the sampling lines.

The Monte Carlo procedure uses information about the expected values and distributions of possible deposition, resuspension, and fixation parameter values to make a series of estimates of the transmission factor. The result of these calculations is a distribution of values of TF_{ee}. That distribution of estimates is typically lognormal, or approximately so, and is characterized by a median (50th percentile value) and a geometric standard deviation (GSD). It reflects the uncertainties in the input parameters and their effect on the estimates of TF_{ee}.

The model was used to compute sampling line losses for elemental iodine under various operating conditions that are discussed below. The whole range of estimates of sampling bias due to line losses was then used in the reevaluation of the radioiodine releases.

Extraction of a Representative Sample

Following the May–June 1961 releases, plant staff also investigated the overall sampling process. Tritium gas was used as a tracer to check whether the sample extraction system and sampling line were functioning properly. Known amounts of tritium gas (~190 Ci) were injected into the stack and measured with a portable Kanne chamber at the sample collection point. The measured tritium concentrations in two tests were an average of 96% of the expected tritium concentrations. These results provide confidence that a representative sample of stack gases was being collected and that the sampling system was functioning properly (Marter 1962).

It was noted that the linear velocity of air in the stack exhaust was about double the linear velocity in the sampling system. This difference in velocities means that sampling was not isokinetic (Marter 1962). Anisokinetic sampling is not an important factor for gaseous iodine, but it is considered in the evaluation of discharges of radioactive particles.

Estimates of Airborne Releases of Radioiodine at the SRS

Revised estimates of radioiodine releases to the atmosphere from the reprocessing facilities, production reactors, and research facilities are presented in the following sections. Original weekly and monthly reports have been used to prepare a history of reported releases. The data have been compared with summaries of releases that have been compiled in recent years (Cummins et al. 1991; Kantelo et al. 1993). Occasional small differences or inconsistencies were found when comparing the documents. In spite of them, the summaries are considered to be accurate reflections of the ^{131}I releases that were reported historically; however, the summaries do not address issues related to the reliability of the historic data.

Reprocessing facilities, which were the largest sources of airborne radioiodine releases and for which monitoring records are most complete, are discussed first and given the greatest attention. Overall, it is estimated that the F-Area and H-Area stacks discharged more than 300 times more radioiodine than all the reactor and research facility stacks combined. The most important discharges, from the point of view of human health, are those of elemental iodine. As was discussed earlier, this is because of its role in exposure via the air-grass-cow-milk pathway. It is estimated below that historic discharges of I_2 from the F-Area and H-Area stacks exceed those from other locations by at least a factor of 50. Those releases, which mostly occurred during early years of operation, are the most significant radioiodine releases at the SRS.

Estimates of Radioiodine Releases from Reprocessing Facilities

Reassessment of the radioiodine releases from the reprocessing facilities must consider the history of sampling techniques and the information on radioiodine species that were described above. The early monitoring and sampling systems were designed for collection of I_2. At that time, there was little or no knowledge of other less reactive chemical forms. After charcoal cartridges were employed (late 1961), more complete collection of radioiodine was accomplished and more attention was given to problems of incomplete sample collection. Subsequently, a new monitoring system was installed (Jolly et al. 1968). Table 4.2-2 shows the time periods considered in this reassessment.

Table 4.2-2 illustrates a gradual improvement of radioiodine monitoring at the reprocessing facilities. The most important changes were made following the 1961 releases, which led to a serious investigation of radioiodine monitoring practices. The only factor important to estimation of releases that was consistently not considered is loss of sample due to deposition in the sampling line. In the more recent summary of reported SRS radioiodine releases (Kantelo et al. 1993), line loss was not considered to be an important factor because most of the releases have been shown to consist of organic iodides. That view overlooks the fact that during the periods when the highest releases were reported only the reactive species were being measured.

The reported amounts of radioiodine released from the separations areas are discussed in the next section. That is followed by the reevaluation of those releases, which takes into account the factors identified in Table 4.2-2.

Table 4.2-2. Overview of Radioiodine Sampling and Monitoring Methods

Period	Method of sample collection	Comments on reported results
Dec 1954 to Oct 1956	NaOH scrubber	Only reactive forms were measured; system was not considered reliable; no corrections for line loss were made
Oct 1956 to Sept 1961	$AgNO_3$-impregnated filters	Only reactive forms were measured; unknown variability in collection efficiency; no corrections for line loss were made
Sept 1961 to Sept 1965	Charcoal cartridges	All chemical forms were measured; attention paid to collection efficiency variations; no corrections for line loss were made
After Sept 1965	Charcoal cartridges	New monitoring system with higher flow rate; all chemical forms were measured; attention paid to performance; no corrections for line loss were made

Reported Releases of ^{131}I. Values of the reported releases of ^{131}I from the F and H stacks during early years of operation (late 1954 to the end of 1961) are plotted in Figure 4.2-2 (Du Pont 1954a-l, 1955a-l, 1956a-l, 1957a-l, 1958a-l, 1959a-l, 1960a-l, 1961a-l). The year of largest releases was 1956, when several of the reported monthly releases exceed 100 Ci. The reported total for that year was slightly under 1600 Ci. Reported releases during 1957, 1959, and 1961 were ~290, ~160, and ~160 Ci, respectively.

Annual releases reported for years following 1961 did not exceed 40 Ci and those reported for 1972–1989 were all less than 3 Ci. Comparison of release estimates from monthly reports from 1954 through December 1971 (Du Pont 1954a-l, 1955a-l, 1956a-l, 1957a-l, 1958a-l, 1959a-l, 1960a-l, 1961a-l, 1962a-l, 1963a-l, 1964a-l, 1965a-l, 1966a-l, 1967a-l, 1968a-l, 1969a-l, 1970a-l, 1971a-l) with the summary of annual releases by Kantelo et al. (1993) and all the annual releases reported by those authors are contained in an Excel® spreadsheet. Clicking on the following hyperlink will provide automatic access to these data: Reported_I-131_Releases_(F,H).xls. Agreement between the sums of values in the monthly reports and the annual totals in the recent summary report is generally very good.

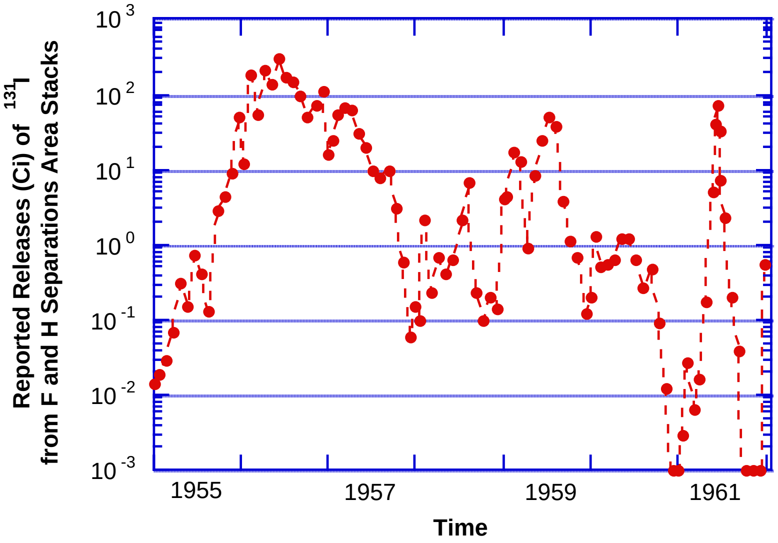

Figure 4.2-2. Reported releases of ^{131}I from the stacks of the F and H separations facilities during the earliest years of operation.

Revised ^{131}I Release Estimates. The revised estimates of ^{131}I releases from the reprocessing facilities reflect estimates of bias and uncertainty that were not addressed in previous reports of those releases. Uncertainties that are addressed include those associated with sample collection and with radioactivity measurements. The biases in the reported results stem from the fact that some sampling media did not collect organic forms of radioiodine and because there were losses of I_2 due to deposition in the sampling line.

As indicated in Table 4.2-2, several time periods must be considered because the methods of radioiodine monitoring changed. Those same time periods are listed in Table 4.2-3, which contains information related to several questions about radioiodine monitoring for elemental iodine and organic iodides. The less reactive inorganic fraction that may have been present is not addressed explicitly because of lack of information; however, identification of that fraction is much less important than the distinction between I_2 and organic iodides. In the two earliest periods, the organic iodide releases were not monitored. They are estimated from the releases of elemental iodine using observations that organic forms comprised 70–90% of the total release from U.S. reprocessing facilities. The calculational procedure that was used is described later.

Estimates of the efficiencies of the collection media (NaOH scrubber solutions and $AgNO_3$-impregnated filters) for I_2 are based primarily upon reported SRS experience. However, the selected ranges are somewhat broader than that to reflect a greater variety of field sampling conditions. For the scrubber efficiency, a range of about ±10% is considered. A broader range is considered for the $AgNO_3$-impregnated filter papers. This range is based upon the results of field tests that were conducted in 1961. A collection efficiency as low as 34% was measured for a sampling flow rate that was lower than that normally used for the filter paper. It is not certain why the field tests did not include sampling at the normal flow rate, but it may have been related to the requirements for the caustic scrubber used for comparison. The lower bound of 0.2 is believed to cover the range of possible efficiencies. Relatively high efficiencies had been measured in field tests in 1956. The factors that changed during the roughly 5-year period and led to lower collection efficiency for I_2 and when the change occurred are not known.

Table 4.2-3. Factors Considered in Preparing Revised Estimates of ^{131}I Releases

Period	Chemical Form of ^{131}I released	Release Measured?	Sample collection efficiency	Estimated measurement uncertainty	Estimated transmission factor
Dec-54 to Oct-56	Elemental	Yes	0.75–0.95	40%	0.28[c]
	Organic	No[a]	NA[b]	NA	NA
Oct-56 to Sept-61	Elemental	Yes	0.2–0.9[e]	30%	0.42[f]
	Organic	No[d]	NA	NA	NA
Sept-61 to Sept-65	Elemental	Yes	[g]	20%	0.28[h]
	Organic	Yes		20%	1
After Sept-65	Elemental	Yes	[g]	15%	0.59[i]
	Organic	Yes		15%	1

[a] Organic iodides were not collected by NaOH scrubber; releases estimated using observations that organic iodides constituted 70–90% of the total radioiodine discharge.

[b] Not applicable.

[c] Median estimate based upon the design flow rate of 944 $cm^3\ s^{-1}$ (2 $ft^3\ min^{-1}$) of the NaOH scrubber system; estimated GSD = 1.94 (see Figure 4.2-3).

[d] Organic iodides were not collected by AgNO3-impregnated filters; releases estimated using observations that organic iodides constituted 70–90% of the total radioiodine discharge.

[e] Estimates based upon reported results of 1956 field tests and measurements following the 1961 releases.

[f] Median estimate based upon the reported total flow rate of 1316 $cm^3\ s^{-1}$ (3 $ft^3\ min^{-1}$); 944 $cm^3\ s^{-1}$ (2 $ft^3\ min^{-1}$) to the $AgNO_3$-impregnated filter and 472 $cm^3\ s^{-1}$ (1 $ft^3\ min^{-1}$) to the NaOH scrubber system; estimated GSD = 1.65 (see Figure 4.2-3).

[g] Although variable, collection efficiencies were checked routinely and corrections made; efficiencies of the charcoal cartridges are estimated to be within 10% of the value estimated.

[h] Estimate based upon a flow rate of 944 $cm^3\ s^{-1}$ (2 $ft^3\ min^{-1}$) through the charcoal cartridge.

[i] Median estimate based upon the design flow rate of 2360 $cm^3\ s^{-1}$ (5 $ft^3\ min^{-1}$) of the stack monitoring system; estimated GSD = 1.34 (see Figure 4.2-3).

Equilibrium transmission factors for organic iodides and elemental iodine through the long sampling lines have been computed using the line loss model discussed earlier. Transmission factors for organic iodides through sampling lines, even for short times after an increase in concentration, are practically identical to 1 and no correction has been made here for line losses of organic iodides.

The transmission factor for elemental iodine depends upon the sampling flow rate, which varied with time. Values of the deposition, resuspension, and fixation parameters of the model are based upon experimental results for lines that are similar and the distributions reflect a range of results. The distribution of deposition velocities was taken to be lognormal with a median value of 0.02 cm s^{-1} and a GSD of 1.4. The distributions of resuspension and fixation rate constants were logtriangular, with the mode equal to the upper bound. For resuspension, the range of values was from 1.1×10^{-7} to $6.8 \times 10^{-6}\ s^{-1}$. For the fixation rate constant, the range considered was 9.2×10^{-8} to $1.0 \times 10^{-5}\ s^{-1}$. The calculations of the elemental iodine transmission factors reflect the observed variability of these parameters as well as a potential 15% variability in the nominal flow rates.

The distributions of estimates of the equilibrium transmission factors for elemental iodine are shown in Figure 4.2-3. The distributions are contained in an Excel® spreadsheet. Clicking on the following hyperlink will provide automatic access to the information: Transmission Factor Estimates.xls. Sampling line losses decrease as the sampling flow rate increases. The median TF_{ee} was estimated to be 0.28 for a flow rate of 944 $cm^3\ s^{-1}$ (1 $ft^3\ min^{-1}$), but for a flow rate of 2360 $cm^3\ s^{-1}$ (5 $ft^3\ min^{-1}$) it was 0.59. Dispersion of the estimates decreased with increasing flow rate. Approximate GSDs are 1.94, 1.65, and 1.34 for flow rates of 944, 1416, and 2360 $cm^3\ s^{-1}$ (1, 2, and 5 $ft^3\ min^{-1}$), respectively. The median transmission factors for the two lower flow rates, 0.28 and 0.42, are approximately the same as the estimate made above by extrapolation of the 1961 SRS result to the entire line.

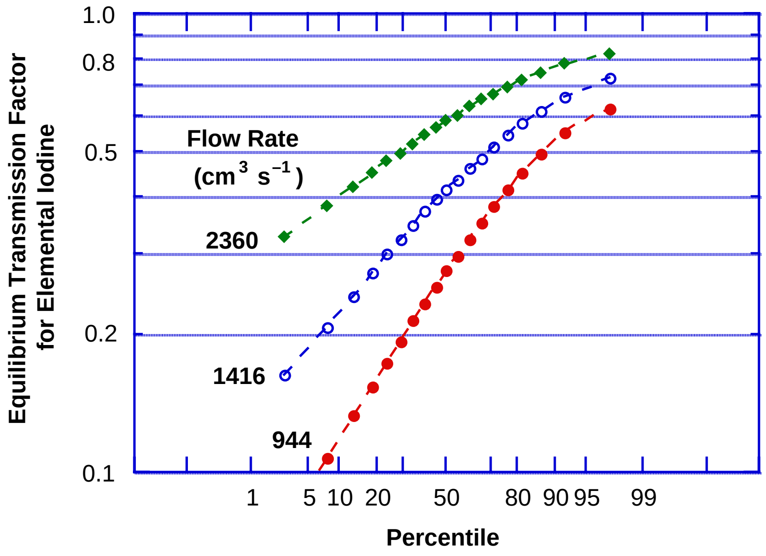

Figure 4.2-3. Equilibrium transmission factor estimates for elemental iodines for three sampling flow rates through a 93-m sampling line.

The method used here to reevaluate releases of ^{131}I at various times depends upon the measurement technique that was used. During the two earliest time periods defined in Table 4.2-3, the measurement procedure only gave an estimate of the elemental iodine release. After the fall of 1961, when routine use of charcoal cartridges was begun, that technique yielded an estimate of the total radioiodine release. The reanalysis of these results relies upon our knowledge of the fraction of the total radioiodine in organic form (f_o), which we estimate to be 70–90%. Designating the total amount of radioiodine released by Q_t and the two fractions by Q_e and Q_o, respectively, we may write:

$$Q_t = Q_o + Q_e$$
$$Q_o = f_o Q_t \qquad (4.2\text{-}5, \text{-}6)$$

Substitution and rearrangement yields:

$$\frac{Q_o}{f_o} = Q_o + Q_e \tag{4.2-7}$$

$$Q_o = \frac{f_o Q_e}{(1 - f_o)} \tag{4.2-8}$$

It should be noted that the quantity Q_e in these and subsequent equations refers to the corrected estimate of elemental radioiodine release. That is, it reflects corrections of the measured amount of elemental iodine (Q_{me}) for incomplete sample collection and for sample transmission losses discussed above. If the sample collection efficiency for elemental iodine is ε_e, and the equilibrium transmission factor for that species is TF_{ee}, then:

$$Q_e = \frac{Q_{me}}{\varepsilon_e TF_{ee}} \tag{4.2-9}$$

Corrections for sample collection efficiency were made historically. However, we consider a range of possible corrections as opposed to point estimates made previously. Corrections for sample transmission losses, not made previously, were discussed above. When the measurement technique could only detect elemental iodine, we first estimate the release of elemental iodine and then estimate the release of organic iodides.

A different approach is necessary to reevaluate radioiodine releases after September 1961, when the measurement technique collected both elemental iodine and organic iodides. The sample collection efficiency (called ε_o) was primarily determined by collection of organic iodides, and probably overcorrected somewhat for the elemental iodine fraction. The initial estimate of the total radioiodine release given in previous reports is:

$$Q_t = \frac{Q_{mt}}{\varepsilon_o} \tag{4.2-10}$$

However, this estimate neglects sample transmission losses. The relationship between Q_t and the amounts of organic and elemental radioiodine released is:

$$Q_t = Q_o + Q_e TF_{ee} \tag{4.2-11}$$

Rearrangement and use of the previous relationship (equation 4.2-8) between Q_e. and Q_o yields:

$$Q_o = Q_t - \frac{Q_o(1 - f_o)TF_{ee}}{f_o} \tag{4.2-12}$$

Solving for the organic iodide release, we have:

$$Q_o = \frac{f_o Q_t}{\left[f_o + (1 - f_o)TF_{ee}\right]} \tag{4.2-13}$$

and then the elemental iodine release is calculated using:

$$Q_e = \frac{Q_o(1 - f_o)}{f_o} \tag{4.2-14}$$

The two procedures just discussed were applied, in appropriate time periods, to previous estimates of radioiodine releases from the separations stacks in the F-Area and H-Area. The median estimates of the revised ^{131}I releases of elemental iodine are shown in Figure 4.2-4. The estimates are highest for 1955–1961 and are much lower for all subsequent years. It should be noted that the vertical scale is logarithmic and that each of the major divisions is ten times greater than the previous one. Thus, the median estimate for 1962 is more than 1000 times lower than the median estimate for 1956.

Figure 4.2-5 contains the release estimates for ^{131}I in the form of organic iodides from the same stacks. The pattern is similar to that in the previous figure and the vertical scale is also logarithmic. Because organic iodides are believed to comprise a large fraction of the total release, estimated organic iodide releases exceed those for elemental iodine. However, as noted earlier, releases of organic iodides are much less important for exposure of the thyroid via the air-grass-cow-milk pathway.

The main (5th, 25th, 50th, 75th, and 95th) percentiles of the distributions of release estimates for both elemental iodine and organic iodides are tabulated in an Excel® spreadsheet. In that listing, the median (50th percentile) value is highlighted to guide the reader to the central estimate of the amount released. The 5th, 50th, and 95th percentile values from this table are the basis for the release estimates plotted in Figures 4.2-4 and 4.2-5. The Excel® spreadsheet can be directly accessed by clicking on the following hyperlink: Revised_I-131_Releases_(F,H).xls.

The revised estimates of ^{131}I releases have been criticized by SRS staff (Heffner 1999) because they found good agreement between environmental concentrations predicted using the originally reported release estimates for 1956 and contemporary environmental measurements. They also cited similar comparisons made in an earlier plant report (Reinig 1959). As was discussed above, in the 1950s, both the effluent and environmental concentrations of ^{131}I were estimated using sampling media that were not highly reliable. Uncertainties in meteorological modeling, even for flat terrain, are on the order of a factor of 3–4 without consideration of plume depletion. In view of the quality of both sets of measurements and the uncertainties involved, the comparisons between predicted and measured concentrations in 1956 are not conclusive. When Reinig made similar comparisons in 1959, he had no knowledge of the unreliability of the sampling systems.

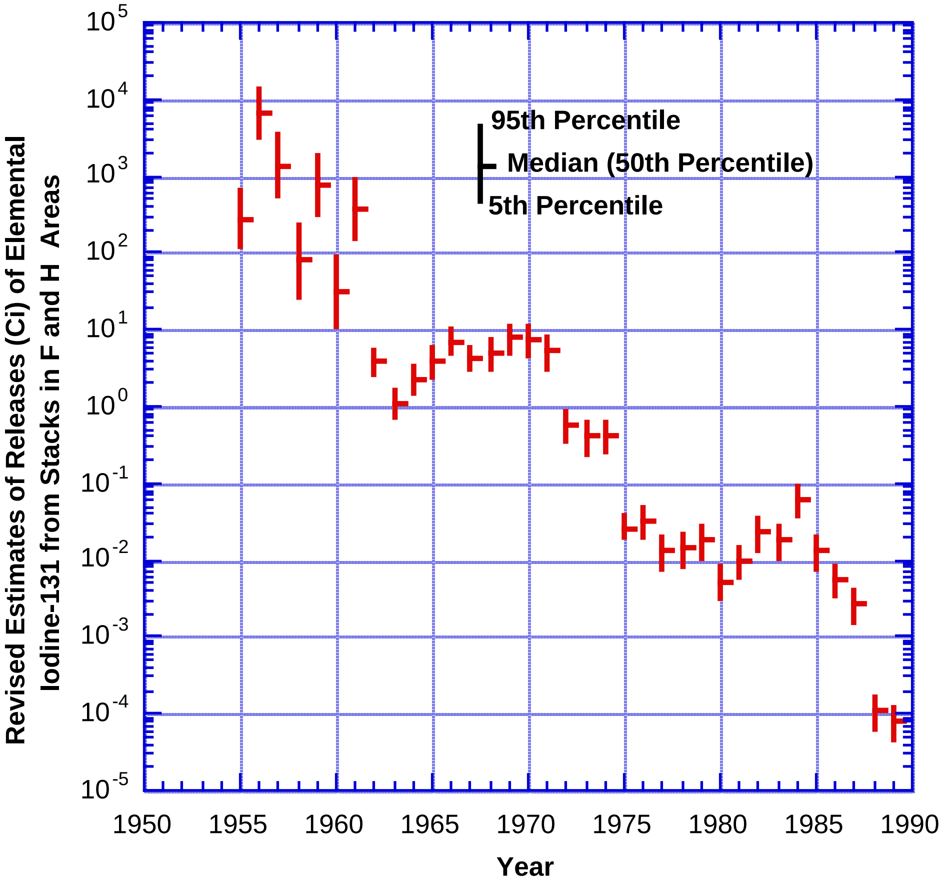

Figure 4.2-4. Revised estimates of releases of ^{131}I in elemental form (I_2) from stacks in the F and H separations areas. For each year, the length of the vertical line shows the range (5th to 95th percentiles) of the distribution of release estimates and the median estimate is indicated by the horizontal bar.

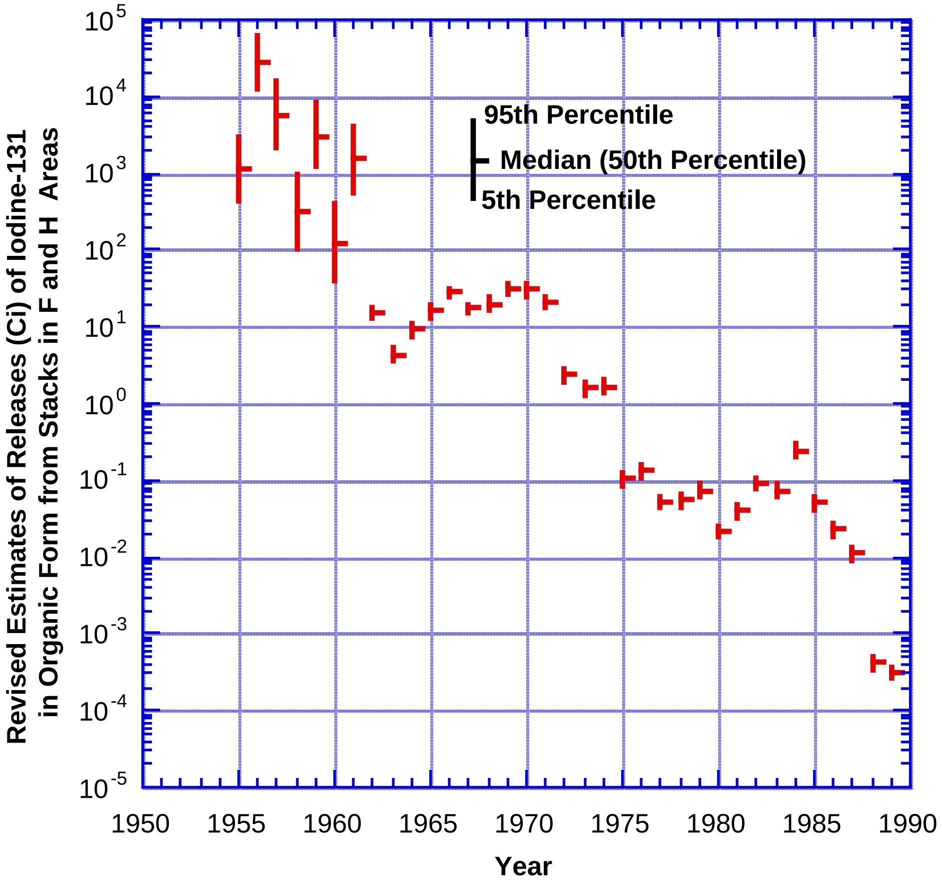

Figure 4.2-5. Revised estimates of releases of ^{131}I as organic iodides (e.g., CH_3I) from stacks in the F and H separations areas. For each year, the length of the vertical line shows the range (5th to 95th percentiles) of the distribution of release estimates and the median estimate is indicated by the horizontal bar.

Releases of ^{129}I. Iodine-129 is a fission product produced in reactor fuel that has a very long (~16 million-year) half-life. Most releases of ^{129}I occur when the fuel is being processed because the fuel cladding is dissolved and then the fuel itself is dissolved. These processes first liberate fission gases that have diffused into the gap between the fuel and the cladding and subsequently provide an opportunity for all volatile elements to be released from the solutions in the dissolver or in other vessels to which solutions are transferred.

The amount of ^{129}I produced in a particular batch of fuel depends upon the time that the fuel is irradiated in the reactor core. Figure 4.2-6, which shows the total power production of the SRS reactors during the years 1954–1989, is a guide to the amount of ^{129}I that was produced at the SRS. Note that the highest production occurred during 1959–1962, when the total power levels were at or above 3000 GW-d per year. During the years 1958, 1963, 1964, 1966, and 1967 the total power production was above 2500 GW-d. In years prior to 1958 and after 1968 reactor power levels were less than 2000 GW-d.

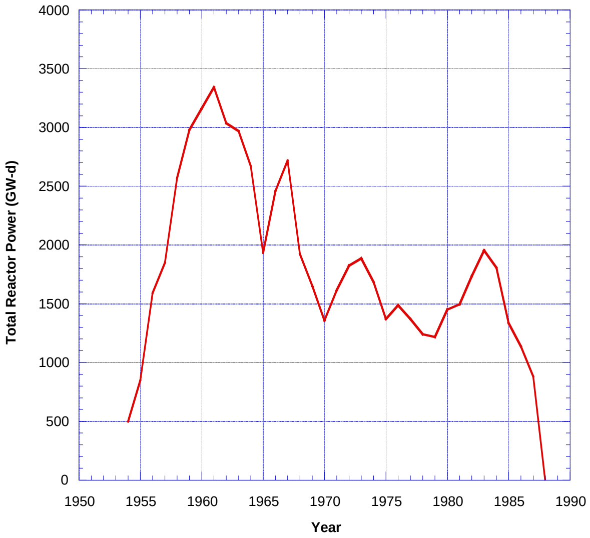

Figure 4.2-6. Total reactor power levels in operating SRS reactors. Data are given in Chapter 2.

The annual production of plutonium at the SRS (DOE 1996) is shown in Figure 4.2-7. That figure provides a guide to the time of releases of ^{129}I as the result of fuel processing. The delays (a) between removal of fuel from the reactor and start of fuel processing and (b) between the start of processing and the shipment of the product are reflected in the plutonium production plot. The peak releases of ^{129}I likely fell between the peak in power production and the peak in plutonium production. The plutonium production data from DOE (1996) are contained in an Excel® spreadsheet, which can be accessed by clicking on the following hyperlink: SRS_Plutonium_Production.xls.

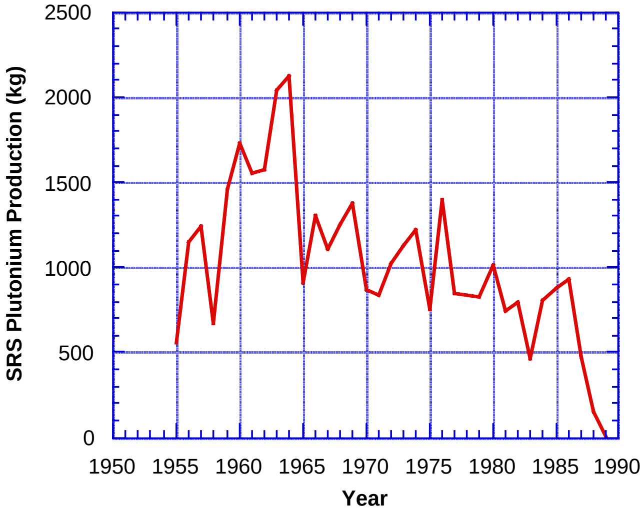

Figure 4.2-7. Amounts of plutonium produced at the SRS (DOE 1996).

Two different ^{129}I release histories have been reported for the SRS. Both are primarily based upon estimates of fuel inventories, release fractions, and cleanup system performance. Cummins et al. (1991) and Kantelo et al. (1993) present an average release of 0.21 Ci y^{-1} of ^{129}I between 1955 and 1973. Although Kantelo et al. (1993) identify a set of estimates based more upon plant operations (Boone et al. 1985), those estimates were not used for dose calculations contained in their report. The estimates made by Boone et al. are more consistent with the data on SRS reactor power levels and processing facility operations illustrated above. Measurements of ^{129}I releases did not begin until 1981. Measurements made in 1984 and later years are reported by Kantelo et al. (1993).

Estimated releases of ^{129}I that reflect historic operations are shown in Figure 4.2-8. The estimated releases are tabulated in an Excel® spreadsheet, which can be accessed by clicking on the following hyperlink: I-129_Releases_(F,H).xls. For the years 1955–1960, we estimate the uncertainty in the plotted ^{129}I releases to be ± 40% because of variations in scrubber performance during the earliest years of operations. We estimate that uncertainties in release estimates are ± 25% for years between 1961 and 1980 and that those for estimates in years after 1980 are ±20%.

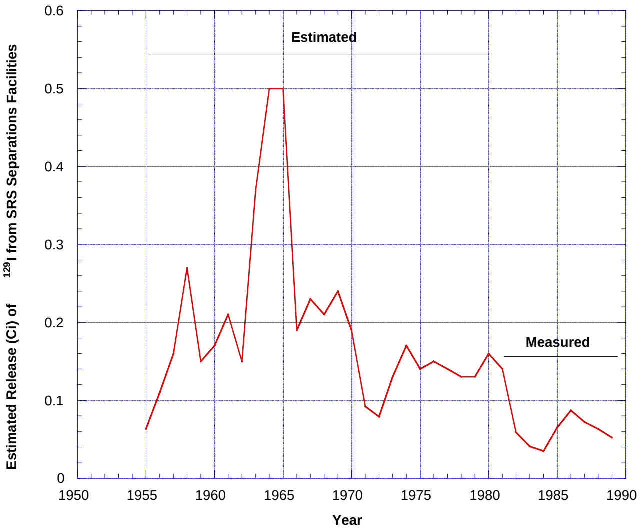

Figure 4.2-8. Estimated releases of ^{129}I from separations facilities in F-Area and H-Area.

Releases from SRS Reactors

This section focuses upon the release of ^{131}I to the atmosphere from the SRS production reactors. Releases of ^{129}I from reactors are expected to be small because of the low level of production of ^{129}I compared with that for ^{131}I. There are no quantitative estimates of the releases of ^{131}I to the atmosphere from SRS reactors for years prior to 1972. The summary document for radioiodine (Kantelo et al. 1993) contains only the releases that were reported after that time. There are some historic reports of releases of nonvolatile beta activity from the reactors. There are no atmospheric release estimates for R reactor, which was shut down in 1964, in Kantelo et al. (1993). The only release estimates given for L reactor are those based upon measurements during the latter years of operation (1986-1988). For the other reactors (P, K, and C), the report contains results of measurements of airborne releases that were performed after 1972.

Although no releases of ^{131}I to the atmosphere are reported for the early years of SRS reactor operations, such releases surely occurred. Considering the nature of the iodine sampling systems at that time, failure to observe airborne ^{131}I releases is not an adequate basis for suggesting that they did not occur. Silver nitrate-impregnated filters were used for collection of ^{131}I in samples of airborne effluent until the early 1960s. As noted above, this technique was inadequate. At about the same time that charcoal cartridges were introduced for sample collection, particle filtration and iodine absorption systems were installed in the SRS reactors to protect against large releases in case of accidents. During power operation, the building exhaust air was discharged through the filtration systems. During outages, part of the building air bypassed the filters. Tests of the activated charcoal from these absorbers were reported by Durant (1967) to show that the medium was quite efficient (> 99.9%) for removal of elemental iodine. The charcoal was not tested for collection of organic iodides but it is believed that the efficiency was not as high for that species.

Estimates of early atmospheric releases of ^{131}I from the SRS reactors are needed to assess the importance of those releases relative to those from the SRS reprocessing facilities. A number of factors affect ^{131}I releases from reactors. The reactor power level determines the maximum inventory of ^{131}I in the fuel. After a month of power operation, the ^{131}I content of the fuel is near its maximum value. Thus, duration of operation is not a major determinant of the release potential. After the inventory of ^{131}I is near the equilibrium level, the potential for release depends primarily upon the integrity of the fuel cladding. If the cladding is intact, radioiodine gas that has diffused into the gap between the fuel and the cladding will be contained. If not, radioiodines can leak from the fuel elements into the reactor moderator or fuel storage pool water and will be available for release in liquid or gaseous effluents.

The following rationale forms the basis for a method to estimate releases of ^{131}I to the atmosphere from the reactors. Because releases of ^{131}I to the atmosphere are a consequence of releases of ^{131}I from fuel elements to moderator or pool water, discharges of ^{131}I in liquid waste indicate that releases from the fuel have occurred and indicate that releases to the atmosphere were likely. The summary of Cummins et al. (1991) contains 26 pairs of estimates of annual releases of ^{131}I to the air and water from three reactors after 1971. The data were used to compute the ratio of airborne release (Ci) to the liquid release (Ci) for a particular year. The reactors and years with data available are: P Reactor (1973), K Reactor (14 years between 1972 and 1987), and C Reactor (11 years between 1973 and 1985). Because the releases were filtered, the radioiodine collected was most likely present as organic iodides. No correction for line losses was made when computing the ratios. Figure 4.2-9 shows the distribution of ratios of releases to the atmosphere to those in liquid wastes. The figure reflects 25 of the 26 ratios; one value of 45 (K Reactor in 1981) was excluded because it was clearly different from the other results and was considered an outlier.

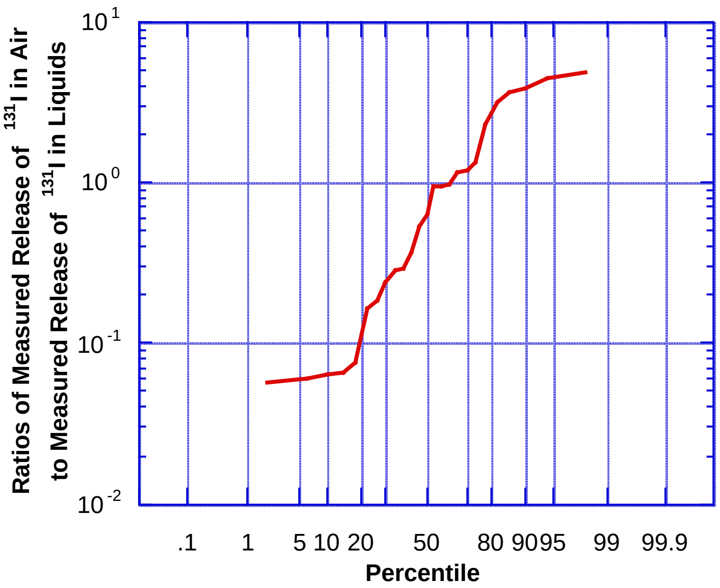

Figure 4.2-9. Distribution of ratios of the annual ^{131}I release (Ci) to the atmosphere to the annual ^{131}I release (Ci) in liquid wastes in years between 1972 and 1987. Gaseous effluents were filtered during power operation.

The distribution of release ratios has three components. About 20% of the ratios are relatively low (0.05–0.08) and a comparable fraction are relatively high (3–5). The remaining 60% of the ratios are roughly lognormally distributed between 0.2 and 2.3, with a median of about 0.6 and a GSD of about 2.3. The release data and computed ratios are contained in an Excel® spreadsheet, which can be accessed by clicking on the following hyperlink: Ratios_of_Releases_(A-LW).xls. These ratios can be used together with previously reported and estimated releases of ^{131}I in liquid wastes to estimate the releases of ^{131}I to the atmosphere during years when no definitive measurements were made.

Figure 4.2-10 shows the reported and estimated releases of ^{131}I in liquid wastes from all the SRS reactors for the years between 1953 and 1975. For the years 1958 and 1959, the release was estimated to be 50 Ci, which is the average of the releases reported for five nearby years (1957, 1960–1964). Releases for 1953–1956 were also estimated. Releases were interpolated between a low release (0.1 Ci) estimated for 1953 to 65 Ci reported for 1957. The release data are contained in an Excel® spreadsheet, which can be accessed by clicking on the following hyperlink: Reactor_Liquid_Waste_Releases.xls.

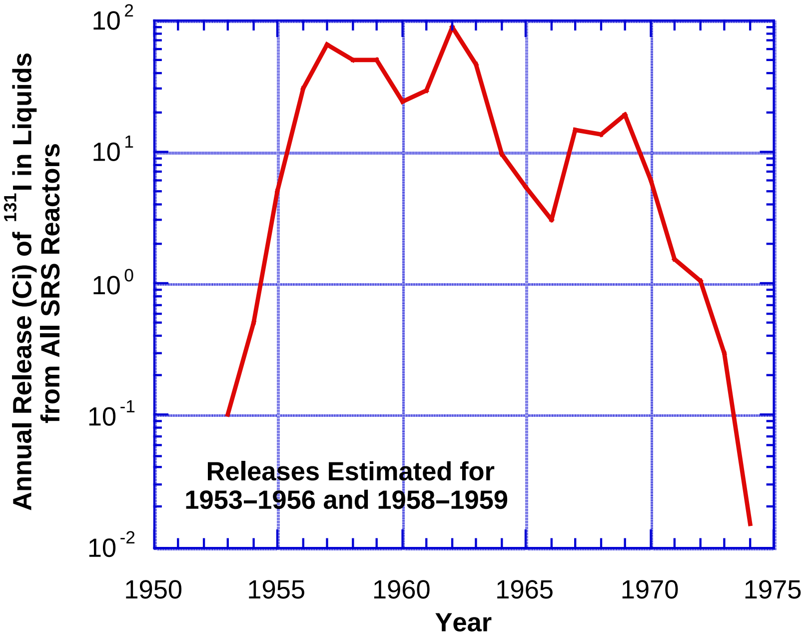

Figure 4.2-10. Reported and estimated reactor liquid waste releases of ^{131}I for all SRS reactors combined. See text for information about the estimates made for 1953–1956 and 1958–1959.

The measured releases of airborne ^{131}I during the years for which ratios were computed were lower than the amounts that would have been released had the charcoal filters not been present. Air purged from lower levels of the reactor buildings was not filtered. Containment purges during shutdown were found to contribute about 55% of the total release from commercial PWRs (Pelletier et al. 1978b) but that result offers only a rough guide to the appropriate fraction for the SRS reactors. A range of 30–70% is assumed for the fraction of the ^{131}I release that was unfiltered. The charcoal filter units were assumed to remove 0.999–0.9995 of the inorganic iodine and 0.8–0.95 of the organic iodides in air discharged through them. It is estimated, based on measurements reported by Pelletier et al. (1978b), that about 40% of the ^{131}I in routine PWR discharges was present as organic iodides. Using these assumptions, it is estimated that the sampling program estimated 31–74% of the release of airborne ^{131}I that would have occurred had the filters not been in place. All of the reactors had filter systems at some time in 1964 (Durant 1967). For the earlier years (1953–1963) the release ratios shown in Figure 4.2-9 must be adjusted for the effects of the charcoal absorbers on the amounts released.

The liquid waste release amounts shown in Figure 4.2-10 and the distribution of release ratios shown in Figure 4.2-9 were used in Monte Carlo calculations of estimated releases of ^{131}I from the SRS reactors. The liquid waste releases were represented by triangular distributions with a mode equal to the reported release. Minimum and maximum values for these distributions were ±25% of the mode for times before 1971 and ±20% of the mode for later years. For years prior to

1964, the effect of filtration on the release ratios was included. A uniform distribution over the range of values discussed above (0.31–0.74) was used for the calculations.

Results of the calculations are shown in Figure 4.2-11 for all years of operation. The plot provides three percentiles (5th, 50th or median, and 95th) of the distributions that were estimated. The release estimates shown are those for all forms of ^{131}I. Based upon measurements at commercial PWRs (Pelletier et al. 1978b), it is estimated that about 30% of the total would be elemental iodine and that the remainder would be divided between less reactive inorganic forms (~40%) and organic iodides (~30%).

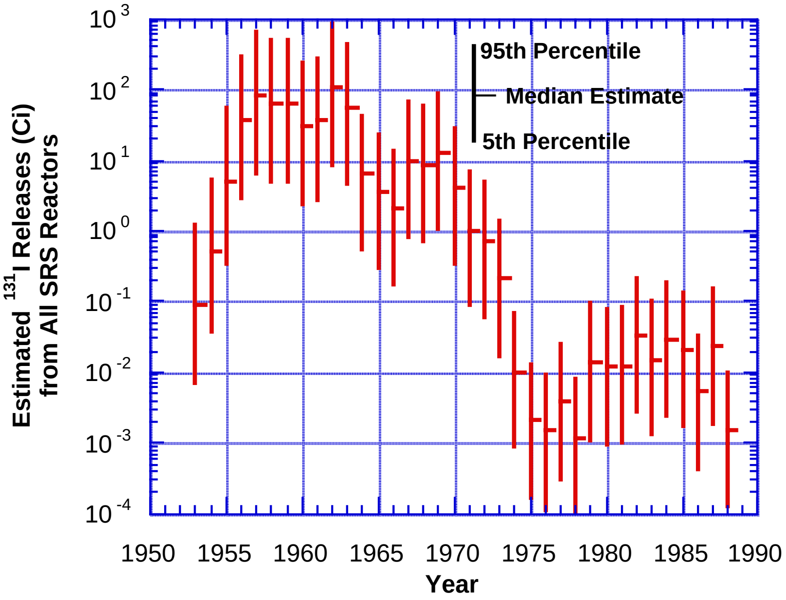

Figure 4.2-11. Estimated releases of ^{131}I from all SRS reactors to the atmosphere. For each year, the length of the vertical line shows the range (5th to 95th percentiles) of the distribution of release estimates and the median estimate is indicated by the horizontal bar.

The main (5th, 25th, 50th, 75th, and 95th) percentiles of the distributions of release estimates for all SRS reactors are tabulated in an Excel® spreadsheet. In that listing, the median (50th percentile) value is highlighted to guide the reader to the central estimate of the amount released. The 5th, 50th, and 95th percentile values from this table are the basis for the release estimates plotted in Figures 4.2-11 and 4.2-12. The spreadsheet can be directly accessed by clicking on the following hyperlink: Est_I-131_Releases_(Reactors).xls.

Figure 4.2-12 shows results of the calculations for only those years when releases were reported. It provides a comparison between the estimates resulting from the estimation procedure and those that were reported. Correspondence between the estimated and reported values is not surprising because the release ratios used in the calculations were taken from this period. However, the comparison does indicate that the estimates are in the expected range.

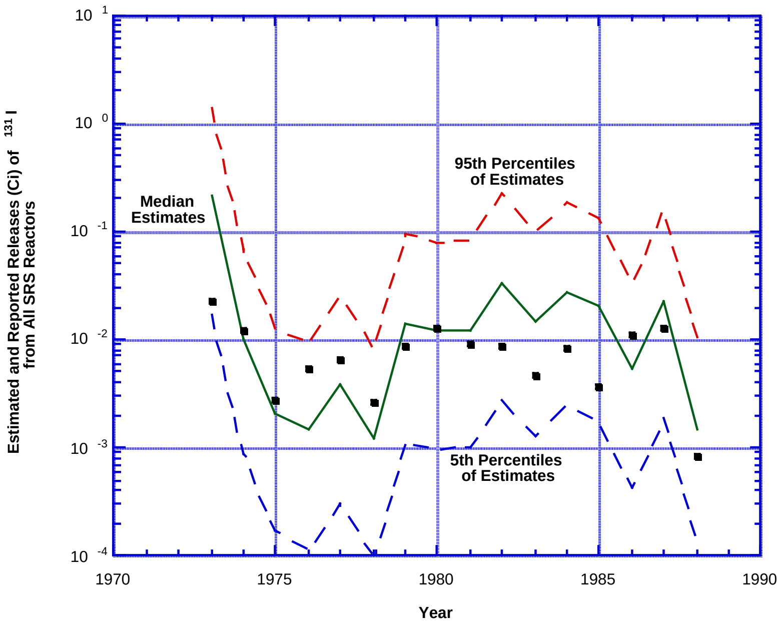

Figure 4.2-12. Comparison of estimated radioiodine releases from all SRS reactors to the atmosphere with measured values (Kantelo et al. 1993). The median and range (5th to 95th percentiles) of the distribution of release estimates are shown by solid and dashed lines, respectively. The reported annual releases are indicated by the solid squares.

Releases from Other SRS Facilities

The only other facility that released measurable amounts of ^{131}I at the SRS was the Savannah River Laboratory (SRL) research facility. Laboratory research in support of processing activities was conducted by the SRL. Some of the research employed short-cooled irradiated fuel that contained ^{131}I and some of the radioiodine was released. Kantelo et al. (1993) indicate that the total measured release was 6.4 Ci of ^{131}I, about half of which was reported to have occurred in 1958 (Cummins et al. (1991). The next largest release, about 1.6 Ci, was reported to have occurred in 1964 (Cummins et al. 1991).

Based upon the previous discussion of radioiodine monitoring at the SRS, it is unlikely that releases occurring prior to 1962 were measured reliably. Recorded releases between 1958 and 1961 total about 3.4 Ci of ^{131}I; no estimates are given for prior years (Cummins et al. (1991). It is believed, based on other experience (Sill and Flygare 1960), that most of the radioiodine released from the shielded cells was probably in elemental form. It seems unlikely, considering previous discussions of corrections of estimated releases, that more than 20 Ci of ^{131}I were actually released from the SRL stacks. The SRL releases are small compared to those from reactors and much smaller than those from reprocessing plants.

RELEASES OF BETA-GAMMA-EMITTING PARTICLES

The term “beta-gamma-emitting particles” refers to a large group of radioactive fission and activation products of many non-volatile elements. These radionuclides are produced in reactors when fuel and reactor components are irradiated. When released, they are generally associated with solid particles and are thus distinguished from the radioactive noble gases and the radioiodines, which are always or much more likely to be in gaseous form. Some radionuclides in this category are very short-lived and decay prior to fuel processing, which (for reasons discussed earlier) provides the principal opportunity for release to the atmosphere. Those that are released generally have half-lives on the order of tens of days (like ^{89}Sr, ^{95}Zr, ^{95}Nb, ^{103}Ru and ^{141}Ce), hundreds of days (like ^{106}Ru and ^{144}Ce), or many years (like ^{90}Sr and ^{137}Cs).

This section also focuses on the F and H fuel processing facilities because fuel dissolution and processing activities occurred there. Measurement records, summarized by Cummins et al. (1991), show that the F and H fuel processing plants were the largest sources of release of beta-gamma-emitting particles at the SRS. Some releases were measured from the reactor facilities and from the SRL stacks, but the quantities were much smaller.

Original weekly and monthly reports have been reviewed and the data on atmospheric releases contained in them has been compiled to prepare a history of reported releases of beta-gamma-emitting particles from the SRS fuel processing facilities. Initial measurements did not quantify releases of specific isotopes, but were estimates of the amounts of “non-volatile beta” activity released from the stacks (Du Pont 1954a-l, 1955a-l, 1956a-l, 1957a-l, 1958a-l, 1959a-l, 1960a). Early in 1960, results of more specific analyses were first reported. During most of the 1960s, release estimates were given for the sums of: two ruthenium isotopes (^{103}Ru plus ^{106}Ru); two strontium isotopes (^{89}Sr plus ^{90}Sr); two cerium isotopes (^{141}Ce plus ^{144}Ce); the zirconium-niobium pair (^{95}Zr plus ^{95}Nb); and for the long-lived cesium isotope, ^{137}Cs (Du Pont 1960b-l, 1961a-l, 1962a-l, 1963a-l, 1964a-l, 1965a-l, 1966a-l, 1967a-l, 1968a-l). Beginning in 1969, routine monthly reports contained separate release estimates for the two ruthenium isotopes and that practice was continued in later years (Du Pont 1969a-l, 1970a-l, 1971a-l). At the start of 1971, the listing was further segregated to provide estimates for ^{95}Zr and ^{95}Nb as well as for ^{141}Ce and ^{144}Ce (Du Pont 1971a-l).

Some additional data are provided in the SRS summary release report (Cummins et al. 1991). Included there are estimates of releases of the two strontium isotopes (^{89}Sr plus ^{90}Sr), the two ruthenium isotopes (^{103}Ru plus ^{106}Ru), and ^{137}Cs from the F and H processing plants during the early years of operation. The following illustrations are based upon the report of Cummins et al. (1991). Figures 4.2-13 and 4.2-14 show, for F-Area and H-Area respectively, the sums of the reported releases of the beta-gamma-emitting particles discussed here. Those results generally agree with the sums of releases of beta-gamma-emitting particles that were derived from the monthly reports. Two significant differences can been seen in the spreadsheet containing the reported releases. For F-Area in 1959, the sum of releases reported by Cummins et al. (1991) is about double that derived from the monthly reports (~8 Ci versus ~4 Ci). For H-Area in 1955, the sum of releases reported by Cummins et al. (1991) is about 19 Ci. That total is about 5 Ci smaller that the total of about 24 Ci derived from the monthly reports.

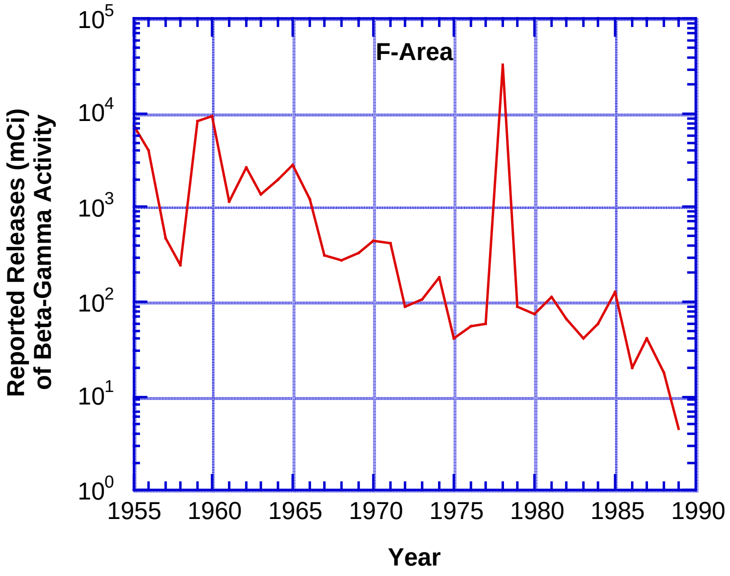

Figure 4.2-13. Reported releases of beta-gamma-emitting particles to the atmosphere from F-Area (Cummins et al. 1991). Estimates for ^{89,90}Sr, ^{95}Zr-Nb, ^{103,106}Ru, ^{137}Cs, and ^{141,144}Ce have been summed.

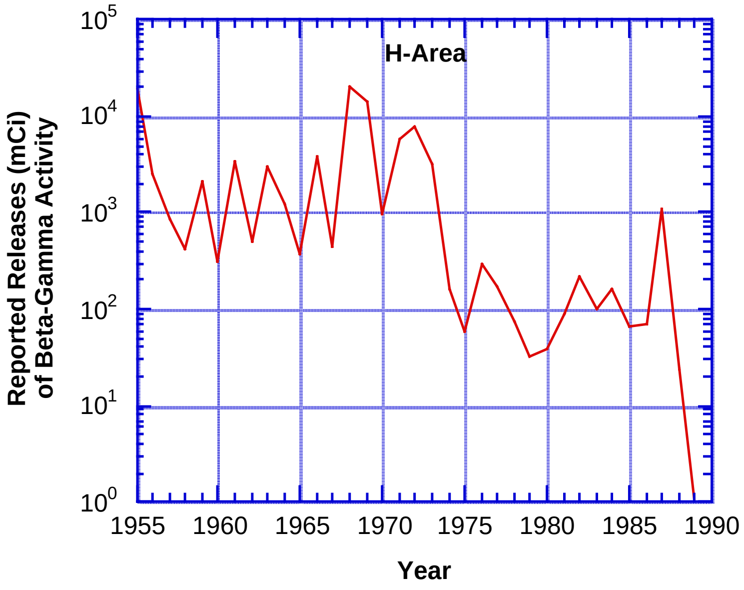

Figure 4.2-14. Reported releases of beta-gamma-emitting particles to the atmosphere from H-Area (Cummins et al. 1991). Estimates for ^{89,90}Sr, ^{95}Zr-Nb, ^{103,106}Ru, ^{137}Cs, and ^{141,144}Ce have been summed.

Figure 4.2-15 shows the releases of ruthenium and cerium isotopes from F-Area. There are no estimates for the latter nuclides prior to 1960. Comparison of this figure with Figure 4.2-13 shows that releases of ^{103}Ru and ^{106}Ru comprise the largest fraction of the total releases of beta-gamma-emitting particles from F-Area. Ruthenium isotopes are identified in some monthly reports as being major components of stack releases; for example, see Du Pont (1956i, 1958j). The large release in 1978 was also of ruthenium (Cummins et al. 1991). Figure 4.2-16 shows the releases of ^{89,90}Sr, ^{95}Zr-Nb, and ^{137}Cs from F-Area. Releases of zirconium and niobium, which are only reported for years after 1959, exceed those of the longer lived isotopes in later years. In some years the releases of ^{95}Zr and ^{95}Nb exceed those of ruthenium isotopes.

Figure 4.2-17 shows the releases of ^{103,106}Ru and ^{141,144}Ce from H-Area and Figure 4.2-18 shows the releases of the isotopes of strontium, zirconium, niobium, and cesium from H-Area. Again, the ruthenium isotopes account for large fractions of the releases for most years, although a release of about 1 Ci of ^{137}Cs is prominent in 1987. Separate reviews of the releases of cesium and strontium, based upon the summary report, have been prepared (Carlton et al. 1992a, 1992b).

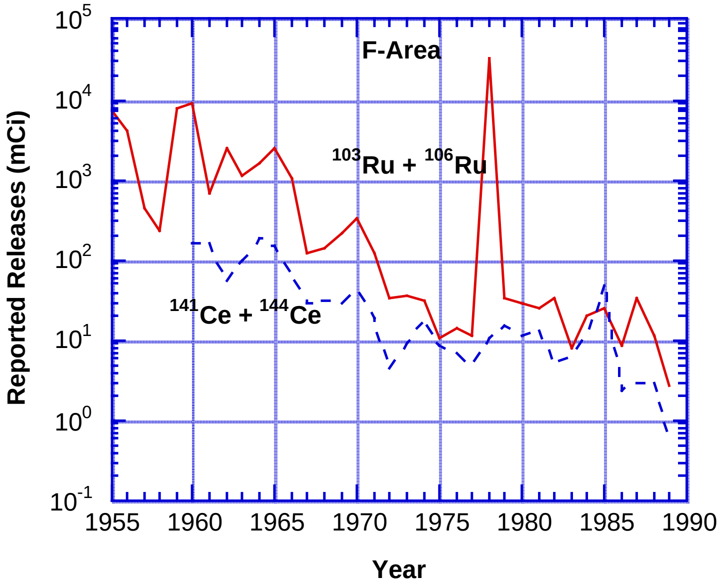

Figure 4.2-15. Reported releases of ^{103,106}Ru and ^{141,144}Ce to the atmosphere from F-Area at the SRS (Cummins et al. 1991).

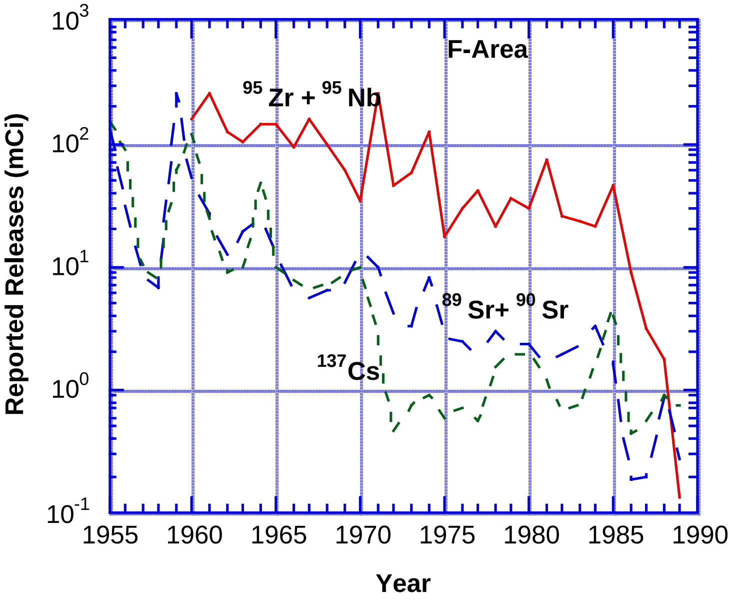

Figure 4.2-16. Reported releases of ^{89,90}Sr, ^{95}Zr-Nb, and ^{137}Cs to the atmosphere from F-Area at the SRS (Cummins et al. 1991).

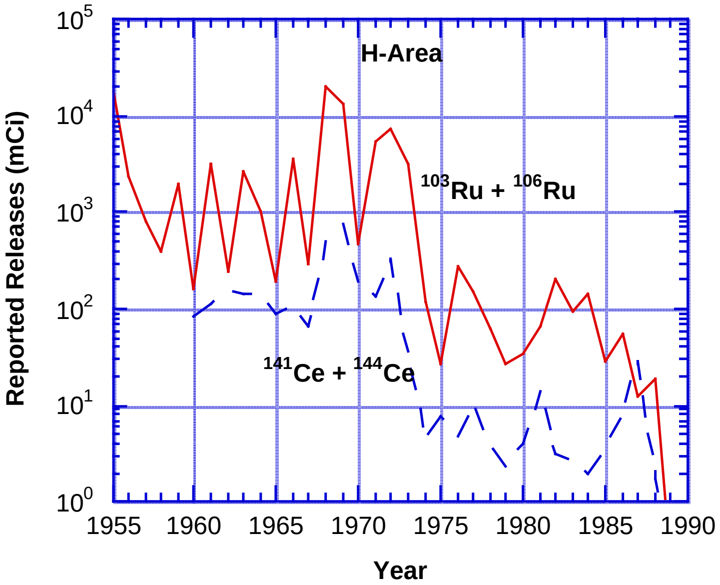

Figure 4.2-17. Reported releases of ^{103,106}Ru and ^{141,144}Ce to the atmosphere from H-Area at the SRS (Cummins et al. 1991).

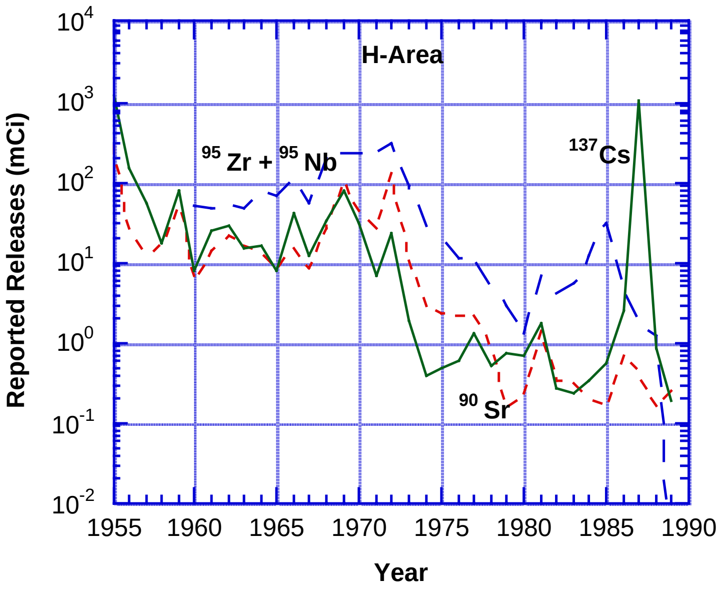

Figure 4.2-18. Reported releases of ^{89,90}Sr, ^{95}Zr-Nb, and ^{137}Cs to the atmosphere from H-Area at the SRS (Cummins et al. 1991).

The release estimates that are shown in these figures do not reflect any corrections for sampling line losses. As discussed previously, the sampling line for the F- and H-Area stacks were long and some deposition of particles would be expected. However, evaluation of such losses is problematic. Information on the sizes of particles in the stack effluents is limited and largely confined to results for plutonium. Those results are presented and discussed in Chapter 4.4. Whether the sizes of particles containing beta-gamma-emitters are comparable to those containing plutonium (from a different area in the facility) is an open question.

Some estimates of sampling line losses are also presented in Chapter 4.4. Losses of 20–30% were estimated for particles with geometric mean diameters less than 1.5 µm. That size range is consistent with normal operation of the effluent filtration systems, which would efficiently remove larger particles. However, sampling line losses during periods when there were filter failures would probably be greater because larger particles could pass through the defective filters. Filter failure events occurred during startup of both fuel processing plants and in H-Area in 1969. Calculations presented in Chapter 4.4 suggest that, for plutonium particles, sampling line losses of 70-80% may have occurred at those times.

Estimates of releases of beta-gamma-emitting particles are tabulated in an Excel® spreadsheet. The spreadsheet can be directly accessed by clicking on the following hyperlink: Ann_B-G_Part_Releases.xls. Because of the great uncertainty about the applicability of the available plutonium particle size data, these estimates (like those shown in the figures) have not been corrected for sampling line losses. For screening calculations, a cautious procedure would be to multiply releases of beta-gamma emitting particles in the spreadsheet by a factor of 4. It is unlikely that all actual releases were more than four times those that were reported; thus estimates of potential doses would likely not be underestimated by such a procedure.

SUMMARY OF RELEASE ESTIMATES

Releases of Radioiodines to the Atmosphere

The largest releases of radioiodines from SRS facilities to the atmosphere were from the tall stacks of the fuel processing facilities located in F-Area and H-Area. The main focus of the analysis was estimation of the releases from these facilities. Estimates of the annual releases of ^{131}I and ^{129}I from the F and H fuel processing plants are contained in Excel® spreadsheets, which can be accessed by clicking on the following hyperlinks: Revised_I-131_Releases_(F,H).xls and I-129_Releases_(F,H).xls. Radioiodine releases from the SRS production reactors were estimated to be more than 80 times smaller than those from the fuel processing facilities. The estimated annual releases of ^{131}I from all reactors combined are compiled in the Excel® spreadsheet, which can be accessed by clicking on the following hyperlink: Est_I-131_Releases_(Reactors).xls. Releases from research facilities at the Savannah River Laboratory were estimated to have been more than 20 times smaller than those from reactors and were not evaluated in detail. We estimated that an upper bound total for those releases is 20 Ci.

Radioiodines released in elemental form (I_2) are more likely to enter the air-grass-cow-milk food chain. For that reason, such releases have greater significance to the health of the public living near the SRS. Releases of elemental iodine were estimated separately for the reprocessing facilities and are shown in the spreadsheet referenced above. Measurements at commercial PWRs suggest that about 30% of the ^{131}I releases from the reactors may have been in elemental form. Experience at other facilities suggests that all of the release from the SRL hot cells was likely to be I_2.

Releases of Beta-Gamma-Emitting Particles to the Atmosphere

The largest releases of particles containing beta-gamma-emitting fission and activation products to the atmosphere also occurred at the fuel processing areas in F-Area and H-Area at the SRS. The particles, which are not volatile like the radioiodines, contained a variety of radionuclides. Releases of the following radionuclides were the most important: ^{89}Sr, ^{90}Sr, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{137}Cs, ^{141}Ce, and ^{144}Ce. Initial estimates were of the composite activity and individual isotopes were not measured separately for many years. Estimates of releases of these radionuclides are contained in an Excel spreadsheet, which can be accessed by clicking on the following hyperlink: Ann_B-G_Part_Releases.xls.

Release estimates for these radionuclides have not been revised to account for deposition in the sampling lines. Adequate information about particle sizes is not available to permit estimation of the correction factors. For screening purposes, a cautious approach of multiplying the release estimates for beta-gamma-emitting particles by 4 is recommended.

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