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ACRONYMS AND ABBREVIATIONS

AEC	U.S. Atomic Energy Commission
AGL	above ground level
Bq	becquerel
CAM	Continuous Air Monitor
cc	cubic centimeter
CEDR	Comprehensive Epidemiologic Data Resource
cfm	cubic feet per minute
Ci	curie
cm^3	cubic centimeter
cpm	counts per minute
CWS	Chemical Warfare Service
d $24h^{-1}$	disintegrations per 24 hours
DOE	U.S. Department of Energy
dpm	disintegrations per minute
EEOICPA	Energy Employee Occupational Illness Compensation Program Act
ft^3	cubic foot
GM	Geiger-Mueller
HFIR	High-Flux Isotope Reactor
hr	hour
kW(t)	kilowatts thermal
LAM	Local Air Monitor
LITR	Low-Intensity Test Reactor
m^3	cubic meter
mA	milliampere
MDA	
MDC	
MeV	mega-electronvolt
MFPs	mixed fission products
mph	miles per hour
mR	milliroentgen
mrad	millirad
ORNL	Oak Ridge National Laboratory
ORR	Oak Ridge Reservation
PAM	Perimeter Air Monitor
RaLa	radioactive lanthanum
RAM	Remote Air Monitor

4.1 INTRODUCTION

This technical basis document (TBD) describes the development of internal and external exposure data from onsite environmental sources of radioactive material for workers at the Oak Ridge National Laboratory (ORNL). Its purpose is to provide guidance to individuals performing dose reconstructions under the Energy Employees Occupational Illness Compensation Program Act (EEOICPA) for workers whose work history includes time at ORNL. The document addresses intakes of radioactive material that could have been present in the ambient air and ambient external exposures from process facilities, waste handling areas, and airborne noble gas emissions.

The existence of site-wide monitoring data for external exposures dating back to early in the Laboratory's history simplified the task of evaluating potential external exposures to unmonitored workers. However, despite the existence of substantial air monitoring networks both on the site and elsewhere, little information was available regarding airborne concentrations of the two principal nuclides of concern for inhalation exposure: ^{131}I and tritium. Thus, a number of conservative approaches and assumptions had to be employed to develop the requisite concentration and intake data. These results should be considered more bounding than representative.

Special Note Regarding Historical External Dosimetry Practices at ORNL
Dose reconstructors responsible for external dose assessments should note there are no known instances where control badges were stored or employed in such a way at ORNL where they would have been exposed to plant-related activity or otherwise utilized where excess exposure (i.e., exposure in excess of natural background) would have been subtracted from a worker's dosimetry result. ORNL went to a "take home" badge (i.e., security badge and dosimeter combined) in the early 1950s, and there are no indications in the records reviewed to date that the Laboratory ever employed control dosimeters for the purpose of subtracting a "background" from personnel exposures. It is known the Lab began subtracting backgrounds from take-home dosimetry results in 1989 or so, but the amount subtracted was based on controlled studies of offsite exposure rates. The signal subtracted was not based on control badge readings. Therefore, assigning external environmental dose (in addition to missed dose) should not be necessary for workers who were monitored with whole-body personnel dosimetry. The external environmental exposures addressed in this document are directed toward unmonitored individuals.

The intake and exposure rate data given in Attachments 4B and 4C of this Technical Basis Document apply to the ORNL site as a whole. Technical Basis Documents and Site Profile Documents are general working documents that provide guidance concerning the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). These documents may be used to assist NIOSH in the completion of the individual work required for each dose reconstruction.

In this document the word "facility" is used as a general term for an area, building or group of buildings that served a specific purpose at a site. It does not necessarily connote an "atomic weapons employer facility" or a "Department of Energy facility" as defined in the Energy Employee Occupational Illness Compensation Program Act of 2000 (42 U.S.C. § 7384l (5) and (12)).

4.2 HISTORIC SOURCES OF AIRBORNE RADIOACTIVE MATERIAL AT ORNL

Table 4-1 lists the principal stacks that were in operation at ORNL over its history that were used to exhaust radioactive materials. The facilities and processes that contributed gaseous radioactive effluents over ORNL's history changed somewhat with changes in the Laboratory's mission and

facilities, coupled with improvements in air cleaning technologies. However, they can be placed in three general categories:

- Cooling air exhausted from the Graphite Reactor
- High-activity offgases from process vessels
- Low-activity ventilation air from hot cells and hoods

Table 4-1. Principal elevated release points for airborne radioactive materials at ORNL.

Initial designation	Subsequent designation	Height in feet (AGL)	Years of operation	Major processes served
105	3018	200	1943–1963	Graphite Reactor, LITR
205 (pilot plant stack)	3020	200	1943–present	Dissolver offgases (pilot plant, RaLa facility), cell ventilation streams
706-D (fan house stack)	N/A	50	1944–1950	RaLa cell ventilation
900 (central offgas system stack)	3039	250	1950–present	Isotopes Area, RaLa, vessel offgas, reactors, and hot cells.
N/A	2026	75	1964–present	High-Level Analytical Laboratory
N/A	7025		1967–ca. 1989	Tritium Target Fabrication Facility
N/A	7512	100	1965–1969	Molten Salt Reactor Experiment
N/A	7911	250	1966–present	HFIR/TRU facility

The two original sources of airborne radioactive emissions at ORNL were the Graphite Reactor and the hot pilot plant (Building 205, later redesignated Building 3019). The gaseous wastes created by these two facilities were exhausted via the 105 stack and the 205 stack, respectively. The 105 stack ventilated cooling air exhausted from the Graphite Reactor, the principal radioactive constituent of which was ^{41}Ar. The 205 stack exhausted offgases from dissolving and other chemical separations processes associated with the development of the bismuth phosphate process for separating plutonium from irradiated fuel. The principal radioactive constituents in this effluent stream were noble gases and ^{131}I.

The ORNL Graphite Reactor operated from November 1943 until November 1963. It was an air-cooled graphite pile, with the exhaust cooling air ventilated via the 105 stack, which was later renumbered as the 3018 stack. The exhaust air system did not provide any holdup or delay, so the effluent stream included short-lived fission gases, their particulate decay products, and air activation products, most notably ^{41}Ar. In addition, particulate mixed fission product activity would be released following incidents of ruptured fuel slugs. The Graphite Reactor was initially operated at a maximum power level of approximately 1,000 kW(t). However, it soon became apparent the system had been significantly over-designed, and modifications were made to allow its maximum power level to be uprated to approximately 5,000 kW(t). The estimated ^{41}Ar emission rate from the 105 stack with the reactor at its initial power level was approximately 200 curies per day in 1944 (Parker 1944). The emission rate increased to approximately 500 curies per day in 1945 (Cheka 1945) following the uprate of the reactor's operating power level.

The exhaust air from the Graphite Reactor was discharged unfiltered until 1948 when a significant particle contamination problem was found on the Laboratory grounds. The Graphite Reactor was found to be one of the principal contributors to this particulate contamination, so a filter house was added between the exhaust plenum and the stack. The filtration system, which became operational in November 1948, consisted of parallel banks of roughing plus high-efficiency filters. Particulate releases from the 105 stack were largely mitigated once the filtration system was added. The 105 stack also ventilated gaseous effluents from the Low-Intensity Test Reactor (LITR) from 1949 until 1968.

Low volumes of high-activity offgases were produced from process vessels such as dissolvers, where the initial steps of chemical separations involving irradiated source materials were carried out. Examples of such operations include plutonium separations processes such as bismuth phosphate or PUREX and its variants, alternative fuel cycle processes such as Thorex, and production of radioisotopes such as ^{131}I or ^{140}Ba/La (i.e., the RaLa process). These high-activity offgas streams were ventilated via the 205 stack prior to completion of the ORNL central offgas handling system about 1950. After 1950, most of these high-activity offgas streams were ventilated via the 900 stack (later renumbered the 3039 stack) following treatment. Offgas treatment methods evolved and improved over the years and often included pretreatment (filters, scrubbers, etc.) at the generating facility prior to entering the central offgas system.

High volumes of low-activity waste gases were produced by ORNL's numerous hot cells and fume hoods. Most of these sources were ventilated locally prior to completion of the central offgas system about 1950. Following completion of the central offgas system, both filtered and unfiltered cell ventilation streams were exhausted via the 205 stack (later renumbered the 3020 stack). The filtered streams passed through a filter bank consisting of roughing plus high-efficiency filters prior to discharge. Unfiltered streams were discharged without additional treatment.

As of the late 1940s, Cheka and McAlduff (1949) reported the principal sources of airborne radioactive emissions at ORNL to be:

- Cooling air exhausted from the Graphite Reactor via the 105 stack
- Emissions from the Redox process in the Pilot Plant (via the 205 stack)
- Dissolver offgas from ^{131}I production in Building 706-C (via the 205 stack)
- Dissolver offgas from RaLa production in Building 706-D (via the 205 stack)
- Cell ventilation air from the RaLa process in Building 706-D (via the 706-D stack)
- Cell ventilation air from ^{131}I and ^{135}Xe production in Building 706-C (via roof vents)

Other sources of airborne radioactive material in this era were hot chemistry work conducted in hoods in Building 706-A. These were exhausted via roof vents. In addition, the tank farm was considered a source of airborne material from spills that subsequently dried and became airborne (Cheka and McAlduff 1949). In reality, studies in later years showed the W-9 tank in the south tank farm to be a significant source of local airborne contamination during waste transfer operations (Bradshaw and Cottrell 1954). This was presumably the case from the beginning, but was not recognized in the late 1940s. Another source of airborne radioactive material cited for this era was fugitive emissions from the 706-D Building during RaLa operations (Cheka and McAlduff 1949). In fact, Cheka and McAlduff (1949) note that offgases from dissolver operations ventilated via the 205 stack seldom contributed to local air activity, whereas emissions from the latter parts of separations operations that were vented through the D-stack, roof vents, or the building itself were large contributors of activity. This situation was mitigated soon thereafter when the 900 stack and central offgas facility were placed in operation about 1950.

Prior to 1950, airborne radioactive effluents from ORNL were discharged to stacks and vents serving individual facilities. However, the temporary nature of the Laboratory's original mission resulted in it quickly outgrowing its waste handling measures once its status was changed to one of indefinite duration. Thus, in approximately 1950, ORNL installed and placed into operation a centralized offgas handling and treatment system as part of an effort to upgrade original, temporary laboratory facilities to those designed for long-term use. The central offgas system expanded over the years to accommodate expansion of the laboratory facilities and mission. A significant expansion in capacity of the system took place between 1956 and 1958. The primary stack used by the central offgas system was the 900 stack, which was renumbered as the 3039 stack around 1953. High-activity

gaseous effluent streams such as those from process vessels used in isotopic separations were exhausted via the 900 stack following treatment. High-volume, low-activity streams, such as those from hot cells and fume hoods, were exhausted via the 205 stack.

Following completion of the centralized offgas facility in the early 1950s, two principal waste gas systems were in place at ORNL. The primary system handled high-concentration effluent streams such as those from chemical processing of materials such as irradiated nuclear fuels. This system discharged via the 250-foot 3039 stack after passing through various filters and precipitators. The secondary system was a common ventilation system for hot cells and laboratory hoods that discharged via either the 3020 or 3039 stack after passing through filter banks. The precise date that this system was completed is unknown, other than it was prior to 1959. The waste gas system for the Graphite Reactor remained unchanged and was independent of the two central systems.

The vessel offgas treatment system of the central offgas facility originally consisted of a Cottrell electrostatic precipitator followed by high-efficiency filters. The precipitator consisted of 23 tubes, each 8 inches in diameter and 12 feet long. It operated at 52,000 volts and 130 mA, and was served by a continuous, recirculating-water flush system (Browder 1959). Additional treatment measures were added over time, such as scrubbers for radioiodines added in 1961 (Ohnesorge 1986). The report of the Applied Health Physics division for the first quarter of 1961 makes mention of the "installation of more efficient cleaning equipment for off-gas streams," which appears to have occurred some time in 1960 (Hart 1961a). Charcoal filtration was added to the offgas line of the liquid waste storage tank used for waste from operations involving ^{131}I about 1970 (Ohnesorge 1986). The Cottrell precipitator was eventually taken out of service, and demolition of the unit began in late 1981 (Auxier and Oakes 1982). The demolition of the precipitator was an element of a larger effort that began in October 1981 to upgrade 3039 stack services. Upgrade and removal efforts for the Laboratory's cell ventilation system were also initiated during this era.

4.3 HISTORIC ONSITE ENVIRONMENTAL MONITORING PRACTICES AT ORNL

4.3.1 Airborne Radioactivity Measurements: November 1943 through March 1945

Initial air monitoring efforts at ORNL focused on measurement of outdoor exposure rates from the radioiodine and noble gas emissions from the two stacks operating at that time: the 205 stack, which vented offgases from dissolving operations associated with the development of the bismuth phosphate process; and the 105 stack, which vented the discharged cooling air from the Graphite Reactor. Both of these stacks are 200 feet tall. The potential for internal exposure associated with radioiodine emissions was recognized, but it was felt that relatively little iodine was released from the Pilot Plant stack relative to the tolerance level of the day, which was 1×10^{-13} Ci cc^{-1} (Parker 1944). For comparison, this same reference states the tolerance levels (concentrations) for xenon and argon to be 2.3×10^{-11} Ci cc^{-1} and 1.8×10^{-12} Ci cc^{-1}, respectively. Parker (1944) also states that selected staff members were checked for radioactivity of the thyroid gland with none being found, but neither the instrumentation used nor its sensitivity are discussed. The principal radioactive constituents of the effluent from the 205 stack were xenon and iodine. The activity in the 105 stack effluent was primarily ^{41}Ar.

Initial monitoring of outdoor airborne radioactivity at ORNL was performed using Lauritsen electroscopes to make exposure rate measurements on occasions when the plume from the 205 stack reached ground level on the site. This monitoring method was employed from November 1943 until March 1944 (Cheka 1945).

In May 1944, systematic monitoring of outdoor exposure rates on the ORNL site was begun using what was known as an X-22 ionization chamber (Cheka 1945). These units, sometimes referred to as "kangaroo chambers," were condenser-type chambers based on the design of the pocket meters in use at the time. The chambers had 0.125-inch-thick bakelite walls and an active volume of 4,900 cm^3. They therefore had a relatively flat energy response and were essentially insensitive to beta activity. The chambers were calibrated using a radium gamma source (Parker 1944). A number of the chambers were modified to make them beta-sensitive by making 48 large holes in the walls and then covering the openings with graphite-backed cellophane or aluminum foil. The aluminum foil was preferred because it stood up better to weathering. Insulators for the collecting electrode in the X-22 chambers were selected to limit leakage.

In 1944, X-22 measurements were collected daily from chambers at eight locations. Some of these were on the site and some were in the surrounding area, out to about a mile away (Parker 1944). Cheka (1945) states X-22 chambers were placed at nine locations on and around the site by June 1944. Of these nine locations, it appears that only the chambers located at the "rabbit cages" (800 feet from the 205 stack) would have provided meaningful data about exposures to onsite personnel during that era. The location at White Oak Dam was not included in the daily monitoring routine.

Each X-22 location had two stands designed to hold two chambers each (four chambers total) at a height of 5 feet off the ground. The intent was to have two of the standard gamma-only chambers and two of the modified beta-sensitive chambers at each location, with one of each pair acting as a control on the other. This was analogous to the Laboratory's external dosimetry practice at the time, under which personnel were issued pairs of pocket meters, with one acting as a backup for the other in the event of a questionable reading. In practice, however, Cheka (1945) reported there were typically two gamma chambers deployed at each monitoring location, but the fact the modified (beta) chambers did not stand up well to weather made them unreliable. Thus, data from the beta-sensitive chambers is said to be "fragmentary."

Observation and trending of readings from the X-22 stations showed the principal source of gamma exposure on the ORNL site during this time was ^{41}Ar discharged from the 105 stack and not xenon/iodine emissions from the 205 stack. [However, it was noted that when the bismuth phosphate process in the 205 building was shut down in early January, 1945, the monthly average exposure per X-22 station dropped 11% even though activity from the 105 stack increased by 8.5% (Wirth, Morgan, and Curtis 1945).] It was also noted that the radiation levels measured by the X-22 chambers were more dependent on their elevation than their distance from the stacks, with the exception of the station at the base of the stacks (which gave the highest reading of all). Diurnal variability was also noted, with the stations giving higher readings during the day than at night. The X-22 data also showed the ratio of the response of the beta-plus-gamma chambers to that of the gamma-only chambers to be independent of the radiation source (i.e., the chambers gave the same ratio whether they showed activity over background or not). Thus, site-related activity of that era does not appear to have had a strong beta component.

In addition to monitoring via the X-22 chambers, film badges were deployed at locations further away from the site to provide monitoring in areas where the need for routine reading made use of the X-22 chambers impractical. According to Parker (1944), the local X-22 chambers were read two or three times a day as of September, 1944. Film badges were placed at additional locations and were collected weekly starting on April 13, 1944. Parker states "regular film badges were used for this purpose and both beta and gamma ray measurements were obtained." He goes on to say, "There has never been a measurable record in a one-week period," which is not surprising given that the limit-of-detection for the film badges of that era was on the order of 30 mR.

Other methods of environmental monitoring at ORNL during its early period of operation included making exposure rate measurements in areas where it was evident, either visually or by odor, that the plume from the 205 stack had touched down locally (on the site). When this occurred, exposure rate measurements would be attempted using thin-walled Lauritsen electroscopes. Parker (1944) reports positive electroscope readings in three of eight such instances, with exposure rates of 1 to 2 mR hr^{-1} being observed for periods of 1 to 2 minutes. Grab sampling was performed early on using evacuated steel cans having a volume of approximately 10.5 liters. However, these samples could not be quantitatively assayed due to the lack of gaseous calibration standards. Precipitator sampling was used to determine natural radon and thoron concentrations. Using an assumed collection efficiency of 70%, Parker (1944) reports the measured range of radon concentration to be 1×10^{-17} Ci cc^{-1} to 5×10^{-16} Ci cc^{-1}. The range of thoron concentrations measured was 8×10^{-19} Ci cc^{-1} to 1×10^{-17} Ci cc^{-1}.

Cheka (1945) described experiments performed in November and December 1944 to determine ^{133}Xe concentrations in outdoor air downwind from the 205 stack. An accumulator was used to collect and liquefy 4,500 ft^3 of air over the course of an hour. The liquefied air was then evaporated through a xenon trap, and the collected sample subsequently counted using a mica-window Geiger-Mueller (GM) tube. Fifteen runs were made at distances ranging from 150 feet to 3 miles from the plant, but most were made between 0.1 and 0.4 mile. The concentrations determined ranged from zero to 2.8×10^{-11} Ci ft^{-3}, and averaged 5.06×10^{-12} Ci ft^{-3}. The highest values were obtained at 0.2 and 0.4 mile from the 205 stack. Note: 2.8×10^{-11} Ci $ft^{-3} = 9.9 \times 10^{-10}$ µCi cc^{-1}.

An experimental monitor consisting of a glass-walled GM tube and a scaler/recorder unit was installed approximately 800 feet northwest of the 105 stack in the latter part of 1944 (Cheka 1945). This unit began giving "fairly good" results in early 1945, with a maximum exposure rate of 0.009 mR hr^{-1} net (0.024 mR hr^{-1} gross) being measured due to ^{41}Ar emission from the 105 stack during a period of southeasterly winds (i.e., blowing toward the monitoring unit).

Cheka (1945) states that the program of monitoring via the X-22 chambers concluded on March 31, 1945, as a "fairly complete" picture of the distribution of gaseous wastes obtained and it had been established that onsite exposure rates were within an order of magnitude of natural background.

4.3.2 Concentration Measurements for Airborne Radioactive Material

Following the conclusion of the X-22 program in March 1945, subsequent monitoring efforts for airborne radioactivity at the Laboratory switched in focus from external exposures to potential internal exposures from respirable materials (such as ^{131}I and particulate matter). The date when they first appeared is unclear, but at some point prior to mid-1947, a system of Continuous Air Monitors (CAMs) began to evolve on the ORNL site. The initial complement of monitors consisted of three units. In October 1947, the three monitors were in the vicinity of Buildings 706-A, 115-B, and 735-B (Burnett 1947a). They were in these same locations a year later in October 1948 (Brown 1948). These three units were the extent of the local air monitoring network until mid-1949, when the system was expanded to four monitors. In December 1948, the unit originally located at Building 115-B was moved to south of Building 105, and the unit originally located at 735-B was moved to west of the south tank farm (Cottrell 1948a). The system was subsequently expanded to 10 monitors around late 1949. Note: A number of CAMs were employed for monitoring airborne radioactivity concentrations inside various ORNL facilities in addition to the three units originally installed to monitor outdoor air on the site.

The CAMs were housed in louvered wooden enclosures, and drew air through a cylinder of filter paper inside a cardboard tube. The filter cartridge was shielded by a lead pig, and a GM tube was "fixed at its axis" (Cheka and McAlduff 1949). The GM detector was connected to a recording device

(tape counters initially, then chart recorders as technology improved). The sample flow rate for the CAMs was reported by Morgan (1948) to be 7,000 ft^3 per 24-hour day, which equates to roughly 5 cfm. Cheka and McAlduff (1949) also assert a 5-cfm flow rate for these units. However, a memo describing an investigation of elevated airborne concentrations noted in July 1947 implies a flow rate of 3 cfm for that time (Burnett 1947b). In 1953, the filter media used in the CAMs is reported by Bradshaw and Cottrell (1954) to have been "H and V Number 70 paper," where H and V stands for the Hollingsworth and Vost Company. The particulate filters used in the CAMs were eventually changed to glass fiber filters.

Sometime around 1949, the ORNL Applied Health Physics Division began reporting data from the 10 onsite CAMs in its periodic summary reports. CAM readings had been included in monitoring reports prior to this time, but only when instances of high air activity were noted, and even then the information was often qualitative. The data provided in the Applied Health Physics reports were weekly average concentration data for the 10 onsite CAMs averaged over the reporting period (either quarterly or semiannually). The last report for a given calendar year gave the annual average of the weekly CAM results. It is not clear if the onset of reporting weekly average results for the CAMs in the Health Physics Division reports coincided with a change to a weekly exchange rate for the CAM filters. However, it is known the exchange rate was reduced to weekly at some point, and this appears to have happened prior to May 1951. In addition, the original scaler recording devices for the CAMs were changed to rate meters sometime in the early 1950s. Prior to this change, CAM results (i.e., *in situ* airborne concentration values) were derived from the number of counts integrated over an arbitrary time interval. The original scalar design, and the associated microswitches in particular, were a source of significant reliability problems for the original CAMs. The recording instruments were upgraded in December 1947 in an effort to improve reliability (Burnett 1948). The waste monitoring report for the week ending November 7, 1948, says the Instrument Division gave a delivery date of January 15, 1949, for the first of 10 logarithmic count rate meters to be used with the CAMs (Cottrell 1948b). However, the scalars were still in use through March 1949, but the transition to logarithmic count rate meters was completed by the early 1950s.

During the first 6 months of 1956, an additional set of seven CAMs was activated. Six of these were placed at the U.S. Atomic Energy commission (AEC) Perimeter Guard Portals (gates). The seventh unit was on the Clinch River at Hickory Creek Bend. These new units included gummed-paper fallout trays and rainout collectors for measuring airborne particulates (fallout). The additional seven CAMs installed at the perimeter locations became known collectively as the Perimeter Air Monitors (PAMs), while the 10 monitors on the ORNL site became known as the Local Air Monitors (LAMs). The LAMs were outfitted with fallout trays and rainout collectors by the latter half of 1956.

Along with the PAMs, another three CAM stations were installed sometime in the first half of 1956 at three remote locations: Berea, Kentucky; Corryton, Tennessee; and Kingston, Tennessee. These three stations were referred to as the Remote Area Monitors (RAMs). Data from the PAMs and RAMs were first reported in the Applied Health Physics semiannual report for the second half of 1956 (Hart 1957). However, the RAM stations at Corryton and Kingston, Tennessee, were used only in 1956. Data are reported for the station at Berea, Kentucky, only in 1957 and 1958.

Subsequently, an additional seven RAMs were activated at various Tennessee Valley Authority (TVA) dams prior to 1959, bringing the RAM network to a total of 8 monitors for the calendar 1958 reporting period. These eight stations ranged in distance from the ORNL site from 12 to 120 miles. The RAM station at Berea, Kentucky, was retired by 1962, reducing the RAM network to seven stations ranging in distance from 12 to 75 miles from ORNL. A new RAM station at the TVA Power Service Center in northeast Knoxville, Tennessee, was added to the network in 1965 to bring the total back to eight.

Note: unlike the LAMs, which included GM tubes coupled first to scalers then to logarithmic count rate meters, PAMS and RAMs contained no monitoring equipment.

The ORNL LAM network was upgraded beginning in 1962 by replacing the fixed filter with a step-type, moving-tape media (Morgan, Davis, and Hart 1963). The filter head housing the moving tape was monitored with an unspecified "beta-gamma monitoring device." The impetus for changing the network from a fixed-filter medium to a moving tape appears to have been to provide greater versatility in the event of a radiological emergency.

Starting in the second quarter of 1963, 12 new CAM stations began to be integrated into the LAM network. These monitors were added to expand coverage in response to a significant increase in the physical size of ORNL due to new facility construction (Davis and Hart 1963). However, data from only two of these stations were ever included in the network averages reported by the Health Physics Division. These two monitors were east of the 4500 building and at the HFIR complex. A 23rd LAM station, at Walker Branch, was added to the network by 1976. The results from this station were included in the network averages beginning with that year, bringing the total number of LAMs in the average to 13: the original 10 units plus the units east of the 4500 Building, at the HFIR complex, and at Walker Branch. The Walker Brach unit was approximately 2.5 miles east of the main Laboratory complex at the Low-Level Counting Laboratory.

The PAM station at Hickory Creek Bend on the north side of the Clinch River was relocated in 1962 to the south side of the river and farther away due to the impending flooding of the Melton Hill Reservoir, scheduled for 1963 (Morgan, Davis, and Hart 1963). Two new PAM stations were added to the monitoring network in 1964, bringing the total number to nine. Another two stations were added by 1983, making a total of 11.

Radioiodine sampling capability was added to the PAM network in the second quarter of 1963 by the installation of activated charcoal cartridges in the air sampling line between the particulate filter and the air pump. The cartridges were removed weekly and assayed for radioiodine activity via gamma spectrometry (Davis and Hart 1963). Radioiodine (^{131}I) concentration data measured via the PAM network were first reported for the fourth quarter of 1963. Radioiodine sampling capability was added to the LAM network by 1966. According to the Health Physics Division's annual report for 1966, the MDA for the ^{131}I sampling protocol employed in the LAMs and PAMs was 20 dpm, which corresponded to concentrations of 1.0×10^{-14} µCi cc^{-1} for the PAM stations and 2.0×10^{-14} µCi cc^{-1} for the LAM stations (Morgan, Snyder, and Davis 1967). ORNL practice at the time was to assign a concentration equivalent to 10 dpm for any charcoal cartridges showing a total amount of ^{131}I less than the 20 dpm MDA (Morgan, Snyder, and Davis 1967).

Beginning in 1975, the ORNL Health Physics Division began reporting isotopic analyses for filters collected from the LAM, PAM, and RAM stations. The results are reported as quarterly composites, but the specifics of how the analyses were performed or if all of the filters from all of the monitors were included are uncertain. The isotopic analyses show the dominant nuclide collected on the filters for all three networks to be ^{7}Be, which is formed naturally in the upper atmosphere by cosmic ray interactions with nitrogen and oxygen. Other nuclides observed on the filters, although in much lower concentrations, included ^{95}Zr/Nb, ^{106}Ru, ^{125}Sb, ^{137}Cs, and ^{144}Ce (Auxier, Davis, and Turner 1976). No appreciable differences are seen in the isotopic mix data for the three air monitoring networks. By the early 1980s, as the airborne beta/gamma concentrations measured by the three air monitoring networks continued to drop, the relative concentration of ^{7}Be continued to climb to nearly 100% of the total. For instance, in 1979 ^{7}Be accounted for 95.5% of the composite concentration for the LAM network, with fission product nuclides accounting for 4.1%. The difference between the two fractions is made up by small concentrations of primordial species. Fission product nuclides present in the

LAM samples for 1979 were ^{90}Sr, ^{106}Ru, ^{125}Sb, ^{137}Cs, and ^{144}Ce (Auxier and Davis 1980). However, the ^{7}Be concentrations reported for calendar 1981 were significantly lower than those for previous years, implying a possible change in the analysis procedure. By 1977, filters collected from the LAM, PAM, and RAM stations were being analyzed for alpha radioactivity in addition to beta/gamma. Silica gel cartridges for tritium monitoring were added to selected LAM stations by 1980; however, no results (i.e., tritium concentration data) from this effort have been found in the references reviewed to date. By 1983, the filters were counted weekly for gross beta and gross alpha, and all filters for a given network (LAM, PAM, or RAM) were composited on a quarterly basis for specific nuclide identification.

In response to changes in the regulatory framework placing increased emphasis on quantification of releases of pollutants from all pathways, a major reorganization of the various air monitoring networks in operation throughout the Oak Ridge Reservation (ORR) took place during 1985. As a result, the ORNL LAM network that had been in place for decades was retired, with the exception of a few stations that remained in service to form two new networks. One of these two new networks was established such that it covered the perimeter of the ORNL site. This new ORNL perimeter network consisted of four of the original LAM stations. The second new network was defined such that it covered the ORR perimeter. This new ORR perimeter network consisted of 11 stations: nine of the original PAM stations plus two of the original LAMs (MMES 1986). The 1985 reorganization does not appear to have affected the RAM network, other than all the air monitoring stations making up all the networks were renumbered during this time. However, annual average concentration data from the ORNL LAM network do not exist after 1984.

4.3.3 Studies and Measurements of Onsite Particulates

With the recognition of the significance of particulate emissions at the Hanford Site in 1948, studies were conducted at ORNL beginning in May of that year to determine if a similar situation existed there. Indeed, a significant “particle problem” (as it became known) was confirmed, and this launched a program of monitoring for particulate radioactivity that continued for decades. Thus, there are voluminous data on particulate monitoring in ORNL’s onsite air monitoring history. However, the focus of the particulate monitoring program was quantification in terms of numbers of particles and fallout rates and, as such, the data are of limited use for estimating potential exposures to workers. For example, data from sedimentation frames were initially quantified in terms of particles per day (and later, particles per unit area), and air concentration data were expressed as particles per unit volume. In addition, the methods used to assess particle fallout rates and concentrations were preferentially biased toward larger nonrespirable particles. There is some information on particle activity distributions in the periodic monitoring summaries, but this information says nothing of particle sizes and thus is not useful for estimating inhalation intakes. This does not mean, however, that inhalation intakes of respirable particulates represent an unmonitored pathway. The numerous LAM stations positioned on the Laboratory grounds were designed to collect respirable particulate matter, and particulate air concentration data from these units are available as far back as 1947.

This section of the TBD provides a summary of ORNL particulate monitoring practices, starting with the original recognition of the “particle problem” back in 1948. The data collected from the particulate monitoring program are of limited use for estimating inhalation intakes. However, the investigations into particulate emissions at ORNL are still of interest because they provide insight into issues such as sources of airborne particulates, different radionuclides that were released, remedial actions taken to reduce emissions, etc. This information was used as an aide to interpreting particulate data collected by the LAM network. In addition, the information was helpful in that it showed consistency between results from the onsite environmental monitoring program (for airborne concentrations and exposure rates) in the early years of Laboratory operation, and events that affected particulate emissions, such as installation of air cleaning measures or process upsets.

Following the recognition of the "particle problem" in May 1948, the Laboratory began an intense, large-scale program of investigations into the sources of particulate emissions, numbers of particles being released, and their physical nature. The initial phase of this effort began in June 1948, and by July of that year it had been shown that large (nonrespirable) particles could be found almost anywhere on the site, and that the principal source of these was the Graphite Reactor (Cheka and McAlduff 1949). This prompted Laboratory management to institute a large-scale program of measures to eliminate or control these particulate emissions in August, 1948 (Cheka and McAlduff 1949). Particle control measures included installing filtration on the gaseous effluent streams subsequently found to be sources of particulate emissions, and dust (resuspension) control measures.

Initially, the most problematic source of particulate emissions at ORNL was the Graphite Reactor. Specifically, the problem was high-specific-activity UO_2 particles from ruptured fuel slugs being entrained in the cooling air exhaust stream. In June 1948, particles collected in the exit air duct of the Graphite Reactor (and thus upstream of the stack) over a period of 4 days showed particles ranging in size from 10 to 100 microns (Unknown 1948). Subsequent surveys around the site showed the largest particle concentrations (in terms of particles per unit area) were in the vicinity of the Graphite Reactor exhaust stack. These particles are reported to have ranged in size from 90 to 100 microns (Unknown 1948). However, Cheka and McAlduff (1949) report sizes ranged from approximately 80 to 400 microns. Newman (1948) reported particles found by Health Physics being on the order of "a few tenths of a millimeter in diameter" and having beta count rates of 50,000 to 100,000 cpm. Follow-up investigations on particles collected in this initial phase of the large-scale investigation showed them to be UO_2, but with a high percentage (in terms of alpha activity) of plutonium. Cheka and McAlduff (1949) reported that plutonium accounted for a weighted average of 88.9% ± 4.2% of the alpha activity. In addition, the particles contained mixed fission products and showed variable decay rates, implying material of different ages was being entrained and released.

Following the discovery of large numbers of particles in the vicinity of the 105 stack, surveys in freshly graded areas and collections performed using sedimentation frames showed some particulate emissions did remain airborne. The recognition that the possibility of an inhalation hazard existed prompted corrective actions, principally, the installation of a filtration system between the Graphite Reactor exhaust air plenum and the stack. A filter house was constructed and put into operation on November 15, 1948 (Unknown 1948). Better surveillance methods for detecting and preempting slug ruptures were also initiated.

Along with the particulate emissions from the Graphite Reactor's cooling air exhaust, the second-most problematic source of onsite particulate emissions at ORNL was the RaLa process and its associated liquid waste handling operations. Particulate sampling of air exhaust streams from Redox operations (in Building 205) and RaLa operations (in Building 706-D) in 1948 showed the RaLa process produced radioactive particles "at a rate greater by several orders of magnitude than any other known source" (Unknown 1948). In contrast, the Redox process was deemed "a very minor contributor of radioactive particles" (Unknown 1948). Identification of the RaLa process as a major contributor of particulate emissions prompted corrective actions similar to those taken for the Graphite Reactor (i.e., installation of filters in the offgas streams).

The nature of the particulate emissions from the RaLa process (specifically, from the cell ventilation air) was different than those from the Graphite Reactor. The particles from the Graphite Reactor exhaust air flow were UO_2 containing plutonium and mixed fission products, and thus were discrete in nature. In contrast, Cheka and McAlduff (1948a) described particulate emissions from the RaLa process as more uniform in activity and in some cases "diffuse" in nature, "as if due to a liquid." Cheka and McAlduff (1949) goes on to say that it is unclear if what they observed on the

autoradiographs were truly particles, or liquid droplets. They also point out that emissions from the RaLa process contained very few, if any, large particles, and the "particles" observed were presumed to be minute and of high specific activity. The particulate activity collected in the vicinity of the 706-D Building (RaLa process) was short-lived in relation to that collected in the vicinity of the 105 stack (Graphite Reactor).

The studies of sources and magnitudes of radioactive particulate emissions at ORNL begun in May 1948 led to increased monitoring of gaseous effluent streams and a program of routine surveillance and monitoring for particulate contamination. The particulate contamination and monitoring program was carried out by the Health Physics department, and initially included grab sampling, collection of particles using sedimentation frames, and the local CAMs. Grab sampling was performed using Filtrons, which drew air through a particulate filter at a rate of approximately 5 cfm. Sedimentation frames were used to monitor the rate of fallout of heavier particles that were contributing to ground concentration, whereas the Filtron and CAM data were used to determine particle densities in air (Cheka and McAlduff 1949). For each of these collection methods, the number of particles collected (either per unit volume or per unit area) was determined via autoradiography using X-ray film [reported by Cheka and McAlduff (1949) to be 14- $\times$ 17-inch Eastman "Blue Brand" film]. Morgan (1948) describes the process as a 24-hour exposure with each visible spot on the film counted. Morgan also states that this process would show particles ranging in activity from less than 100 to several thousand dpm. Cheka and McAlduff (1949) report the detection threshold for the 24-hour autoradiography to be equivalent to 0.8 count per minute with a mica-window counter in a 10% geometry. Note: The autoradiography method used for counting particles could only resolve the larger, higher-activity particles.

In the case of the sedimentation frames, the particles were originally collected in a fiberglass mat in a 3- by 4-foot frame. Following collection, the material in the mat was shaken onto an area sized such that it could be covered by a sheet of X-ray film (Cheka and McAlduff 1948b). However, it was later found that the fiberglass mat was retaining too much of the collected material, so it was replaced with heavy brown wrapping paper (Cheka and McAlduff 1948a). The process was again modified; rather than being shaken onto an area for radiography, the collected material was vacuumed using a "Sanitizor," which was a vacuum cleaner with a replaceable filter paper bag (Cheka and McAlduff 1949). The vacuuming technique is said to have done a better job of collecting the material adhered to wrapping paper (Cheka and McAlduff 1949). Counting the material in the filter bag worsened the detection threshold for the 24-hour radiography to 3.5 cpm equivalent (for a mica-windowed counter in a 10% geometry).

With regard to particle sizes, Newman (1948) says experiments showed the Filtron units had a low efficiency for large particles (due to horizontal opening and low inlet velocity), but that those units should have been "reasonably efficient" for particles smaller than 10 microns. Morgan (1948) gives a sample flow rate for the Filtrons of approximately 300 cfm. However, Cheka and McAlduff (1948b) report the sampling rate as 300 ft^3 per hour, or 5 cfm. The latter value is consistent with the flow rate reported for CAMs in both Morgan (1948) and Cheka and McAlduff (1948a). Newman (1948) asserts the sedimentation frames would probably not have been effective for capturing particles in the 5- to 10-micron range, but should have been satisfactory for larger particles. A caveat is that sedimentation frames would have shown fewer particles as distance from the source increased, because larger particles would have settled out. Comparisons between particulate concentration data from collections using Filtrons and those made using Sanitizors in the fourth quarter of 1948 showed significant differences between the two. The Filtrons showed much higher concentrations than the Sanitizors, suggesting particulate emissions from chemical separations activities included a submicron component that passed through the Sanitizor filter bag. Studies conducted to determine solubilities of particulate material collected in the vicinity of the Graphite Reactor and Building 706-D

(RaLa operations) showed material collected near the reactor to be almost exclusively insoluble in water, whereas most material collected in the vicinity of Building 706-D was water-soluble (Cheka and McAlduff 1949).

Other methods used for air sampling during this initial intense period of investigation into the "particle problem" included precipitrons, large air filtering units borrowed from the U. S. Public Health Service, and airway "Sanitizors," which are described as a type of vacuum cleaner with a removable filter bag (Cheka and McAlduff 1948c). The Public Health Service units drew 240 cfm of air through 10 square feet of CWS 6 filter paper, which is asserted to have a collection efficiency of 99.97% "for the most penetrating particle size" (Cheka and McAlduff 1949). For the Sanitizor units, a flow rate of 56 cfm was used for calculation (Cheka and McAlduff 1949). The same reference states that the filter bag was estimated to have a collection efficiency of better than 95% for particles larger than 1 micron, and the inlet flow and collection nozzle were such it was unlikely that particles larger than 10 microns would be collected.

Another element of the large-scale investigations of the "particle problem" was the start of an investigative nose swabbing program on September 17, 1948 (Cheka and McAlduff 1948a). However, the program was discontinued shortly thereafter (December 1948), "since no positive evidence of the presence of radioactivity in the lungs was obtained and no reliable quantitative measurements could be made" (Unknown 1948). As conceived, the nose swabbing program was to involve the selection of 50 individuals who would be requested to stop by the dispensary each day on completion of their shifts. Participation was good at first, but waned considerably over time, and this might have played a part in the decision to discontinue it. Of the individuals who did show up at the end of a given day, a nasal smear was collected using a cotton swab. The swabs were sliced open, counted in a "standard beta-gamma counter at 10% geometry," and autoradiographed for 24 hours (Cheka and McAlduff 1948c). A total of 767 nasal smears was collected between September 17 and November 17, 1948, with particles detected on 66 of these (8.6%) (Cheka and McAlduff 1948c). Cheka and McAlduff (1948c) noted the percentage of positive nasal smears (for particles) correlated with the number of persons participating in the program for a given period. However, they also noted the incidence rate for positive nasal smears was 12.3% for persons working in the 105 Building (Graphite Reactor) versus a rate of 7.8% for other workers. The apparent incidence rate of 12.3% for 105 Building workers came from a sample size of 138 smears, and the 7.8% rate from the remaining 629 smears. The authors note the classic problem associated with nasal smear data, the interpretation of negative results.

Following the initial, large-scale investigation into airborne particulate emissions at ORNL in the latter half of 1948, studies in 1953 documented by Bradshaw and Cottrell (1954) showed a significant correlation between particulate contamination and the RaLa process. However, the principal source of this contamination was not the 900 area stack (to which all RaLa gaseous wastes were vented starting in 1950). Rather, the primary pathway through which the RaLa process contributed to local air contamination was the W-9 underground storage tank in the south tank farm. The tank, which received liquid wastes from RaLa separations, was vented to the atmosphere. Beta concentration measurements in the immediate vicinity of the tank vent were as high as 2×10^{-5} µCi cc^{-1} averaged over a 24-hour period between November 5 and November 25, 1953 (Bradshaw and Cottrell 1954). This period bracketed RaLa Run No. 54. Samples collected in this area prior to the first wastes being jetted to the W9 tank showed half-lives of greater than 136 days, and two principal components with beta energies of 0.35 and 3 MeV (Bradshaw and Cottrell 1954). In contrast, samples collected during various stages of the RaLa run showed initial half-lives of 8.5 to 22 days. The same samples showed half-lives of 21.5 to 58 days after approximately 1 month of decay. Radiochemical analyses were performed on some of these samples and are reported in Bradshaw and Cottrell (1954, Table VI). These analyses showed roughly 50% to 65% of the activity to be from total rare earths (cerium et al.),

with the next largest constituent (on an activity basis) being strontium, primarily ^{89}Sr. Other constituents included ruthenium, barium, zirconium, niobium, and tellurium/iodine. The difference in total activity collected on filter paper media versus that collected using a bubbler sampler indicated "that the major portion of the activity escaping the W-9 tank was in particulate form" (Bradshaw and Cottrell 1954).

Bradshaw and Cottrell's data prompted a subsequent experiment during RaLa Run 57 (in July 1954) to assess the effectiveness of a fiberglass filter on the W-9 tank vent (Bradshaw and Cottrell 1954). The filter reduced local air concentrations in the vicinity of the tank vent by about 2 orders of magnitude over those measured during RaLa Runs 52 and 54. However, a bigger reduction in local air activity was achieved by sealing off the opening around the tank liquid level indicator. Other openings in the tank were sealed prior to the start of the experiment. Note: It is not clear from the report if the seals on the tank openings and the filter on the vent line were left in place after the experiment was completed.

A significant increase in particle concentrations detected by the onsite CAMs in the first half of 1956 prompted an investigation into the cause. It was found the filters downstream of the Cottrell precipitator in the central offgas system serving the 3039 stack had been eaten away by acid fumes (Hart 1957). The filters were replaced and a "considerable decrease" in particle concentrations was subsequently noted (Hart 1957).

By June 1953, the methods used to measure particulate fallout rates and concentrations on the ORNL site consisted of the network of 10 LAMs, sedimentation trays (then referred to as collecting frames), and four U. S. Public Health Service air samplers. However, by July 1953, it appears that use of collecting frames had been discontinued, and only particulate concentration data were reported in terms of particles per 1,000 ft^3 of air. Data from the collecting frames were reported on a rate basis, as particles per 24 hours. Starting in the fourth quarter of 1954, particulate data from the 10 LAMs were reported in terms of concentration in particles per 1,000 ft^3 of air and in terms of an activity distribution. Activity was determined by comparing autoradiograph images from field samples to those from a particle of known activity. Activity was reported in terms of disintegrations per 24 hours. The activity distributions were given as numbers of particles in each of four activity ranges: less than 10^5 d $24h^{-1}$, 10^5 to 10^6 d $24h^{-1}$, 10^6 to 10^7 d $24h^{-1}$, and greater than 10^7 d $24h^{-1}$. (These distributions do not provide information on particle sizes beyond what is known about the physical characteristics of the LAMs and the filter papers employed.) By the first half of 1955, use of the Public Health Service samplers appears to have been discontinued, leaving the onsite LAMs as the sole means for determining onsite particulate concentrations. This was the case until sometime in the first half of 1956, when gummed-paper fallout trays were collocated with new PAM stations at the AEC perimeter guard stations and at three remote locations. The onsite LAMs had fallout trays added, and collectors for measuring fallout in rainwater were added to all of the CAM stations during this time.

The gummed-paper fallout trays deployed at the air monitoring stations are described as cellophane sheets 1 square foot in area with a gummed surface on one side. The sheets were mounted on an aluminum tray with the gummed side facing up (Hart 1957). The sheets from the fallout trays (LAMs, RAMs, and PAMs) were collected weekly and assayed in terms of microcuries per square foot.

The material collected in the gummed-paper fallout trays was assayed first by autoradiography and then by beta/gamma counting. Autoradiography was performed by covering the gummed paper with a sheet of rubber hydrochloride, cutting it from the collection frame, and mounting it on a sheet of Kodak Blue X-ray film (Browder 1959). The film was exposed in a dark room for 24 hours, after which spots created by radioactive particles were counted via the unaided eye. Activity ranges for the

particles were determined by comparing the diameter of the spots to those created using particles of known activities.

Following radiography, the material collected on the gummed paper was beta/gamma counted by folding the paper into a small square, placing it in a crucible, and ashing it in a muffle furnace. The ash was transferred to a stainless-steel counting dish and counted in a gas-flow proportional counter (Browder 1959). Activity was asserted in microcuries and results were reported as microcuries per square foot.

Experiments performed in 1953 using Millipore filters suggested the autoradiographic methods and filter papers used in the LAMs to assert average airborne particle concentrations suffered from low bias due to particles embedded in the LAM filters and the method used to count spots on the exposed X-ray films (Bradshaw and Cottrell 1954).

The practice of radiographing the material collected from the gummed-paper trays was eventually discontinued, and the fallout data from the trays was reported only in terms of activity per unit area. The gummed-paper fallout trays and the rainout collectors remained part of the complement of collection media utilized by the ORNL onsite air monitoring network through 1984.

4.3.4 Estimation of Radioactive Airborne Emissions

The initial emphasis on effluent monitoring at ORNL was on liquid waste discharges, either directly or indirectly, to White Oak Creek. Early data on gaseous effluent discharges are scarce, as airborne releases were not considered a significant contributor to on- or offsite contamination or personnel exposures. The combination of airborne concentration data from the CAM stations and the numerous exposure rate measurements indicated minimal impact from airborne releases. Therefore, the focus of stack monitoring practices for the first decades of Laboratory operations was the identification of off-normal events resulting in abnormally high stack effluent concentrations. The original stack sampling methods focused on sampling offgas streams more so than monitoring, with samples drawn through filter media for subsequent counting for gross beta and gross alpha. Counting results were divided by the volume sampled to yield average concentration over the sampling interval. Gaseous releases were quantified in this manner until the early 1960s, when the Laboratory began reporting ^{131}I releases in total curies. However, quantification of other components of the Laboratory source term did not begin until the early 1970s, when the Laboratory began reporting discharges for noble gases (in 1969), tritium (in 1972), and “unidentified alpha” (in 1972). Improvements to stack monitoring practices in 1970 allowed the Libratory to report separate release estimates for ^{85}Kr and ^{133}Xe rather than for total noble gases.

From an offsite dose perspective, the most significant airborne radioactive effluent from ORNL was ^{131}I from the period 1943 until around 1972. The importance of ^{131}I to collective offsite dose diminished after 1972 due to a decrease in production and improvements in the offgas treatment systems. As a result, tritium became roughly equal to ^{131}I in terms of contribution to collective dose in the period from 1972 to 1979 (Ohnesorge 1986). After 1979, ^{131}I fell out of the picture, leaving tritium as the principal contributor until 1997, when tritium releases dropped to a point where they were no longer significant. At this time, ^{41}Ar releases from the HFIR (7911 stack) became the principal contributor to offsite collective dose, supplanted with ^{138}Cs if the HFIR was not operating for extended periods. Note: these materials are discussed in terms of contribution to offsite dose since that was the driver for the effluent monitoring and reporting program. This is not meant to imply that different materials were important contributors to onsite dose to ORNL workers.

By the late 1950s, approximately 90% of the gaseous radioactivity generated by the Laboratory was discharged via the principal stack, referred to as 3039 (also referred to in older literature as the 900 area stack) (Manneschmidt undated). The other contributors to the site's airborne source term in this era were the 3018 and 3020 stacks, formerly known as the 105 and 205 stacks, respectively. The 3039 stack discharged gaseous wastes from the isotope processing areas, the ORNL Research Reactor, and various buildings housing research and development groups. The 3018 (reactor area) stack vented cooling air discharged from the Graphite Reactor, consisting primarily (on an activity basis) of ^{41}Ar. The 3020 (pilot plant) stack discharged ventilation air from hot cells and operating areas where processing of irradiated reactor fuel was performed.

Routine monitoring of the gaseous waste discharges at the lab began in 1956 when the Health Physics Department installed a filter tape monitor on the Graphite Reactor (3018) stack (Manneschmidt undated). A similar device was installed on the pilot plant (3020) stack in 1957, and coverage was extended to the principal stack (3039) in 1959. Manneschmidt asserts nonroutine grab sampling was performed prior to the installation of these devices. The tape monitors did not provide a measurement of activity discharged, but simply a means to detect unusually high activity (via a side-window GM tube). Manneschmidt reports that tapes were changed daily, and used tapes were counted for gross beta/gamma and gross alpha activity following 72 hours of decay. No nuclide identification was performed. These systems suffered from significant sample line losses.

A report prepared by ORNL for public hearings on radioactive waste disposal before the Joint Congressional Committee on Atomic Energy (January 28 through February 3, 1959) provides more detail on the stack monitors in use at that time. The system for the 115 (Graphite Reactor) stack is consistent with the description by Manneschmidt (Undated), and the collection media was HV 70 paper (Browder 1959). The systems for the 205 and 900 stacks are described as sample lines drawing the extracted air through Millipore filters. Unlike the system for the 105 stack, no mention is made of count rate meters or chart recorders. Browder (1959) reports the filters were changed daily and counted for gross beta and gross alpha activity. No mention is made of how long the filters were allowed to decay prior to counting or if any sort of decay correction was performed. Results obtained were compared with MPC values for gross beta and gross alpha.

Gaseous waste data reported for the second quarter of 1961 were in terms of "long-lived" gamma activity for the principal Laboratory stacks. These data do not include, for instance, the contribution from ^{41}Ar discharged from the 3018 stack (Cox 1961). However, ^{131}I is included, so it is not clear how long the filter papers were allowed to decay before counting (or if the assay results were corrected to account for decay during collection and between collection and counting). Cox (1961) asserts that 90% of the total long-lived beta/gamma activity discharged came from the 3039 stack, and that 50% of this activity came from iodine processing operations in the isotopes area. Principal components of the activity are ^{131}I, ^{133}I, and traces of ^{103}Ru. The same report states the use of paper filters had been discontinued in favor of membrane filters, with the latter giving better collection efficiency, and retention characteristics that made for higher alpha counting efficiencies.

Reliable estimates of quantities of airborne radioactive effluents discharged by ORNL are not available prior to the installation of the sampling system at the 50-foot level of the 3039 stack in the Fall of 1961 (Jordan 1974). The monitoring equipment for the 3039 and the 7911 stacks was modified in April 1969 to provide quantitative information on noble gas releases for the first time. Noble gas releases were initially reported in terms of ^{133}Xe equivalence, with this practice transitioning to reporting equivalent quantities of ^{133}Xe and ^{85}Kr shortly thereafter. Either way, ORNL felt the reported releases were quite conservative.

Quantities of tritium released to the atmosphere asserted by ORNL are upper bound estimates based on differences between material received and the sum of material sold plus that held in inventory. This method was necessary due to the lack of reliable tritium monitoring equipment at the largest source of the Laboratory's airborne tritium emissions, Building 3033. This facility received tritium primarily from the Savannah River Plant, purified it, and repackaged it for industrial and research applications. The tritium released in the Building 3033 gaseous waste stream was discharged via the 3039 stack. The 3033 building did have monitoring equipment in its offgas system, but the monitor could not differentiate between tritium and the ^{85}Kr that was also processed there. Another source of atmospheric tritium emissions at ORNL was the Tritium Target Fabrication Facility (TTFF), Building 7025. This facility operated intermittently in relation to Building 3033 and handled tritium in much smaller quantities. Tritium was released from the TTFF via its own local stack.

Summary data of annual discharges of radionuclides to the atmosphere began appearing in the annual reports of the Health Physics Division in 1976. Discharge estimates were provided for individual stacks for tritium, ^{85}Kr, ^{131}I, and ^{133}Xe (Auxier and Davis 1977).

By 1984, noble gas discharges via the 3039 stack were quantified via "a real-time (continuous) monitor with an electronic integrator" (MMES 1985). Prior to this time, discharges were estimated via manual integration of stripchart data. Tritium discharges, 99% of which were via the 3039 stack, were quantified for the first time using a real-time monitor for that release point. All tritium discharge estimates asserted prior to 1984 are based on inventories. Remaining tritium releases (from the local stack at Building 7025) were quantified via silica gel samplers. Releases of alpha and beta particulates and iodine were quantified using filters and charcoal cartridges, respectively. These media were collected three times a week from the 3039 stack sampling system and weekly for other stacks. Iodine-131 was released primarily from the 3039 and 7911 stacks from operations involving fuel elements and isotope production activities. Radon-220 was released from the 7911 stack from storage of a neutron-irradiated ^{226}Rn source at the HFIR complex. The source was sealed in 1983, but radon releases continued from contamination of the storage area (MMES 1985).

A summary of total annual airborne releases of ^{131}I, tritium, ^{85}Kr, and ^{133}Xe reported by ORNL is included as Attachment 4A to this document. All values are given in total curies discharged to the atmosphere. The data date to 1961 for ^{131}I releases, 1969 for tritium releases, and 1970 for noble gases. For completeness, data for ^{41}Ar and ^{138}Cs have been included for later years when these nuclides represented the maximum contributions to collective offsite dose.

4.3.5 Onsite Exposure Rate Measurements

By the late 1940s, ORNL had an established program of routine measurements to monitor the ambient "background" at the Laboratory. Measurements were made at approximately 50 specific locations in the Laboratory area using a portable GM counter and scaler. The measurements, referred to as "background checks," were performed approximately monthly and were originally quantified in terms of count rate. Starting in February 1949, a radium source was used to determine a calibration factor in cpm per mR hr^{-1} for the detector used, and results from then on were reported in mR hr^{-1}. However, data for 1947 and 1948 have been found in which the results have been converted from cpm to mR hr^{-1} (Morgan 1959).

The GM counter used for background measurements was mounted in a 2- by 2-inch piece of lumber that extended approximately 3 feet from the bed of a pick-up truck (Cottrell 1949). This arrangement placed the counter approximately 3 feet off the ground. The detector was coupled to a scaler and a portable power supply (if AC power wasn't available at the measurement location). Counting intervals

ranged from 2 to 5 minutes. Note: The exposure rates at some measurement locations were such that the GM tube could not be used. In these instances, an electroscope was used.

It is suggested in some of the periodic reports of the ORNL Health Physics Division that a scintillation detector was sometimes used rather than a GM counter for the background measurements, but the protocol appears to have remained otherwise unchanged until 1962. Experimental, weather-proofed film packets were tested for continuous environmental monitoring during 1961 and found to be satisfactory. Morgan and Hart (1962) report the results from the film packets differed from survey meter results by less than 10%, and thus the film packets became the principal means for monitoring background exposure rates beginning with 1962. The packets were placed near the original stations where monthly background readings had been taken. The packets were processed quarterly. It appears that onsite ambient exposure rate measurements were performed using film packets through 1970. Annual average exposure rate data do not appear in the Health Physics Division reports for 1971 through 1975, and have not been found in other references reviewed to date.

By 1973, the practice used by ORNL for measuring exposure rates at offsite locations in East Tennessee changed from static measurements using GM or scintillation detectors to the deployment of thermoluminescent dosimeters (TLDs). The TLDs were placed at the PAM and RAM stations at a height of 4 feet off the ground (Auxier, Davis, and Turner 1974). The thermoluminescent material was calcium fluoride (Harshaw TLD-200). Control badges were irradiated to 20 mR at the midpoint of a deployment period to provide a fade correction (Auxier, Davis, and Turner 1975). The TLD measurement protocol for the PAM and RAM stations was modified slightly in subsequent years, with the dosimeters suspended at a height of 1 meter off the ground and the exchange frequency for the dosimeters at the RAM locations extended to semiannually (Auxier, Davis, and Turner 1976). The exchange frequency for the dosimeters at the PAM stations remained monthly.

TLDs were included at the onsite LAM stations as of 1976. Commensurately, annual average exposure rates on the ORNL site were again reported in the summary reports of the Health Physics Division. By 1982, the dosimeters used for environmental exposure measurements were changed to a combination of lithium fluoride and calcium fluoride units, suspended 1 meter off the ground. The exchange frequency for the dosimeters remained the same: monthly for PAM stations and semiannually for LAM and RAM stations (ORNL 1983). The practice of reporting onsite average exposure rates for ORNL appears to have ceased after 1984, presumably an artifact of the major reorganization of the air monitoring networks in operation throughout the ORR that took place during 1985.

4.4 INTERPRETATION AND ASSESSMENT OF AVAILABLE MONITORING DATA

4.4.1 Airborne Concentration Measurements

Until 1966, measurements of airborne radioactivity concentrations for local onsite air at ORNL were performed by sampling with particulate filters alone. Thus, while results from the Laboratory's onsite air monitoring network should be reliable for mixed fission product activity, the results were not deemed reliable for estimating ^{131}I concentrations for dose reconstructions under the EEOICPA. Iodine-131 was a significant constituent of ORNL airborne releases from early in site history until the early 1970s; however, charcoal cartridges were not added to the sampling train of the local air monitoring network until 1966. Similarly, Laboratory onsite air monitoring results in the periodic reports of the Health Physics Division do not address tritium concentrations. Tritium became a relatively significant constituent of airborne releases once ^{131}I releases dropped off following a reduction in isotope sales. The largest airborne tritium releases from ORNL occurred between 1980 and the early 1990s.

It appears the protocol for determining the long-lived airborne beta concentration from the CAM filters (from all three monitoring networks) remained fairly consistent over the Laboratory's history. As described in ORNL-2601 (Browder 1959), CAM filters were changed weekly unless significant contamination occurred. For the LAMs, which included GM detectors and count rate meters, count rate data were tabulated hourly, with air concentrations indicated by the rate of increase of the count rate (under the assumption that the half-life of the activity collected was long in relation to collection time). Following collection, the filters were analyzed first by autoradiography, then by gross beta counting. Autoradiography was done by placing 12 filters at a time on a 14- by 17-inch sheet of cardboard and placing the cardboard in direct contact with a sheet of Kodak Blue X-ray film (Browder 1959). This filter/film arrangement was exposed for 24 hours in a dark room. Afterward, spots (i.e., particles) were counted via the unaided eye, with particle activities assigned by comparing the diameter of the spots to those created using particles of known activity.

Following radiography, the CAM filters were counted for beta/gamma activity by placing them inside a 1-inch-diameter aluminum cylinder containing an axially mounted GM tube (Browder 1959). This geometry essentially replicated that of the LAMs in the field. The activity (in microcuries) was determined and divided by the volume sampled to get average concentration. The ORNL-2601 report (Browder 1959) mentions performing gamma spectrometry and chemical separations in instances where high activity was encountered to try to determine its source. However, it is not clear if this was done for the CAM filters or just material collected on the gummed-paper fallout trays deployed at the CAM stations in 1956.

In the process described by Gupton (1973), air samples were collected using 6- by 6-inch HV-70 filters at a flow rate of approximately 5 cfm for 7 days. After 7 days, the filters were collected and replaced. After approximately a week of decay, the filters were counted using a beta scintillation detector with an efficiency of 0.5 for ^{90}Sr/Y. If the apparent concentration was low, the result based on the ^{90}Sr/Y efficiency was retained, and all activity was considered "long-lived." However, if the result was elevated, the isotopes were identified and an appropriate efficiency applied. The MDC of this technique for respirable activity based on the ^{90}Sr/Y efficiency was "about 5×10^{-15} µCi/ml" (Gupton 1973). Review of the periodic waste monitoring reports from the late 1940s suggests filters were collected on a shift basis (every 8 hours), and thus the MDC would increase commensurately. Cottrell (1948a) reports the background of a CAM after an hour of operation was about 300 to 800 cpm. How long the filters were allowed to decay prior to counting and the specifics of how they were counted during this era are not known. In addition, by the late 1940s, the filters were autoradiographed to determine the number of discernible particles collected. Alpha counting was not performed, though special sampling for alpha air activity could be conducted on an as-needed basis using grab samplers.

Note: The gross beta results reported for the ORNL CAM stations have always been in terms of "long-lived" activity, meaning the filters were decayed for long enough that short-lived particulate decay products of noble gases and radon were not included in the results.

The average concentration reported for the LAM network for 1949 through 1951 is notably higher than for all subsequent years, with the exception of 1954, when the average is driven by the RaLa accident that occurred that April (see the summary of gross beta results from the LAM network in Attachment 4B). Thus, the expansion of the LAM network from four units to ten that occurred around late 1949 does not appear to have had a significant impact on the site-wide average. This implies the network was providing representative coverage. The reason for the nearly order of magnitude drop in reported concentrations between 1951 and 1952 is not known, but it might have been the result of a change in analytical methods or assumptions. The electronics used in the LAMs were switched during this time from scalers to ratemeters, but this is not believed to have affected reported average concentrations.

Referring to emissions from the 900 (3039) stack during RaLa runs 52 and 54, Bradshaw and Cottrell (1954) report "No correlation was found between the stack activity and the response of the CAMS on the Laboratory area in either run." They go on to add that good correlation was observed between airborne emissions from the vent of the W-9 underground waste storage tank and the response of the CAMs. Thus, ground-level radioactive material concentrations in air on the ORNL site were primarily due to ground-level and low-elevation release points, and not stack emissions. That was especially true until about 1970, when charcoal filtration was added to the offgas line of the liquid waste storage tank used for waste from operations involving ^{131}I.

Though it is doubtful the LAMs ever provided an accurate representation of onsite ground-level ^{131}I concentrations prior to the addition of charcoal cartridge sampling media in 1966, the filter paper media did collect some radioiodine activity. Presumably the filter media would collect iodine-bearing aerosols with particle sizes in the respirable range, and airborne radionuclide concentrations were treated as ^{131}I equivalent (i.e., were compared to the MPC for ^{131}I) in the early years of Laboratory operations. Decay curves produced using CAM filters in service during a period of increased air concentration resulting from a temperature inversion during RaLa Run No. 21 showed a mixture of ^{131}I, ^{133}I, and ^{135}I (Burnett 1947a). A description of an investigation of elevated airborne activity in July 1947 includes the assumptions used in computing airborne concentration from the count rate from the CAM filter. These assumptions include a collection efficiency of 100% for the activity, which from decay curves appeared to be ^{131}I, and was assumed to be ^{131}I for comparison against tolerance levels (Burnett 1947b). There are several instances in the late 1940s where the CAMs "blocked out," meaning they went off-scale for a period. These instances typically occurred during periods of RaLa dissolvings, when radioiodines would have been a principal constituent of the activity. However, instances of off-scale activity would occur during RaLa runs after dissolving operations were completed, suggesting ground-level particulate emissions from waste jetting and sparging operations could peg the monitors, though radioiodines might have been the significant contributor in these instances as well. Other isotopes associated with short-duration instances of increased air activity include ^{140}Ba (in January 1948) (Burnett 1948). In some cases, upsurges in air activity associated with temperature inversions could not be associated with a specific Laboratory process, and the activity decayed rapidly. Cases of increased air concentrations accompanying temperature inversions that were primarily ^{41}Ar activity are described in (Morgan 1947). Several short-duration instances of elevated air activity are mentioned in the waste monitoring group reports from the late 1940s, albeit in a nonquantitative manner. The reports note instances of increased air activity if the measured value exceeded 10% of the ^{131}I tolerance level for that era of 8.5×10^{-8} µCi cc^{-1}. Note: Reliability of the CAMs during this period was an issue, during 1947 in particular, with data loss rates as high as 56% noted. Loss rates appear to have been significantly reduced by mid-1948.

The airborne radioparticulate concentration results determined from the three CAM networks were primarily driven by fallout from above-ground nuclear weapons testing between 1960 and 1965. Fallout affected the concentration data for other periods as well, but between 1960 and 1965 contributions from fallout to airborne gross beta concentrations reported by the ORNL LAM network dominated those from site operations. Fallout from a number of Soviet tests beginning in September 1961 caused average concentration levels as determined by the LAM and RAM stations to increase by approximately 2 orders of magnitude for the third quarter of 1961 (Hart 1961b). Fourth quarter 1961 data were similarly affected, although data for September represented the peak for CAM results. (The results from gummed-paper fallout trays did not peak until November of that year.) LAM and RAM data for September through December 1961 emphasize the magnitude of the effect of fallout on airborne concentration measurements, as the average concentration from the 8 RAM stations exceeded that of the 10 LAM stations. Thus, the LAMs were effectively blinded to airborne radioactivity originating from ORNL during that time. The same arguments hold true for particulate data from autoradiographs of the CAM filters and from material collected on the gummed-paper fallout

trays. Gamma spectrometry performed on fallout material from Soviet tests collected in September 1961 showed a combination of ^{237}Np and mixed fission products, with the latter consisting primarily of ^{140}Ba/La, ^{132}Te/I, ^{99}Mo, ^{95}Zr/Nb, and ^{131}I (Hart 1961b).

Figure 4-1 is a plot of annual average gross beta concentrations for the LAM and RAM networks for 1956 through 1983. The effect of fallout on LAM data between 1960 and 1965 is apparent.

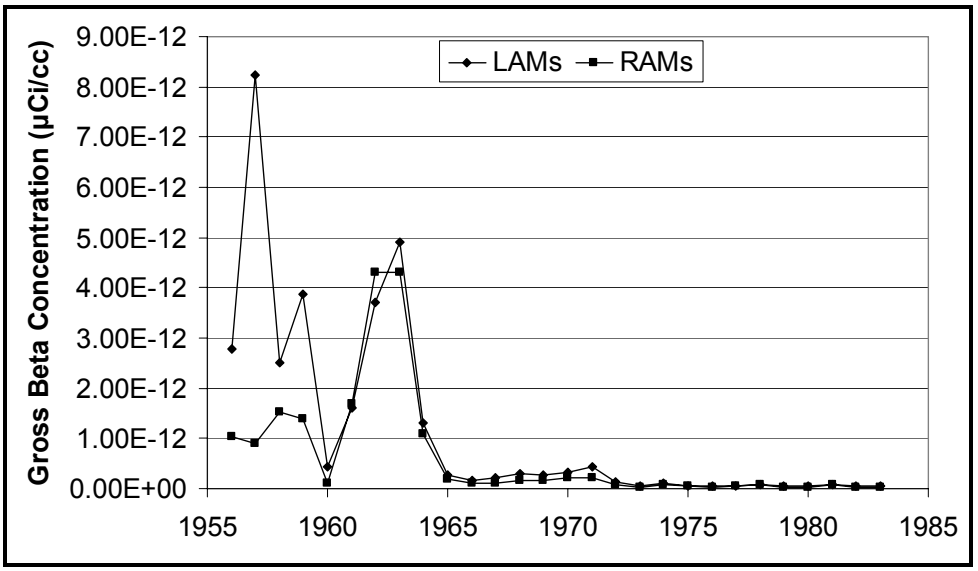

Figure 4-1. Annual average gross beta concentration data from LAM and RAM networks for 1956 through 1983.

The fact that gross beta concentration results for the ORNL LAM network consist of little more than fallout and ^{7}Be from 1960 on makes the matter of assigning a nuclide mix for dose assessment somewhat arbitrary. The true net inhalation dose an individual might have received from material originating from ORNL would be only a small fraction of that assigned using the gross beta concentration. Further, the dose associated with gross measurements would be minimal regardless of the nuclides assumed, given the low concentrations. The same is true for the early years of Laboratory history; however, for this period most of the activity collected on the filters originated from Laboratory operations. Much is known about nuclides that would have been present in the gross beta activity collected during this period; thus, a representative, albeit conservative, nuclide identification can be assigned to gross beta results prior to 1960 for dose assessment.

The principal contributors to airborne gross beta activity collected by the LAM stations in the early period of Laboratory history would have been emissions associated with the RaLa process. RaLa operations were carried out at ORNL from 1944 to 1956. On an activity basis, the most abundant radionuclides present in RaLa emissions (excluding noble gases and their decay products) would have been isotopes of iodine, ruthenium, cerium, barium, lanthanum, zirconium, and niobium. Iodine and ruthenium were the more volatile of these, and thus made up the majority of the activity collected in periods of RaLa operations (RaLa was a batch process carried out several times a year). While the particulate filters in the LAMs would have collected some of the airborne radioiodines present, the highly variable and uncertain nature of the retention efficiency precludes making a defendable estimation of how much. Thus, while the gross beta results included some contribution from radioiodines (primarily ^{131}I), the uncertainty associated with this contribution is such that it makes subtracting it impractical. Subtracting the ^{131}I contribution to gross beta results would be desirable because ^{131}I concentrations are treated separately in this document. However, including them with

gross beta results is conservative and, in reality, of little consequence given the small magnitude of the concentrations.

Therefore, in light of the above, if it is desirable to assign a dose contribution from the gross beta (mixed fission product) concentration data provided in Attachment 4B, it is recommended the activity be treated as either ^{106}Ru or ^{144}Ce, depending on the organ of interest. Both should be assumed to be absorption type S. Along with ^{131}I, the principal constituent of the particulate activity collected on LAM filters in the RaLa years was probably ruthenium (in a mixture of ^{103}Ru and ^{106}Ru); and Class S ^{106}Ru is the bounding radionuclide of those that might have originated from ORNL. However, there may be cases (organs) where ^{106}Ru is not bounding relative to ^{144}Ce, in which case the latter should be selected. While it is true that isotopic analyses performed on LAM filters in later years did sometimes show the presence of ^{90}Sr, it is not thought ^{90}Sr would ever have been a significant component of the gross beta activity that actually originated from ORNL. Treating gross beta results as ^{106}Ru or ^{144}Ce should bound any real contributions to a worker's dose from ORNL emissions after 1960 since very little of the total activity actually originated at the Laboratory. With all that said, however, dose reconstructors might find it easier to simply ignore gross beta (mixed fission product) intakes altogether since they likely will be only a minute fraction of the dose from other contributors (i.e., ^{131}I or tritium). This is especially true given the conservative nature of the concentration data provided for ^{131}I and tritium in Attachment 4B.

The gross beta (mixed fission product) concentration data in Attachment 4B are site-wide averages reported by the Laboratory. Specifically, they are the average of results from the stations in operation around the main ORNL campus plus, in later years, those for the HFIR area and the Low-Level Counting Laboratory about 2.5 miles east of the main complex. The results do not include contributions from outlying areas that could have brought down the average. As with the onsite exposure rate data described elsewhere in this document, use of a site-wide average is appropriate, given it would seem to be the likely outdoor exposure profile for an individual working at or visiting the ORNL site. However, as discussed above, contributions to a claimant's dose from inhalation of mixed fission products in the outdoor air should be negligible in relation to that from ^{131}I or tritium. This is especially true given that results after 1960 include very little contribution from materials originating from ORNL.

4.4.2 Radioactive Airborne Emissions

A study was performed in the early 1960s to assess the stack sampling system for the 3018 stack vis-à-vis sample line losses (Youngblood and Fish 1961). At this time, the 3018 stack served both the ORNL Graphite Reactor and the Low-Intensity Test Reactor (LITR). The investigators determined that a quantitative estimate of the sampling error for particulates could not be made due to the lack of information on particle size distributions and the proximity of the sampling nozzle to flow disturbances. They estimated the loss of iodine vapor in the system to be at least 30%, but could not perform a more precise estimation due to turbulent flow conditions. They did assert that "sample losses are expected to be considerable in the present system" (Youngblood and Fish 1961).

Youngblood and Fish (1961) did not directly evaluate the sampling system for the principal ORNL stack (3039), but did mention the fact that the filter for the 3039 monitoring system only showed a trace of ^{106}Ru following the November 1959 fallout incident. In contrast, the sample line upstream of the filter is reported to have contained enough deposited material to give a direct reading of 10 mrad hr^{-1}. The authors cite this as evidence that the sampling system for the 3039 stack was insensitive to the release of larger particles from agglomeration on interior surfaces of the fan and duct systems (Youngblood and Fish 1961).

ORNL began reporting noble gas releases to the AEC in 1970 in terms of annual totals (in curies) of ^{85}Kr and ^{133}Xe. The quantities (activities) reported were determined via a conservatively selected ratio method for demonstrating compliance with contemporary MPC values. ORNL felt that these totals represented a gross upper bound and that actual releases were probably an order of magnitude lower (McVey 1972). ORNL used the conservative ratio method to assert noble gas releases because it did not have the capability to monitor the individual streams feeding to the central offgas system and the nature of the various processes that generated noble gas effluents was such that the nuclide mix varied widely (McVey 1972). Thus, noble gas releases asserted by ORNL appear to be conservative by a significant margin.

ORNL did not begin reporting estimates of airborne tritium releases until 1972, and did not begin monitoring for tritium releases until 1984. The release estimates for 1972 to 1983 were based on throughput (i.e., the amount of tritium handled). The most significant airborne tritium releases in ORNL history occurred from approximately 1980 through 1991 (see Attachment 4A).

ORNL began reporting annual releases of ^{131}I in 1961 and continues to do so. Although the problems and issues associated with sampling for radioiodines (i.e., accounting for the large differences in behavior and reactivity among the different chemical species present in an effluent stream) were beginning to be understood around that time, it was nonetheless felt the releases reported for that era should be adjusted upward to account for potential losses in the sampling system and to ensure favorability to the claimant.

Organic iodides are carried through long sample lines with little loss. However, reactive iodine (I_2) can suffer from significant line losses, though some of the I_2 that deposits on the walls is later resuspended. Because the true mix of iodine species that were present in the 3039 stack exhaust cannot be known, a factor of 2 was applied to the ^{131}I releases asserted by ORNL for 1961 through 1965. The asserted release data were not needed to compute onsite airborne concentrations for 1966 through 1984 because representative monitoring data were available. A correction was not applied to the asserted releases after 1984 (when monitoring data are no longer available) because the releases reported at that time should meet modern standards, though they are minimal in terms of dose consequence.

Although it was chosen somewhat heuristically, the factor of 2 applied to the asserted ^{131}I release data for 1961 through 1965 should be reasonably accurate in light of what is known about sample line loss mechanisms for reactive iodine and the fact that releases in this period were small in terms of dose consequence.

4.4.3 Onsite Exposure Rate Measurements

The instruments and gamma dosimeters used to perform exposure rate measurements on the ORNL site over its history were originally calibrated to ^{226}Ra. Between 1970 and 1976, the method used to measure onsite exposure rates at ORNL switched from using film badges to using TLDs. TLDs were in use at the PAM stations to measure environmental exposure rates as early as 1973. At some point the photon source used to calibrate the dosimeters changed from ^{226}Ra to ^{137}Cs; however, for this document all exposure rate measurements have been treated as equivalent regardless of the calibration spectrum used or the reporting units (some measurements are reported in rad rather than R).

Given the number of measurements taken, the fact that they reflect known off-normal releases of radioactive material at the ORNL site and compare favorably with continuous monitoring practices once these were initiated, it has been assumed that periodic exposure rate measurements performed

using portable instrumentation adequately represented ambient conditions at the times they were performed.

All site-wide annual average exposure rate values provided in this document include natural background and contributions from non-ORNL-related activities, such as fallout. Fallout from atmospheric nuclear weapons testing was observed to have a minor effect on local exposure rate measurements on the ORNL site. Significant fallout (such as that from Soviet weapons tests beginning in September 1961) was observed to drive local ambient exposure rates higher, but only slightly so in relation to local sources. Hence, while fallout did have a positive bias on local exposure rate measurements, the magnitude of this bias does not ever appear to have been larger than around 10% or so. For comparison, fallout from 1961 Soviet weapons tests raised background levels at remote locations by around 30% (Hart 1962).

Annual average site-wide exposure rate data are not available for 1944 through 1946. The annual site-wide average determined for 1947 was therefore assigned for 1944 to 1946 as well. The largest contributor to elevated local exposure rates during that time was the RaLa process, from exposure rates around the process building itself while runs were in progress and from high-level liquid wastes eventually discharged to the settling basin. Data from the X-22 chambers from 1944 and 1945 show noble gas emissions to be only a minor contributor to onsite exposure rates. The number of short-decayed fuel slugs from the Graphite Reactor used in the RaLa program in 1944 and 1945 was much less than that for 1946, and the amount of material processed in 1946 was comparable to that processed in 1947 [see ChemRisk (1999) for details]. Thus, applying the 1947 exposure rate for the prior years should be claimant-favorable.

No onsite exposure rate data have been located to date for the LAM stations for 1971 through 1975. Annual average site-wide exposure rate data began being reported again in 1976, by which time dosimeters deployed at the LAM locations had changed from film badges to TLDs. To fill the period of missing data, the annual site-wide average exposure rate measured for 1970 was assumed for 1971 through 1975. The result for 1970 was 0.072 mR hr^{-1}. In 1976, when the annual average site-wide exposure rate was again reported, the value was 0.056 mR hr^{-1}.

No annual average exposure rate data for the ORNL site have been located for any years after 1984. The 1984 average was therefore assumed for 1985 through 2003. The same value was applied for 1982 because a quantitative measure of the average exposure rate was lacking for that year as well. The annual average exposure rate for the ORNL site did not show appreciable change from 1979 through 1984.

4.5 DERIVATION OF ONSITE AIRBORNE CONCENTRATIONS FOR RADIONUCLIDES OF CONCERN

The three principal contributors to inhalation dose individuals could have been exposed to in the outdoor air on the ORNL site were ^{131}I, particulate mixed fission products (MFPs), and tritium. Lack of available monitoring data, for ^{131}I in particular, required that bounding values of dispersion coefficients coupled with available source term estimations be used to establish the needed onsite airborne concentration and intake data for certain periods. The dispersion coefficients used for these calculations are discussed in the next subsection, followed by discussion of the derivation of the airborne concentration data for outdoor air at ORNL for the three principal contributors to onsite dose. All concentration data are given in terms of annual averages.

Attachment 4B to this document provides a summary of the annual average airborne concentration data and the associated intakes for an individual assumed to be exposed to the average concentration for 2,000 hours breathing at a rate of 1.2 cubic meters per hour.

4.5.1 Dispersion Coefficients for Principal Elevated Release Points

The lack of reliable measurements of airborne tritium and ^{131}I concentrations at ORNL meant an alternative method had to be used to derive estimates of these values. Available source term information (if available) or release estimates, coupled with dispersion coefficients were therefore used. However, given the small size of the ORNL site, bounding values for dispersion coefficients were used for elevated release points. A bounding value was used for low-elevation releases such as fugitive emissions directly from buildings and short, local stacks, as appropriate.

Dispersion data asserted for the more important ORNL stacks are summarized in Table 4-2. These values were used as needed to provide estimates of local onsite airborne concentrations of tritium and ^{131}I for periods when reliable concentration measurements were not available.

Table 4-2. Dispersion coefficients used for ORNL airborne release points.

Stack	Factor	Dimensions	Reference	Remarks
3039 (formerly 900)	2.226×10^{-7}	sec m^{-3}	(Binford, Hamrick, and Cope 1970)	Maximum, annual-average χ/Q value; occurs 2,000 meters northeast of stack.
3020 (formerly 205)	2.615×10^{-7}	sec m^{-3}	(Binford, Hamrick, and Cope 1970)	Maximum, annual-average χ/Q value; occurs 2,000 meters northeast of stack.
Roof level (50 feet)	2.1×10^{-4}	sec m^{-3}	(Browning 1962)	Assumes worst-case meteorological conditions

In a report prepared by ORNL for public hearings on radioactive waste disposal held before the Joint Congressional Committee on Atomic Energy in early 1959, reference is made to an unpublished study performed by the U. S. Weather Bureau by R. E. Meyers and D. R. Purdy titled “Calculations for Unit Emissions of Air-borne Contaminants.” According to Browder (1959), this study included calculations of maximum ground concentrations per unit discharge rate for varying weather conditions for the ORNL stacks, and asserted a probable minimum dilution factor of 10^{-3} for each of the three stacks. This dilution factor was the basis for the ORNL stack monitoring program in the early 1960s, or more specifically, was the basis for calculations of maximum allowable discharge rates. A determination of the maximum allowable release of ^{131}I via rooftop hood exhausts from 1962 included calculation of a worst-case dispersion coefficient for releases 50 feet above the ground. The dispersion coefficient computed for a roof-level (50-foot) release under a night-time temperature inversion and 5-mph winds was 2.1×10^{-4} sec m^{-3} (Browning 1962). This value was used to estimate ground-level concentrations resulting from local, low-elevation releases on the ORNL site.

4.5.2 Onsite Airborne Concentrations of ^{131}I

The largest airborne releases of ^{131}I were associated with RaLa operations, which took place at ORNL between 1944 and 1956. Processing of short-decay irradiated fuel in the Thorex process in 1956 and 1957 was the next largest source of airborne ^{131}I emissions, followed by the production of ^{131}I for distribution under the ORNL isotopes program. ORNL did not start reporting airborne releases of ^{131}I until 1961, by which time releases had diminished considerably. Similarly, the Laboratory did not start monitoring onsite airborne concentrations of ^{131}I until 1966, when it added charcoal cartridges to the sampling chain of the LAM stations. The Laboratory maintained an air monitoring program for airborne particulate radioactivity from early in its history, but the data cannot be used to establish

reliable estimates of local airborne ^{131}I concentrations. Particulate filters are in general a poor collector for radioiodines. They will collect iodine that has sorbed onto particles, plus a small fraction of the reactive I_2 species, but the retention efficiency for the reactive species is highly variable and a function of the incident concentration and the relative humidity, among other things. Thus, given the fact that neither the retention efficiency of the particulate filters used in the LAMs nor the overall mix of the iodine species in the local air at ORNL can be known, no defendable assertions of airborne radioiodine concentrations can be drawn from particulate monitoring results. Thus, for 1944 through 1965, airborne ^{131}I concentrations on the ORNL site had to be established using information on emission rates and bounding values of dispersion coefficients.

For 1944 through 1956, the principal source of airborne ^{131}I emissions at ORNL was the RaLa program. ^{131}I releases associated with RaLa processing at ORNL were studied in detail as an element of the Oak Ridge Dose Reconstruction and are documented in ChemRisk (1999). Iodine-131 releases from the RaLa program are documented in Chapter 3 of this reference in terms of annual total emissions. Separate source terms are provided for the stacks used to ventilate dissolver offgases and for the process building itself. This source term information was used to derive local onsite concentrations of ^{131}I needed for this document by converting annual releases to release rate and then multiplying by bounding dispersion coefficients for either the stack or roof-level emissions, as appropriate. The dispersion coefficients used were as follows:

- 2.62×10^{-7} seconds per cubic meter for the 3020 stack, which ventilated dissolver offgases from 1944 until 1950
- 2.23×10^{-7} seconds per cubic meter for the 3039 stack, which ventilated dissolver offgases from 1950 until 1956
- 2.1×10^{-4} seconds per cubic meter for direct (fugitive) emissions

All of the dispersion coefficients used are bounding values, as described in Section 4.5.1. The results of the calculations of bounding, local ^{131}I concentrations at ORNL determined from the RaLa source term are summarized in Table 4-3. Note these values include releases associated with the April 1954 accident, as documented in Section 3.15 of ChemRisk (1999).

Table 4-3. Bounding values of onsite airborne ^{131}I concentrations at ORNL from RaLa processing.

Year	Total Released from Stack (Ci)	Total Released from Building (Ci)	Annual Average Concentration from Stack (µCi/cc)	Annual Average Concentration from Building (µCi/cc)	Total Annual Average Concentration (µCi/cc)
1944	262.1	0.5	2.2E-12	3.5E-12	5.7E-12
1945	1371.4	62.4	1.1E-11	4.2E-10	4.3E-10
1946	830.1	16.1	6.9E-12	1.1E-10	1.1E-10
1947	1090.1	19.1	9.1E-12	1.3E-10	1.4E-10
1948	640.9	16.1	5.3E-12	1.1E-10	1.1E-10
1949	953.1	12.1	7.9E-12	8.0E-11	8.8E-11
1950	500.1	5.3	3.5E-12	3.6E-11	3.9E-11
1951	571.0	2.6	4.0E-12	1.7E-11	2.1E-11
1952	2010.3	17.1	1.4E-11	1.1E-10	1.3E-10
1953	3290.4	40.2	2.3E-11	2.7E-10	2.9E-10
1954*	3546.8	305.0	2.5E-11	2.0E-09	2.1E-09
1955	2330.2	28.2	1.6E-11	1.9E-10	2.0E-10
1956	3370.3	26.1	2.4E-11	1.7E-10	2.0E-10

*includes totals from April, 1954 accident.

After RaLa processing at ORNL was terminated in October 1956, the principal source of ^{131}I emissions from the site appears to have been the short-decay Thorex runs that took place in the 3019 Building between July 1956 and November 1957. The total quantity of ^{131}I available for release from these runs was estimated by ChemRisk (1993a) to be 906 Ci for 1956 and 14,600 Ci in 1957. These estimates were used to determine bounding, ground-level concentrations at the ORNL site using essentially the same approach as that used for RaLa releases. The total ^{131}I inventory for each year was multiplied by a factor of 0.027 to provide a total released. The factor of 0.027 is the ratio of ^{131}I releases across all RaLa runs to the available inventory. The assumption that releases from the Thorex and RaLa processes were comparable is based on the commonalities between the process equipment used for each, despite the fact Thorex was conducted in the Pilot Plant (Building 3019). The assumption of similar release characteristics for ^{131}I between the two processes is supported by interviews with individuals responsible for the design of the offgas treatment systems for the Thorex plant conducted as an element of the Oak Ridge Dose Reconstruction Project. (For example, see the interview with O. Yarbro documented as Accession Number 2562 in the Oak Ridge Dose Reconstruction Database, which is an element of the DOE Comprehensive Epidemiologic Data Resource (CEDR) hosted on the Internet by Lawrence Berkeley National Laboratory.) The total releases computed using the factor of 0.027 were partitioned between the fraction released directly from the 3019 Building and that released via the 3020 stack by applying a ratio of 0.01 (i.e., 1% of the total was assumed to be released directly, and the remainder via the stack). The 1% value was selected based on the RaLa source term, as documented in (ChemRisk 1999). The resulting total releases for the two pathways were then converted to ground-level concentrations in the same manner as that used for the RaLa releases. Calculations of the bounding ground-level ^{131}I concentrations associated with the short-decay Thorex runs of 1956 and 1957 are summarized in Table 4-4.

Table 4-4. Bounding values of onsite airborne ^{131}I concentrations at ORNL from short-decay Thorex processing.

Year	Total Released from Stack (Ci)	Total Released from Building (Ci)	Annual Average Concentration from Stack (µCi/cc)	Annual Average Concentration from Building (µCi/cc)	Total Annual Average Concentration (µCi/cc)
1956	24.2	0.2	2.0E-13	1.6E-12	1.8E-12
1957	390.3	3.9	3.2E-12	2.6E-11	2.9E-11

Neither local concentration measurements nor source term estimates for ^{131}I are available for 1958 to 1960. The only data located to date for this period are isotopic sales summaries for 1958, in which the Laboratory reported ^{131}I production in the amount of 4,040 Ci and sales of 1,074 Ci (ChemRisk 1993b). The difference between the quantity produced and that sold is attributed to radioactive decay. No isotopic sales data have been located for 1959 and 1960, although the program was still in operation at that time. Thus, assumptions were made based on the growth in ^{131}I sales. From sales data presented in Table 2-6 of ChemRisk (1993b), it appears that ^{131}I sales experienced fairly consistent growth between 1955 and 1958. In fact, the growth between 1956 and 1957 and between 1957 and 1958 are identical at 21%; thus, this factor was used to project ^{131}I production for 1959 and 1960. Doing so, one gets projected ^{131}I production values of 4,889 Ci and 5,915 Ci for 1959 and 1960, respectively.

The ^{131}I production values for 1958 to 1960 were converted to annual releases by applying the factor of 0.027 determined for RaLa releases, knowing this should be conservative for Laboratory facilities used for ^{131}I production. Doing so yields average release estimates of 109 Ci, 132 Ci, and 160 Ci for 1958, 1959, and 1960, respectively. These values compare favorably with ^{131}I releases reported by

ORNL for subsequent years. Annual average ground-level concentrations were computed by applying the bounding dispersion coefficient for the 3039 stack (2.23×10^{-7} sec m^{-3}).

As discussed in Section 4.4.2, a factor of 2 was applied to ^{131}I release data reported by ORNL for 1961 through 1965 to account for sample line loss. The doubled release estimates were used to compute ground-level concentration by applying the bounding dispersion coefficient for the 3039 stack.

The average values reported by the Laboratory from charcoal cartridge measurements from the LAM stations were used for 1966 through 1984, when these data ceased being reported. The annual average concentration for 1984 was assigned to 1985 through 2003 for completeness.

The ^{131}I concentration data described above are summarized in Attachment 4B. It has been assumed that ^{131}I emissions from isotope production activities from 1944 through 1957 are negligible in relation to bounding assessments performed using source term information for the RaLa and Thorex programs. For dose assessment, it is recommended the airborne ^{131}I be considered to be in a soluble or reactive (vapor) form.

4.5.3 Onsite Airborne Concentrations of Mixed Fission Products

The concentration and intake data for mixed fission products in Attachment 4B are reported annual averages for the ORNL LAM network. These data are gross beta measurements and include contributions from fallout and other sources, such as the ^{7}Be naturally present in the atmosphere. These contributions are the dominant constituent of the gross beta activity measured after 1960. The concentration values are such that the dose contribution from airborne mixed fission products should at all times be negligible in relation to that from ^{131}I or tritium. However, if it is desired to include the dose contribution from mixed fission products, it is recommended the activity be assumed to be Type S ^{106}Ru or ^{144}Ce, depending on which yields a higher dose for the organ of interest. Gross beta (mixed fission product) concentration results are described in more detail in Section 4.4.1. Note: The average concentration value for 1949 was assumed for 1944 through 1948.

4.5.4 Onsite Airborne Concentrations of Tritium

There have been two principal sources of airborne tritium emissions from ORNL over its history: the 3039 (central offgas system) stack and the local stack for the TTFF, Building 7025. The TTFF was not operational prior to 1967; hence, all airborne tritium releases prior to that time were via the 3039 stack alone. The combination of the large dilution afforded by the 3039 stack and the small magnitude of releases during that time was such that airborne tritium emissions from ORNL prior to 1967 are negligible in relation to the dose contribution from airborne ^{131}I. Isotope sales of tritium in that period were too little to have resulted in significant onsite concentrations from airborne emissions. Ohnesorge (1986) provides the following isotopic sales information for tritium for the period prior to 1972:

- Total sales prior to 1965 were 363,000 Ci.
- Sales varied from 121,000 to 334,000 Ci from 1965 through 1971.
- Sales for 1972 were 161,000 Ci, corresponding to a total reported release of 1800 Ci.

ORNL did not begin routinely reporting airborne tritium emissions until 1972; however, a March 1971 letter summarizing liquid and gaseous releases from ORNL asserts airborne tritium releases of 4,500 Ci and 300 Ci for 1969 and 1970, respectively (Sapirie 1971).

The period of peak airborne releases of tritium from ORNL was from 1980 through 1991. Large-scale tritium operations at ORNL ceased in 1989, but releases persisted for some time thereafter from off-gassing of tritium-bearing systems. By 1988, Building 3033 was the principal source of tritium emissions from ORNL, handling roughly 1,000,000 Ci per year in the purification of tritium received from the Savannah River Plant for redistribution to industrial and research users (Shor 1988). Airborne releases of tritium from Building 3033 were via the central (3039) stack. In addition, tritium was handled in Building 7025 in the manufacture of accelerator targets and material test samples. Quantities handled in Building 7025 were roughly 30,000 Ci per year around 1988 (Shor 1988). Thus, during its period of peak emissions, the quantity of tritium handled in Building 7025 was roughly 3% of that handled in Building 3033.

To date, no monitoring data for airborne tritium concentrations on the ORNL site have been located. Concentrations were therefore established using a combination of release data and bounding dispersion coefficients, as was done to obtain ^{131}I concentrations for the period prior to 1966. Onsite airborne tritium concentrations were established in this manner for 1967 to the present. Tritium releases were negligible in terms of onsite concentrations prior to operation of the TTFF, which released tritium to the atmosphere via a short local stack rather than the 250-foot central stack that served the 3033 Building. The fact that airborne tritium emissions from the TTFF were via its own local stack makes this facility the dominant source of onsite tritium concentrations at ORNL (under the conservative assumptions employed in this assessment).

Tritium release data asserted by ORNL are available for 1969, 1970, and from 1972 through 2002 (see Attachment 4A). Because the release estimations are for the most part based on material handling and inventory evaluations, no corrections for sampling errors were deemed necessary. Tritium release estimates were therefore needed for 1967, 1968, and 1971. From the annual tritium sales data provided by Ohnesorge (1986), the largest quantity of tritium sold from 1965 through 1971 was 334,000 Ci. Ohnesorge (1986) also provides annual sales for 1972, which is the first year ORNL began routinely reporting tritium releases. The ratio of the reported release for 1972 (1,800 Ci) to annual sales (161,000 Ci) gives the factor 0.011, which is the value ORNL used to compute the release. Applying this factor to the largest quantity of tritium sold in the period of interest (334,000 Ci) gives a release of 3,674 Ci. This value was assigned as the amount released for 1967, 1968, and 1971, implying the conservative assumption that the maximum sales occurred in all three of those years.

Annual average airborne tritium concentrations for the ORNL site were computed using the annual release data described above. Concentrations were computed by assuming 97% of the total release was via the 3039 stack and the remainder was via the local 7025 stack. The dispersion coefficients used for these two release points were 2.23×10^{-7} sec m^{-3} and 2.1×10^{-4} sec m^{-3}, respectively (see Section 4.5.1). The concentration computed for 2002 was assigned for 2003.

The concentration values and associated intakes computed as described above are summarized in Attachment 4B. Note: The intake values include a factor of 1.5 applied to account for direct absorption through the skin. Dose reconstructors should assume the tritium is in the physical form of HTO (tritiated water vapor). A geometric standard deviation of 3 should be assumed for the intake values given in Attachment 4B.

4.6 ONSITE EXPOSURE RATE DATA

Annual-average exposure rate data for the ORNL site are summarized in Attachment 4C for 1944 through 2003. The methods used by ORNL to determine these results are discussed in Section 4.3.6.

Assumptions associated with these data, including those used to fill gaps where measured values were not available, are discussed in Section 4.4.3. Two important points concerning these data are:

- The exposure rates are gross values, i.e., background is included.
- The exposure rate data are site-wide averages.

The annual average exposure rate data in Attachment 4C of this report represent site-wide averages. "Site-wide" in this context means the average value reported by the Laboratory in the periodic reports of the Health Physics Division. These average values consisted only of measurements made in the core areas of the Laboratory and areas where exposure rates where known or suspected to be elevated. The values, therefore, do not suffer from low bias from having a disproportionate number of measurements at or near background. The site-wide average was selected as the measure of interest because it was felt to be the most representative of an unmonitored worker's exposure profile. ORNL is a relatively small open campus where visitors and workers tend to move around during the time they are on the site. An individual's exposure, therefore, would include a number of different sources and areas as he/she moved about. The site-wide average exposure rate data should therefore provide a defendable basis for assigning external environmental dose for unmonitored workers. Indeed, the site-wide averages include measurement locations where exposure rates were chronically much higher than those for the other measurement locations and thus served to drive the average higher. These locations include the vicinity of the 706-D fan house, which operated until about 1950, and the vicinity of the settling basin. Other locations would raise the site-wide average depending on what activities were taking place, such as locations in the vicinity of Buildings 706-A or 706-C in the mid-to-late 1940s. The fact that these instances are included in the average exposure rate data means the site-wide averages should be sufficiently bounding for an unmonitored worker. The ORNL monitoring program kept up with the footprint of the Laboratory as its facilities expanded over the years, so site-wide averages should always represent the complement of areas of elevated exposure an individual might have encountered.

A caveat associated with the use of site-wide average exposure rates is that these values might not be appropriate for cases in which a composite exposure profile for an unmonitored individual is inconsistent with a known or asserted exposure history. If a worker's dose for a particular period has to be considered to have come from a specific source or location, the dose reconstructor will have to make a judgment about the magnitude of the site-wide average in relation to the nominal exposure rate for the specific source or location. However, the site-wide average values in this document should be higher than exposure rates that were present on most of the ORNL site because they include locations where local exposure rates were significantly elevated. For example, exposure rate measurements for a location 15 feet from the southwest corner of the settling basin for 9 of the 13 months from September 1948 through September 1949 average 3.5 mR hr^{-1}, and range from 1.7 to 8.5 mR hr^{-1} (in August 1949). Similarly, eight measurements at a location 10 feet north of the 706-D (RaLa Building) fan house for the same period average 9.4 mR hr^{-1} and range from 2.8 to 29 mR hr^{-1}. In comparison, the annual averages for the Laboratory grounds for 1948 and 1949 were 0.29 and 0.46 mR hr^{-1}, respectively. These are the two highest averages recorded for the Laboratory over its history, and are based on periodic (approximately monthly) measurements performed at 45 specific locations on the Laboratory grounds, including those in the vicinity of the settling basin and the 706-D fan house. Note. The 706-D fan house was not used after 1949 [i.e., after the RaLa Building was tied into the (then new) central offgas system]. Also note the substantial drop in site-wide averages after the 706-D fan house was retired (see Attachment 4C).

Differences between site-wide average exposure rates and the exposure rate for a specific location would probably have been the largest earlier in the Laboratory's history. This is when such

differences would have been the most important as far as an unmonitored external dose a claimant might have received.

Figure 4-2 is a plot showing the trend of the annual average exposure rates for the ORNL site. Note: The periods 1944 to 1946, 1971 to 1975, 1982, and 1985 to 2003 reflect assumed rather than measured values.

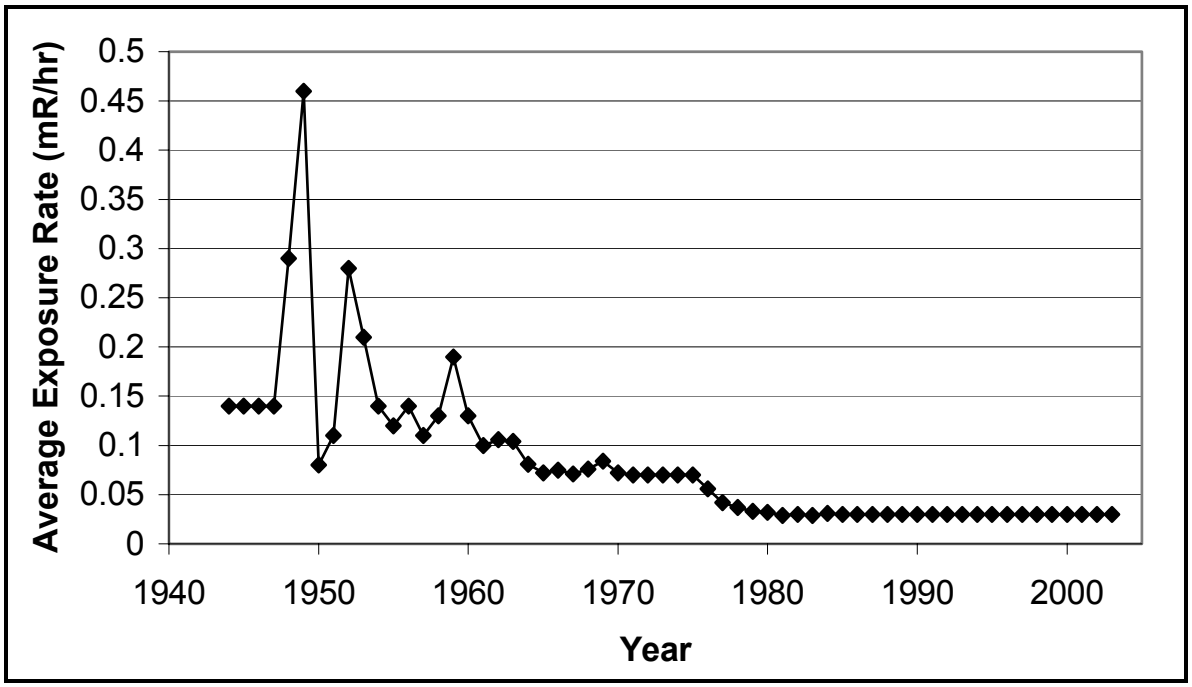

Figure 4-2. Annual average exposure rates for ORNL.

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ATTACHMENT 4A
ANNUAL AIRBORNE RELEASES REPORTED BY OAK RIDGE NATIONAL LABORATORY (CURIES)

Year	^{131}I	^{3}H	^{85}Kr	^{133}Xe	^{41}Ar	^{138}Cs
1961	42	--	--	--	--	--
1962	121.2	--	--	--	--	--
1963	54	--	--	--	--	--
1964	84.5	--	--	--	--	--
1965	18.4	--	--	--	--	--
1966	15.79	--	--	--	--	--
1967	22.3	--	--	--	--	--
1968	10.38	--	--	--	--	--
1969	16.38	4500	--	--	--	--
1970	1.43	300	15000	75000	--	--
1971	3.46	--	15000	71000	--	--
1972	1.7	1800	15400	64900	--	--
1973	2.18	9100	14000	68600	--	--
1974	1.97	555	20000	99200	--	--
1975	2.1	534	17700	87500	--	--
1976	1.25	6019	11500	54000	--	--
1977	1.37	2524	8606	42030	--	--
1978	1.7	2500	12000	59000	--	--
1979	0.3	5109	10500	51190	--	--
1980	0.22	14800	8800	42800	--	--
1981	0.5	11300	6700	32400	--	--
1982	0.13	19000	11700	57100	--	--
1983	0.05	22200	11900	57700	--	--
1984	0.1	33400	14900	72700	--	--
1985	0.09	20180	6623	32280	--	--
1986	0.035	31000	10600	51000	--	--
1987	0.02	44050	4700	22700	--	--
1988	0.056	21000	6300	31000	--	--
1989	0.03	28000	16000	79000	--	--
1990	0.02	12000	89000	1500	--	--
1991	0.046	16000	5400	3100	--	--
1992	0.14	3000	--	--	--	--
1993	0.05	410	--	--	--	--
1994	0.065	450	21	260	--	--
1995	0.16	300	--	--	--	--
1996	0.28	603	280	1.1	--	--
1997	0.055	148	110	0.0068	10000	--
1998	0.062	110	260	320	7950	--
1999	0.058	103	480	4.6	12500	--
2000	0.075	107	283	2.21	3620	--
2001	0.13	49	490	0.488	--	1360
2002	0.09	86	314	5.88	--	1590

Notes:

Missing data implies release is unknown. It does not imply there was no release.

^{41}Ar and ^{138}Cs data are only given for years when these releases represented maximum collective dose.

^{131}I release reported for 1961 is an estimation. The values for subsequent years are based on in-stack sampling.

Tritium releases prior to 1984 are estimates based on inventory.

ATTACHMENT 4B
ANNUAL AVERAGE AIRBORNE CONCENTRATION AND INTAKE DATA FOR IODINE-131, TRITIUM, AND PARTICULATE MIXED FISSION PRODUCTS AT ORNL

	Annual Average Concentration (Bq m^{-3})			Annual Intake (Bq)		
Year	^{131}I	Tritium	MFP	^{131}I	Tritium	MFP
1944	2.1E-01	--	7.4E-01	5.0E+02	--	1.8E+03
1945	1.6E+01	--	7.4E-01	3.8E+04	--	1.8E+03
1946	4.2E+00	--	7.4E-01	1.0E+04	--	1.8E+03
1947	5.0E+00	--	7.4E-01	1.2E+04	--	1.8E+03
1948	4.2E+00	--	7.4E-01	1.0E+04	--	1.8E+03
1949	3.3E+00	--	7.4E-01	7.8E+03	--	1.8E+03
1950	1.4E+00	--	6.7E-01	3.5E+03	--	1.6E+03
1951	7.9E-01	--	6.3E-01	1.9E+03	--	1.5E+03
1952	4.7E+00	--	1.1E-01	1.1E+04	--	2.5E+02
1953	1.1E+01	--	1.7E-01	2.6E+04	--	4.1E+02
1954	7.6E+01	--	1.8E+00	1.8E+05	--	4.2E+03
1955	7.6E+00	--	1.8E-01	1.8E+04	--	4.4E+02
1956	7.4E+00	--	1.0E-01	1.8E+04	--	2.5E+02
1957	1.1E+00	--	3.0E-01	2.6E+03	--	7.3E+02
1958	2.9E-02	--	9.3E-02	6.8E+01	--	2.2E+02
1959	3.5E-02	--	1.4E-01	8.3E+01	--	3.4E+02
1960	4.2E-02	--	1.7E-02	1.0E+02	--	4.0E+01
1961	2.2E-02	--	5.9E-02	5.3E+01	--	1.4E+02
1962	6.3E-02	--	1.4E-01	1.5E+02	--	3.3E+02
1963	2.8E-02	--	1.8E-01	6.8E+01	--	4.4E+02
1964	4.4E-02	--	4.8E-02	1.1E+02	--	1.2E+02
1965	9.6E-03	--	1.0E-02	2.3E+01	--	2.5E+01
1966	8.5E-03	--	6.3E-03	2.0E+01	--	1.5E+01
1967	1.6E-03	2.8E+01	8.1E-03	3.8E+00	1.0E+05	2.0E+01
1968	4.8E-03	2.8E+01	1.1E-02	1.2E+01	1.0E+05	2.6E+01
1969	1.1E-02	3.4E+01	9.6E-03	2.8E+01	1.2E+05	2.3E+01
1970	1.1E-03	2.3E+00	1.2E-02	2.7E+00	8.3E+03	2.9E+01
1971	1.5E-03	2.8E+01	1.6E-02	3.6E+00	1.0E+05	3.8E+01
1972	1.4E-03	1.4E+01	4.8E-03	3.3E+00	5.0E+04	1.2E+01
1973	1.7E-03	7.0E+01	2.4E-03	4.2E+00	2.5E+05	5.8E+00
1974	1.1E-03	4.2E+00	3.7E-03	2.6E+00	1.5E+04	8.9E+00
1975	9.3E-04	4.1E+00	2.1E-03	2.2E+00	1.5E+04	5.2E+00
1976	9.6E-04	4.6E+01	1.7E-03	2.3E+00	1.7E+05	4.1E+00
1977	9.3E-04	1.9E+01	2.3E-03	2.2E+00	6.9E+04	5.5E+00
1978	1.1E-03	1.9E+01	3.3E-03	2.8E+00	6.9E+04	7.9E+00
1979	7.8E-04	3.9E+01	1.6E-03	1.9E+00	1.4E+05	3.9E+00
1980	1.3E-04	1.1E+02	1.9E-03	3.0E-01	4.1E+05	4.6E+00
1981	1.3E-04	8.6E+01	3.1E-03	3.1E-01	3.1E+05	7.4E+00
1982	1.3E-04	1.5E+02	1.6E-03	3.0E-01	5.2E+05	3.8E+00
1983	1.9E-04	1.7E+02	1.9E-03	4.4E-01	6.1E+05	4.5E+00
1984	1.5E-04	2.6E+02	1.0E-03	3.6E-01	9.2E+05	2.4E+00
1985	1.5E-04	1.5E+02	1.0E-03	3.6E-01	5.6E+05	2.4E+00
1986	1.5E-04	2.4E+02	1.0E-03	3.6E-01	8.5E+05	2.4E+00
1987	1.5E-04	3.4E+02	1.0E-03	3.6E-01	1.2E+06	2.4E+00
1988	1.5E-04	1.6E+02	1.0E-03	3.6E-01	5.8E+05	2.4E+00
1989	1.5E-04	2.1E+02	1.0E-03	3.6E-01	7.7E+05	2.4E+00
1990	1.5E-04	9.2E+01	1.0E-03	3.6E-01	3.3E+05	2.4E+00
1991	1.5E-04	1.2E+02	1.0E-03	3.6E-01	4.4E+05	2.4E+00
1992	1.5E-04	2.3E+01	1.0E-03	3.6E-01	8.3E+04	2.4E+00
1993	1.5E-04	3.1E+00	1.0E-03	3.6E-01	1.1E+04	2.4E+00
1994	1.5E-04	3.4E+00	1.0E-03	3.6E-01	1.2E+04	2.4E+00
1995	1.5E-04	2.3E+00	1.0E-03	3.6E-01	8.3E+03	2.4E+00
1996	1.5E-04	4.6E+00	1.0E-03	3.6E-01	1.7E+04	2.4E+00
1997	1.5E-04	1.1E+00	1.0E-03	3.6E-01	4.1E+03	2.4E+00
1998	1.5E-04	8.4E-01	1.0E-03	3.6E-01	3.0E+03	2.4E+00
1999	1.5E-04	7.9E-01	1.0E-03	3.6E-01	2.8E+03	2.4E+00
2000	1.5E-04	8.2E-01	1.0E-03	3.6E-01	2.9E+03	2.4E+00
2001	1.5E-04	3.7E-01	1.0E-03	3.6E-01	1.3E+03	2.4E+00
2002	1.5E-04	6.6E-01	1.0E-03	3.6E-01	2.4E+03	2.4E+00
2003	1.5E-04	6.6E-01	1.0E-03	3.6E-01	2.4E+03	2.4E+00

Notes:

Intake data assume inhalation rate of 2400 m^3 per year.

^{131}I should be assumed to be in vapor form.

Tritium releases were not significant in terms of dose prior to 1967.

Tritium should be assumed to be in the form HTO.

Tritium intake values include a factor of 1.5 for skin absorption.

Dose from mixed fission products (MFPs) may be negligible relative to other contributors.

MFP concentrations included for completeness. Data are site-wide averages.

Assume MFP concentrations are Type S ^{106}Ru or ^{144}Ce if dose assessment is desired.

Assume geometric standard deviation of 3 for all values.

ATTACHMENT 4C
ANNUAL AVERAGE SITE-WIDE EXPOSURE RATES FOR OAK RIDGE NATIONAL LABORATORY

Year	Average Exposure Rate (mR/hr)
1944	0.14
1945	0.14
1946	0.14
1947	0.14
1948	0.29
1949	0.46
1950	0.08
1951	0.11
1952	0.28
1953	0.21
1954	0.14
1955	0.12
1956	0.14
1957	0.11
1958	0.13
1959	0.19
1960	0.13
1961	0.1
1962	0.106
1963	0.104
1964	0.081
1965	0.072
1966	0.075
1967	0.071
1968	0.076
1969	0.084
1970	0.072
1971	0.07
1972	0.07
1973	0.07
1974	0.07
1975	0.07
1976	0.056
1977	0.042
1978	0.037
1979	0.033
1980	0.032
1981	0.029
1982	0.03
1983	0.029
1984	0.031
1985	0.03
1986	0.03
1987	0.03
1988	0.03
1989	0.03
1990	0.03
1991	0.03
1992	0.03
1993	0.03
1994	0.03
1995	0.03
1996	0.03
1997	0.03
1998	0.03
1999	0.03
2000	0.03
2001	0.03
2002	0.03
2003	0.03

Notes: Precision of the results reflects that reported by ORNL, except where values were assumed based on results for other years. GSD of 3.0 should be applied to these values.