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ACRONYMS AND ABBREVIATIONS

ANL-E	Argonne National Laboratory-East
Ci	curie
CP-5	Chicago Pile No. 5
DOE	U.S. Department of Energy
EBWR	Experimental Boiling Water Reactor
EEOICPA	Energy Employees Occupational Illness Compensation Program Act
ft	foot
hr	hour
in.	inch
IPNS	Intense Pulsed Neutron Source
m	meter
mm	millimeter
MPC	Maximum Permissible Concentration
mph	miles per hour
mrem	millirem
NIOSH	National Institute for Occupational Safety and Health
pCi	picocurie
rem	Roentgen-equivalent man
RM	radioactive material
s	second
TBD	technical basis document
TLD	thermoluminescent dosimeter
U. S. C.	United States Code
WSA	Waste Storage Area
yr	year
ZGS	Zero Gradient Synchrotron

4.1 INTRODUCTION

Technical basis documents (TBDs) 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 the National Institute for Occupational Safety and Health (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 [DOE] facility" as defined in the Energy Employees Occupational Illness Compensation Program Act [EEOICPA; 42 U.S.C. § 73841(5) and (12)]. EEOICPA defines a DOE facility as "any building, structure, or premise, including the grounds upon which such building, structure, or premise is located … in which operations are, or have been, conducted by, or on behalf of, the Department of Energy (except for buildings, structures, premises, grounds, or operations … pertaining to the Naval Nuclear Propulsion Program)" [42 U.S.C. § 73841(12)]. Accordingly, except for the exclusion for the Naval Nuclear Propulsion Program noted above, any facility that performs or performed DOE operations of any nature whatsoever is a DOE facility encompassed by EEOICPA.

For employees of DOE or its contractors with cancer, the DOE facility definition only determines eligibility for a dose reconstruction, which is a prerequisite to a compensation decision (except for members of the Special Exposure Cohort). The compensation decision for cancer claimants is based on a section of the statute entitled "Exposure in the Performance of Duty." That provision [42 U.S.C. § 7384n(b)] says that an individual with cancer "shall be determined to have sustained that cancer in the performance of duty for purposes of the compensation program if, and only if, the cancer … was at least as likely as not related to employment at the facility [where the employee worked], as determined in accordance with the [probability of causation] guidelines established under subsection (c) …. " [42 U.S.C. § 7384n(b)]. Neither the statute nor the probability of causation guidelines (nor the dose reconstruction regulation) define "performance of duty" for DOE employees with a covered cancer or restrict the "duty" to nuclear weapons work.

As noted above, the statute includes a definition of a DOE facility that excludes "buildings, structures, premises, grounds, or operations covered by Executive Order No. 12344, dated February 1, 1982 (42 U.S.C. 7158 note), pertaining to the Naval Nuclear Propulsion Program" [42 U.S.C. § 7384l(12)]. While this definition contains an exclusion with respect to the Naval Nuclear Propulsion Program, the section of EEOICPA that deals with the compensation decision for covered employees with cancer [i.e., 42 U.S.C. § 7384n(b), entitled "Exposure in the Performance of Duty"] does not contain such an exclusion. Therefore, the statute requires NIOSH to include all radiation exposures in its dose reconstructions for employees at DOE facilities, including radiation exposures related to the Naval Nuclear Propulsion Program. As a result, all internal and external dosimetry results are considered valid for use in dose reconstruction. No efforts are made to determine the eligibility of any fraction of total measured exposure for inclusion in dose reconstruction.

Argonne National Laboratory was established on July 1, 1946 and this TBD is intended to cover since that date. The work was a continuation of that done by the Metallurgical Laboratory of the University of Chicago beginning in 1941 which is an Atomic Weapons Employer under EEOICPA. The job locations did not change until land and buildings were acquired for the laboratory.

This TBD addresses radioactive material (RM) releases from areas or facilities at the Argonne National Laboratory – East (ANL-E) site that could affect employees at that site. These releases represent potentially unrecorded or missed doses, as either direct gamma or beta-gamma from immersion in the radioactive cloud, for individuals who did not have personal dosimetry to record the dose, or as internal doses from RM inhalation.

In addition, this TBD addresses direct gamma doses resulting from facility operations. In general, these doses, if not controlled by management, increase with time and create a facility background dose. At ANL-E, these doses could have been recorded by film badges before 1970, but references containing these data have not been found. Facility background doses have been recorded by thermoluminescent dosimeters (TLDs) on a routine basis since 1972. These facility background doses (or facility fenceline doses, as they are sometimes called) are a nebulous indication of a dose that workers could receive if they inhabited outside areas at ANL-E. Golchert and Kolzow (2004) stated: "Three locations were added to the network in 1999 to monitor radioactive waste management activities." This seems to have been the practice during earlier years because some of the higher annual doses are associated with areas that temporarily stored irradiated material or radioactive waste.

4.2 EXTERNAL DOSE

As stated in Section 4.1, TLDs have been used to monitor on- and offsite environmental areas since 1972. The measurements of the gamma dose were made with dysprosium-activated, extruded calcium fluoride chips "calibrated with a National Bureau of Standard (NBS) radium-226 source. Each measurement was the average of 3 to 6 chips exposed in the same package. The measurements were made during six successive exposure periods that averaged 61 days in length and were weighted according to their exposure times in calculating the annual average for a location." (Sedlet, Golchert, and Duffy 1973). Six TLD chips were not always used; by 1975 only four were used in a packet.

The available literature does not indicate the use of film badges for this purpose prior to 1972. In 1971, 16 TLD environmental measurements were made around the ANL-E perimeter to characterize the radiation field at the site boundary (ANL 1972). The TLDs might have been part of an experiment to determine their suitability for environmental measurements, because Sedlet (1971k) describes other tests to determine fading, aging, and sensitivity characteristics and suitability of different TLD readers. Of these 16 measurements, based on a maximum of 52 days, the highest value corresponds to the location of the heliport (map grid 8H, Figure 4-1) and was projected from a reading made over 27 days to be 149 mrem/yr. In addition to these 16 TLD readings, readings at six offsite locations averaged 119 mrem/yr.

Since 1972, TLDs have been used to monitor and record suspected radiation fields that were higher than normal. TLDs have been placed at 10 to 30 ANL-E locations. As shown in Figure 4-1, the ANL-E environmental reports divide the site into a grid with letters running from west to east and numbers running from south to north. The spacing is 1,000 ft or about 300 m.

In discussing placement of the TLDs, Sedlet, Golchert, and Duffy (1975, p. 55) and Golchert, Duffy, and Sedlet (1976, p. 53) state:

> *Dosimeters were exposed at a number of locations at the site boundary to determine the dose, if any, due to Argonne operations at the closest uncontrolled approaches to the Laboratory, and at several locations on the site. The latter were chosen for two*

purposes: to determine where abnormal doses might be encountered, and where the results might be useful in determining the origin of any abnormal dose readings obtained at the boundary. Readings were also taken at five off-site locations for comparison purposes.

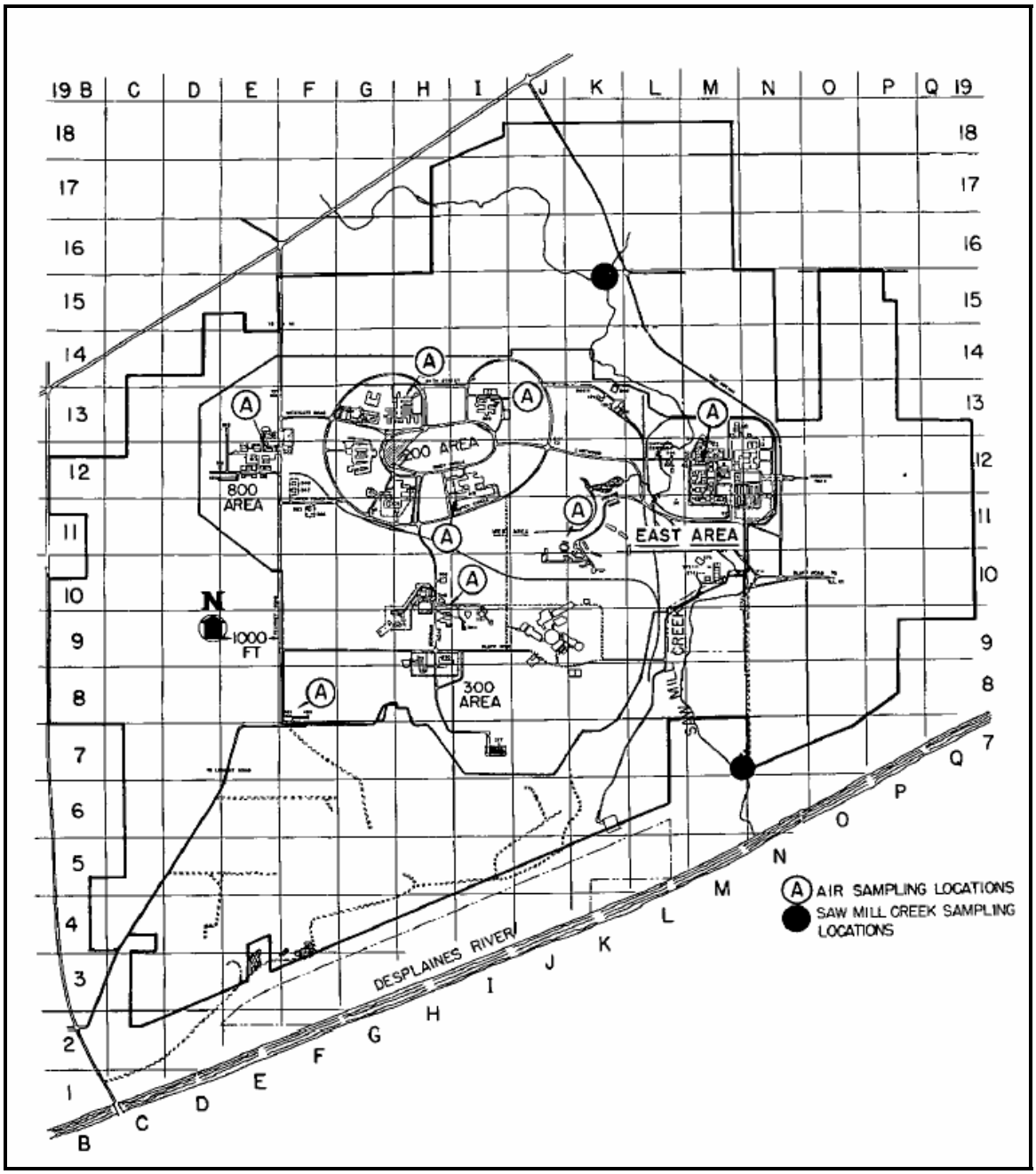

Figure 4-1. Map of ANL-E showing grid locations.

The off-site locations were Downers Grove, Lockport, Lombard, Oak Lawn, and Oakbrook; towns that are distant from ANL-E, chosen so that a background value could be determined so that TLD values on-site could be compared with the background values. Over a couple years, measurements made at these distant locations showed the background gamma radiation level

to be about 100 mrem/yr. As stated in the 1974 environmental report (Sedlet, Golchert, and Duffy 1975):

> *The off-site results averaged 99 mrem/yr with a standard deviation (for a single result) of 8.5 mrem/yr. The standard error of the mean was 1.6 mrem/yr. For 1972 and 1973, averages and standard deviations were 105 ± 5 mrem/yr and 100 ± 11 mrem/yr, respectively. Thus, the off-site readings have remained constant, within the observed statistical variations.*

With respect to TLD measurements made on-site, the environmental monitoring report for 1976 (Golchert, Duffy, and Sedlet 1977) states:

> *Dose variations from period to period at the same location, which are at least in part statistical, and the difference between locations make it difficult to determine with high certainty when site boundary doses are only a few mrem/yr above normal and due to Argonne operations. Three criteria are used here to identify such locations: 1) the results for each sampling period are frequently above the off-site average for the same period; 2) the annual; average at a location exceeds the off-site average (90 mrem/yr) plus twice the standard error in the average, 9 mrem/yr; or 3) occasional results at a particular location are significantly above the normal value for that location, although such results were still in the off-site normal range.*

For each dose measurement cited, the authors (Golchert, Duffy, and Sedlet 1978) state "The error given for an average is the 95% confidence limit calculated from the standard error."

The large error for the doses reported for several locations including 7I are due to movements into and out of the waste storage area and the repackaging of the waste that occurs at these locations. The quarterly TLD measurements for 1987 at 7I illustrate this convincingly. The quarterly readings were 7710, 8700, 11300, and 283 mrem/yr. The report for that year (Golchert and Duffy 1988) states:

> *A dramatic reduction in dose between the third and fourth quarters of 1987 at location 7I is obvious. To verify that the fourth quarter values were accurate, additional measurements were made with a Reuter-Stokes environmental radiation monitor. The results of the measurements at three different locations, 7I Center, 7I Boundary, 9H, agree well with the TLD values in Table 4.20. Subsequent discussion with operating personnel at the Waste Storage Facility indicated that radioactive waste had been shipped off-site for disposal in the late summer and early fall, and very little material was stored in this area for the remainder of the year.*

Similar variation can be seen with the TLD measurements made for the other RM storage areas located at the ANL-E site.

With time, the number and locations of TLDs have not been constant. However, some locations have been consistently monitored with time. The Waste Storage Area (WSA; 317 Area) on the southern boundary of the site, at map grid 7I, is an example of a consistently monitored location. The irradiated material storage area in the 300 Area, 50 m southeast of CP-5 (map grid 9H), is another example. Both of these areas are inside fenced areas because of the RM stored there (ANL 1982; Golchert and Kolzow 2005).

Table 4-1. Area TLD data for ANL-E areas (mrem/yr) (1972 – 2003).

Year	Heliport(8H)	200 Area	300 Area	360 Area	WSA (7I)
1972	141	121	1250; 399; 269	95; 160	1,330; 1,105
1973	145±54	99 ±34	1,645 ±1870; 358	142	2,600 ±1,440; 423 ±300
1974	128+21		2,020 ±2,320; 180 ±18; 175 ±153; 19 1±158		3,460 ±1,400; 282 ±84
1975	125±10		2600±320; 432±182; 632±1484; 200±286; 162±13		3180±709; 292±50; 245±24
1976	155±13	1,670 ±310; 823 ±24	2,420 ±200; 1,440 ±1,420; 455 ±485; 125 ±31		7,080,±1,570; 528 ±75
1977	146±30	1,110 ±350; 268 ±15	1,820 ±150; 162 ±27; 374 ±134; 104 ±7		2,640 ±900; 206 ±31
1978	136±14	770 ±47; 325 ±33; 150 ±9; 152 ±11	2,560 ±165; 124 ±17; 198 ±103; 108 ±10	260 ±140	4,750 ±1,630; 281 ±73
1979	154+19	250 ±5	2,070 ±210; 104 ±10; 118 ±44; 94 ±1	240 ±53	6340+2130; 447+61
1980	186+12	281 ±19	1,820 ±113; 118; 140 ±10; 113 ±12	144 ±15	3,710 ±2,500; 648 ±79
1981	136+7	182 ±7	953 ±258; 108 ±7; 136 ±7; 114 ±8		15,300 ±4,030; 578 ±78
1982	129±20	198 ±10	673 ±35; 100 ±4; 88 ±12		9,650 ±8,340; 509 ±288
1983	116±10	185 ±6	647 ±118		4,090 ±1,830; 292,±139
1984	122±21	164 ±18	561 ±34; 98 ±20		5,450 ±590; 457 ±221
1985	106±52	254	582 ±99		7,600 ±1,170; 941 ±379
1986	<100	102 ±6	436 ±62		4,610 ±1,200; 444 ±72
1987	103±10	112 ±5	608 ±375; 107 ±46		7,000 ±4,730; 458,±241
1988	<100	120 ±5	1,370 ±81		2,010 ±3,000; 144 ±56
1989	<100	110±3	1,216 ±47		6,000 ±112; 222 ±3
1990	<100	<100	1,015 ±46		4,330 ±1718; 165 ±45
1991	<100	<100	874 ±33		1,423 ±35
1992	<100	<100	327 ±148		3,086 ±880
1993	<100	<100	157 ±26		1,195 ±460; 103 ±15
1994	<100	<100	<100		1,455 ±553
1995	<100	<100	<100		5,516 ±2324; 129 ±12
1996	<100	<100	157 ±57		4,023 ±3921; 127 ±12
1997	<100	<100	139 ±105		1,021 ±785; 100 ±8
1998	<100	<100	170 ±66		3,237 ±417; 106 ±10
1999	<100	<100	256 ±192; 367 ±294		2,952 ±493; 105 ±21
2000	101±6	<100	179 ±108; 405 ±37; 141 ±78	783 ±504	2,333 ±1013; 114 ±21
2001	106±17	105 ±18	106 ±17; 110 ±22; 479 ±88; 230 ±149		443 ±120; 128 ±20
2002	<100	<100	967 ±276; 349 ±298; 100 ±5	735 ±52	255 ±63; 111 ±11
2003	<100	<100	1,028 ±499; 457 ±409; 515 ±88		116 ±56; 103 ±23

At least since 1972, the areas at ANL-E (200, 300, 360, 400, 500, 600, 800 areas and the east area) have operated relatively consistently. Other than the WSA (location 7I on Figure 4-1), only the 300 Area has had gamma measurements significantly above background, at an irradiated material/waste storage area. In the 200 Area, large-scale irradiation of animals with ^{60}Co and the JANUS Reactor occurred in the Biology Building (202). Because of the design of the irradiation experiments and the number of animals irradiated, when the relatively large ^{60}Co source (up to thirty-six each up to 1,500 Ci) was in the irradiation position, an area east of Building 202 had the potential for large radiation exposures. This area was monitored for excessive radiation fields and posted, and personnel were excluded from the area during irradiations. TLD readings occurred at locations near the irradiation area, which resulted in higher-than-background readings for the 200 Area.

Because of the average background value of 100 mrem/yr, this TBD analysis considered only TLD measurements above 100 mrem/yr (see Table 4-1). This limitation narrows onsite TLD measurements to the 200 Area, the 300 Area, the WSA in the 7I grid, the Heliport location in the H8 grid, and a few yearly measurements in the 360 Area.

ANL-E accelerators could generate radiation fields to which personnel could have been exposed. The argument is made that health physics personnel were trained to recognize conditions that would create personnel exposure potentials (Moe, Lasuk, Schumacher, and Hunt 1972, Moe 1988) and that these practices were put into operation to prevent personnel overexposure potentials (Wheeler 1960; Moe et al. undated).

In general, the highest TLD doses tabulated correspond to areas in which irradiated hardware is temporarily stored (i.e., 9H for the 300 Area). Personnel around or adjacent to these locations would normally wear badges, as did all personnel who entered areas or buildings where RM was used. Table 4-1 lists all environmental TLD data recorded above the 100-mrem/yr criterion. For example, the highest reading in column 4 (300 Area) was recorded for the irradiated material storage area (map grid 9H), and the highest reading in column 6 (WSA) was recorded for the center of the WSA (map grid 7I).

With respect to unmonitored gamma doses at ANL-E, because of the confined area where RM was stored outside of buildings where RM was used, unbadged workers would not be expected to receive a dose in excess of the normal background dose of 100 mrem/yr. Although operations at the WSA at 7I have been determined to perturbate the normal background TLD readings at the site boundary and at the Heliport, the perturbations are $\pm$ 50% of the background value and are not constant with time (i.e., those variations are infrequent). Since the area around the 7I WSA is wooded, workers are not expected to have inhabited this area.

High values in the 200 area are from the large ^{60}Co sources in the Biology building 202 and were located to the NE of the building. In 1977, the source arrangement was rearranged to reduce these doses.

4.3 EFFECTS FROM ONSITE AIRBORNE RADIONUCLIDE CONCENTRATION

Prevention of inhalation of hazardous material was practiced at ANL-E in the late 1940s, and keeping concentrations below “permissible levels” was attempted. A “Hazards Evaluation Division” (HED) report for August, 1946, (HED 1946) documents this culture of awareness and the attempt at prevention of inhalation of hazardous materials at Site B:

> *Activities in this site for the month produced only the usual low levels of contamination which are usually cleaned up with no general increase in hazards. Air counts show*

> *consistently the existence of air-borne materials, as a rule, well below permissible levels. The machine shop operations with BeO are under careful surveillance for proper ventilation and handling. The personnel involved have been acquainted with the probable seriousness of exposures and the necessary cooperation is expected to result in effective control of any possible danger from such operations.*

However, as mentioned in a monthly Health Physics Division (HPD) report (HPD 1946) for November, 1946, operational control was not as tight as management wished. That report states:

> *The routine surveys turned up more than the usual number of hot spots due to an apparent increase in the handling of active materials. . . An Individual was found grinding uranium bars on an open grinder in Room 121 with the result that his clothing, shoes, tools, and the floor and work bench were heavily contaminated. . .*

These two instances are provided to illustrate that, early on, operations were occurring that produced airborne concentrations of hazardous and radioactive material, that ANL management was monitoring for unusual situations, and that operations were occurring that could produce airborne materials that could be released to the environment if effluent from these operations were not filtered. Indeed, as pointed out in a health physics report (Crain 1946a, Crain 1946b, Crain 1946c), radioactive material was carried out of the plant by personnel to their homes. These reports (April 29, May 11, and May 22, 1946) document results of surveys of personnel homes where personnel had carried substantial quantities of radioactive contamination to their homes.

In November 1948, the Health Physics Division reported (HPD 1948) operations that indicated an interest in what material was being released from various facilities. The following quote is the first indication in the available literature that there was an interest in material that was being released to the environment by way of building exhaust stacks.

> *Film exposure of two shingles from around the New Chem, Room 2 stack (4-5 M and 6-8 M) and a U filter paper from Site B (683 c/m) did not show detectable tracks; the exposure time of one hour was evidently too short. A later exposure of the filter paper and a Pu standard (2212 d/m) for a 17 day period showed countable track patterns from the filter paper, while the tracks from the standard were present as a maze on the outer edge.*

In June, 1949, the Health Physics Division reported (HPD 1949a), under a subtitle “Background Radioactivity” of the report:

> *The vibrating reed electrometer and pressure chamber have been in continuous operation for a week. Twenty-four-hour blank runs with the chamber unshielded show a background of about 5 ion pairs/cc/sec. Sample counting will begin when the shielding is completed. Water samples have been obtained from most of the wells in the immediate site periphery. Rain water samples are being collected routinely from the meteorology rain gauge. Very little animal sampling is going on; adult specimens are not desired during the mating and nesting season.*

In the same report, under a subtitle, Site D, an operation is described that could have been a source of environmental contamination:

> *Uranium machining and grinding was carried out in D-17, Rooms 25 and 28 on June 9 and 10; a coolant was used. The exhaust system has not been installed and a 50-*

> *minute air sample taken during the grinding process showed three times the permissible level. All uranium operations that can produce high levels of contamination should be kept at a minimum until proper ventilation exists. The use of Comfo respirators during the interim should be regarded as a temporary emergency procedure. Personnel who are not concerned with the specific operation should be excluded from the area in which a level above permissible limits may be produced.*

The July 10 to September 1, 1949, Health Physics Division report (HPD 1949b) states:

> *The background radioactivity group has had a pressure chamber and vibrating reed in operation for about two months, studying average background levels. To date 52 air samples have been collected by Filter Queens. Additional animal specimens are being collected to supplement species not represented. Equipment for low level counting has been obtained for test.*

Although no results for the 52 air samples taken could be found in the literature, the foregoing statements quoted from ANL Health Physics Division reports shows that an environmental monitoring program was in the formative stages as ANL operations were moving to Site D. Indeed, by at least 1953, an environmental monitoring program that sampled soil, air, water, plants, and animals, on-site and off-site, had been established principally for the purpose of determining if any radioactive material was released to the environment by the operations at ANL. As stated in ANL-5446 (Sedlet 1955): *"Samples were collected from the ANL grounds and from zones approximately 10, 25, and 100 miles from the Laboratory."* For this sampling program, *"counting techniques, and most of the analytical procedures are given in detail in ANL-5069 and ANL-5289"* (Sedlet 1955). In 1954, 1625 samples were taken and *"Most of the samples were analyzed for total alpha and beta activities. In addition, some of these samples were analyzed for certain fission products; uranium, neptunium, plutonium, thorium, and potassium. Approximately 5% of the samples were not analyzed, but were stored for future use."*(Sedlet 1955).

Although all of the earlier environmental reports (Sedlet and Stehney 1954, Sedlet 1955, Sedlet 1957, Sedlet 1958a, Sedlet 1958b, Sedlet 1959, Sedlet 1961, Sedlet and Iwami 1963, Sedlet and Iwami a 1965,Sedlet and Iwami 1965b) are formal published internal reports, at least some of the information was shared with the State of Illinois and private utilities that were in the proposal stages of nuclear power use. These reports were reviewed to determine if any ANL operations resulted in releases of RM to the environment that could have resulted in significant exposure to ANL employees. The only report that identified RM resulting from ANL operations was the report for the years 1960 and 1961 (Sedlet and Iwami 1963). Quoting from that report:

> *Radioactivity originating at Argonne and leaving the site was found only in air during March 1961, and in most Sawmill Creek water samples collected during the reporting period. The airborne activity was due to iodine-131 accidentally released to the atmosphere in the exhaust air from two of the Argonne buildings. Iodine-131 was found in air samples collected on the ANL site during the latter half of February and most of March 1961. In March it was also present at two of the four off-site sampling locations (southwest and southeast of the site) at average monthly concentrations of 0.41 and 0.36 pCi/m^3, or 0.14 and 0.12% of the maximum permissible concentration (MPC), respectively. Iodine-131 was not detected in any of the off-site air samples during February 1961, the limit of detection being 0.035 pCi/m^3 (0.012% of the MPC). The average concentrations on the site during February and March were 1.9 and 3.3 pCi/m^3, respectively. The maximum concentration in any single air sample on the site was 16.7 pCi/m^3 (5.6% of the MPC). The release had essentially ceased by the end of*

> *March, and during the remainder of the year only two on-site air samples, one collected in April and one in May, contained detectable amounts of iodine-131 originating at Argonne. The iodine content of these samples was equivalent to 0.006 and 0.011% of the MPC, respectively. Except during March 1961, no significant or consistent difference could be detected between the radioactivity in air samples collected on and off the site.*

During the period of these 1950s and 1960s reports, ANL was involved in a vigorous effort to monitor and define radioactive components resulting from fallout from atmospheric and underground nuclear tests conducted in the United States and abroad. The fallout data were sent to the U.S. Atomic Energy Commission Radiological Data Appraisal Center as part of a program for characterizing fallout levels throughout the United States. Internal monthly progress reports between 1963 and 1973 document the effort to characterize this fallout and distinguish it from releases from the ANL-E site. During this time frame, airborne incidents occurred that had the potential to expose ANL personnel to radioactive particulates, but the exposure was confined to buildings as demonstrated by the internal published reports.

The purpose of these published internal environmental monitoring reports was to demonstrate that ANL operations was responsible for very little, if any, increase in the radioactive material inventory of the environment surrounding the ANL and that essentially all of the increase in environmental radioactivity was due to radioactive fallout from atmospheric and underground nuclear testing, both here and abroad. With the advent of gamma scintillation technology in the late 1960s, an effort was made to characterize gamma-emitting radioactive gaseous releases from the CP-5 reactor as reported in an internal monthly progress report (Sedlet 1968a). In the ensuing months, ambient air concentrations measured in the vicinity of Building 330, which housed the CP-5 reactor, were shown to be high. Table 4-2 lists these measurements. Because the CP-5 stack was only 15 m high, ambient air concentrations of ^{41}Ar were high with respect to the environmental maximum permissible concentration (MPC) for that radionuclide (4×10^4 pCi/m^3). All the tabulated average concentrations are above the environmental MPC value but this does not imply that the average daily concentration was above the environmental MPC since the grab samples were acquired at a location downwind where the concentration was expected to be at maximum concentration. In each case the detection limit was 50% of environmental MPC. Film badges or TLDs, had they been used and placed in appropriate locations, would have been able to measure gamma radiation from these concentrations.

Table 4-2. 1968 concentration measurements for ^{41}Ar near CP-5 (Building 330).

Ref.	No. of samples	No. of samples (<20 nCi/m^3)	Max. conc. (nCi/m^3)	Avg. conc. (nCi/m^3)
Sedlet 1968b	17	5	850	177
Sedlet 1968c	31	20	1400	110
Sedlet 1968d	34	13	730	134
Sedlet 1968e	34	19	266	78
Sedlet 1968f	30	17	990	130
Sedlet 1968g	32	14	750	150
Sedlet 1969a	26	8	570	160

Beginning in 1972, Environmental Monitoring Reports were published by ANL that had not only internal distribution, but external distribution. These later reports included sampling efforts for chemical and radioactive materials.

The later of these environmental reports, i.e., from 1979 on (Golchert, Duffy, and Sedlet 1980) contain calculations that document the highest maximum calculated offsite doses resulting from annual

releases of Ar-41 and H-3 from ANL-E facilities at that time. However, isopleths for radionuclide concentrations within the site boundary that would have been useful for determining ANL employee intakes are not available. Beginning in 1974, the environmental reports included tabulations of annual releases of airborne radionuclides. Annual releases might have been tabulated prior to 1974, but the literature sources have not been found.

4.3.1 Operational Releases

Sedlet (1969a) stated: "*Following the shut-down of CP-5 on January 5, 1969, a search will be made for argon-41 from the Juggernaut reactor*." Sedlet (1969b) stated:

> *Two air samples were collected from the Juggernaut exhaust stack after the reactor had been in operation about 4 hours. These samples contained 2.62 × 10^5 and 2.57 × 10^5 pCi ^{41}Ar/liter (2.62 E+8 and 2.57E+8 pCi/m^3), fairly substantial values. Six samples were collected downwind outside of the building on 3 different occasions, but no ^{41}Ar was detected. The wind was strong on these occasions, making detection difficult because of rapid gas dispersion. The sampling will be continued.*

Sampling over the next 9 months provided the concentration measurements near the Juggernaut reactor building (Building 335) listed in Table 4-3.

Table 4-3. 1969 Measurements of ^{41}Ar near Juggernaut Reactor (Building 335).

Ref.	No. of samples	No. of samples <20 nCi/m^3	Max. conc. (nCi/m^3)	Avg. conc. (nCi/m^3)
Sedlet 1969c	22	10	133	43
Sedlet 1969d	24	13	140	33
Sedlet 1969e	28	17	86	21
Sedlet 1969f	30	22	152	24
Sedlet 1969g	28	26	40	2
Sedlet 1969h	16	11	77	25
Sedlet 1969i	16	14	26	<20
Sedlet 1969j	8	7	49	<20
Sedlet 1970a	24	20	106	22

In early 1971, monitoring resumed in the vicinity of CP-5. Sedlet (1971a) states: "^{41}Ar sampling was resumed around CP-5 when the reactor was brought back into operation during the last quarter." Reports following this 1971 report document sampling results, which are lower than those of earlier operations as shown in Table 4-4.

Table 4-5 lists operational airborne releases of RM for ANL-E as reported in environmental reports from 1975 to the present (Golchert, Duffy, and Sedlet 1976 – 1986; Golchert and Duffy 1987 – 1990; Golchert, Duffy, and Moos 1991 and 1992; Golchert and Kolzow 1993 – 2000; Golchert, Kolzow, and Moos 2001; Golchert and Kolzow 2002 – 2005). Prior to 1975, environmental reports contained no operational release data.

Since the CP-5 reactor began operation in 1954, operational releases for the ANL-E site from 1954 to 1969 can be estimated using the statement from Sedlet, Golchert, and Duffy (1974, p. 22), Sedlet, Golchert, and Duffy (1974, 1975), and Golchert, Duffy, and Sedlet (1976, 1977): "Argon-41 and

Table 4-4. 1971 and 1972 concentration measurements for ^{41}Ar near CP-5 (Building 330).

Ref.	No. of samples	No. of samples (<20 nCi/m^3)	Max. conc. (nCi/m^3)	Avg. conc. (nCi/m^3)
Sedlet 1971b	36	29	300	35
Sedlet 1971c	32	22	218	42
Sedlet 1971d	38	24	400	18
Sedlet 1971e	34	25	495	49
Sedlet 1971f	32	18	1110	69
Sedlet 1971g	16	14	428	40
Sedlet 1971h	4	2	453	152
Sedlet 1971i	12	8	620	401
Sedlet 1971j	14	9	620	150
Sedlet 1971k	16	12	1150	107
Sedlet 1972a	16	11	335	56
Sedlet 1972b	18	10	1160	100
Sedlet 1972c	12	5	1160	179
Sedlet 1972d	12	10	1000	180

Table 4-5. Operational airborne releases at ANL-E (Ci).

Year	Ar-41	HT	HTO	Kr-85	C-11	Rn-220	I-131
1974	4.3E+4	60.	450	2.4			0.015
1975	4.5E+4	0.7	420	4.2			
1976	4.1E+4	1.9	360	7.2	1.3		
1977	4.0E+4	0.4	360	15.			
1978	3.2E+4		950	13.			
1979	7.1E+3		660	9.			
1980			9.	5.			
1981	0.4			6.6			
1982	0.56			8.4	100.		
1983	0.75			2.2	119.		
1984	0.88		62.9	71.3	85.6	76.9	
1985	1.6		46.6	128.6	152.	2,788	
1986	1.5		51.2	1.7	90	6,982	
1987	1.7		42.5	4.0	202.	6,377	
1988	2.8	9.1	40.8	7.1	86.5	3,731.2	
1989	4.5	3.6	34.3	4.0	97.7	1,858	
1990	4.77	5.25	14.71	5.18	86.7	2,606	
1991	5.29	18.9	27.12	6.8	81.8	2,946	
1992	4.84	26.6	11.38	4.64	186.4	3,000	
1993	9.4	26.2	8.51	12.87	316.8	2,024.1	
1994	4.4	40.15	67.41	20.96	306.8	1,752.7	
1995	5.3	388.1	64.61	29.8	431.2	1,034.4	
1996	9.5	363.5	79.05	1.68	709.	388.7	
1997	9.9	123.4	12.79	1.04	642.6	286.8	
1998	8.4	157.0	46.1	2.8	564.9	239.4	
1999	1.6	131.4	11.4	1.35	118.2	193.1	
2000	115.7	119.3	10.4	4.6	1610.6	46.9	
2001	81.	73.1	5.6	13.2	1250.1	35.8	
2002	94.4	91.8	9.4	25.9	1459.9	30.1	
2003	88.0	78.8	9.6	13.6	2205.6	27.28	

hydrogen-3 represent the major airborne radioactivity released from the Laboratory…." Operational releases for these two radionuclides were averaged to provide the 1954 through 1968 release values listed in Table 4-6.

The heavy-water moderated reactors CP-3 and CP-3' had tritium in the cooling water and probably also generated ^{41}Ar. Table 4-6 lists estimates of these releases based on an assumed power for CP-3 of 500 kW or less and CP-5 release values. It was also assumed that tritium control in CP-3 was less effective than in CP-5..

The Zero Gradient Synchrotron (ZGS) at its peak performance delivered about three-fourths of the proton beam power as the Intense Pulsed Neutron Source (IPNS) currently does. The reported ^{11}C and ^{41}Ar releases in Table 4-5 for IPNS show considerable fluctuation, and no explanation is provided in the annual environmental reports. We estimate the ^{11}C and ^{41}Ar releases from the ZGS listed in Table 4-6 are comparable to three-fourths of the IPNS releases for the year 2000.

Table 4-6. Estimated airborne releases (Ci).

Source	Year	Ar-41	HTO	C-11
CP-3, CP-3'	1946-1953	4,000	100	
ZGS	1963-1979	90		1,200
CP-5	1954-1973	43,000	400	

4.3.2 Dose Implications of Operational Releases

Most ANL-E airborne releases are gases that are gamma emitters such as ^{41}Ar, ^{11}C, ^{13}N, and ^{15}O. The last three are positron emitters with half-lives of 20, 13, and 2 min, respectively, and the same dose conversion factor (Sv m^3 Bq^{-1}). It is slightly claimant-favorable to sum the activities of these three and assume the release of only ^{11}C. Tritium and ^{220}Rn releases will lead to internal doses, and should be considered separately. Although ^{220}Rn is a gas, the principal dose pathway is from alphas emitted inside the lung.

U.S. Environmental Protection Agency computer code CAP-88 PC vs.2.1 was used to calculate onsite impacts. In the code, ingestion was turned off by specifying that all agricultural products are imported because there are no onsite gardens. CAP-88 is designed to calculate doses without consideration of building wake effects. However, the effect of the building wake is to spread the plume from the narrow cone established by the stack; thus, it reduces the air concentration. Neglecting it will result in a dose that is larger than the actual value and thus claimant-favorable.

To determine the doses or concentrations to which people will be exposed, the distances and directions of nearby buildings from the release buildings were determined from a map of the site and calipers. The nearest buildings are about 150 m away. The dose calculation was done by making CAP-88 calculations using the 2004 ANL-E weather file with a standard assumed release of 100 Ci for each principal nuclide released, as listed in Table 4-5, at distances of 150, 250, 350, 450, 500, 600, 650, 750, 900, 1,050, 1,500, 1,600, 1,700, 1,800, and 2,200 m for each of 16 directions of the wind rose. All calculations assumed a stack height of 15 m (based on CP-5) and an exit velocity of 10 m/s (22 mph). To calculate the dose at each distance, a table of releases for the various nuclides and years was multiplied by these calculated doses and grouped for internal and external doses. The CAP-88 calculations are for calendar year doses of 8,766 hr rather than the 2,000-hr work year, so they need to be reduced by a factor of 0.23.

Table 4-7 lists calculated doses at 150 m in all directions for the external pathway. Since a dose of 1 mrem/yr for 2000 hour occupancy or 4.4 mrem as calculated by CAP-88 will have a very small impact

on the probability of causation, values much less than that are not reported, so only the external dose before 1979 from CP-5 ^{41}Ar releases which exceed this value are reported.

However, people do not have high occupancy in all directions at the 150-m distance, so it is appropriate to consider the actual distance and direction of building locations from the major release points. In addition, for EEOICPA internal dose reconstructions, inhaled concentrations are needed to calculate organ doses.

For the period from 1946 to 1954, dose reconstructors should assign an external dose from assumed releases of airborne tritium and ^{41}Ar from CP-3 and CP-3' of 1 mrem/yr to all workers at Site A based on Table 4-7.

Table 4-7. External Dose from airborne releases at 150 m (mrem).

	External dose															
Direction	1946-1953	1954-1962	1963-1973	1974	1975	1976	1977	1978	1979	1980-1995	1996-1999	2000	2001	2002	2003	2004
N	1.0	10.8	11.0	11.0	11.5	10.5	10.3	8.3	2.0	<0.1	<0.2	0.3	0.3	0.3	0.5	0.5
NNW	0.6	6.0	6.1	6.1	6.4	5.8	5.7	4.6	1.1	<0.1	<0.2	0.2	0.2	0.2	0.3	0.3
NW	0.4	4.4	4.5	4.5	4.7	4.3	4.2	3.4	0.8	<0.1	<0.2	0.1	0.1	0.1	0.2	0.2
WNW	0.3	3.4	3.5	3.5	3.7	3.4	3.3	2.6	0.6	<0.1	<0.2	0.1	0.1	0.1	0.2	0.2
W	0.4	4.0	4.1	4.1	4.3	3.9	3.8	3.1	0.7	<0.1	<0.2	0.1	0.1	0.1	0.2	0.2
WSW	0.7	7.5	7.7	7.7	8.1	7.4	7.2	5.8	1.4	<0.1	<0.2	0.3	0.2	0.2	0.3	0.4
SW	0.9	9.2	9.4	9.4	9.9	9.0	8.8	7.1	1.7	<0.1	<0.2	0.3	0.2	0.3	0.4	0.4
SSW	0.6	6.9	7.0	7.0	7.3	6.7	6.5	5.3	1.3	<0.1	<0.2	0.2	0.2	0.2	0.3	0.3
S	0.2	2.4	2.4	2.4	2.5	2.3	2.2	1.8	0.4	<0.1	<0.2	0.1	0.1	0.1	0.1	0.1
SSE	0.8	8.3	8.5	8.5	8.9	8.1	7.9	6.4	1.5	<0.1	<0.2	0.3	0.2	0.3	0.4	0.4
SE	0.6	6.1	6.2	6.2	6.5	5.9	5.8	4.7	1.1	<0.1	<0.2	0.2	0.2	0.2	0.3	0.3
ESE	0.7	7.4	7.5	7.5	7.9	7.2	7.0	5.6	1.4	<0.1	<0.2	0.2	0.2	0.2	0.3	0.3
E	0.7	7.4	7.5	7.5	7.9	7.2	7.0	5.6	1.4	<0.1	<0.2	0.2	0.2	0.2	0.3	0.3
ENE	0.7	7.4	7.5	7.5	7.9	7.2	7.0	5.6	1.4	<0.1	<0.2	0.2	0.2	0.2	0.3	0.3
NE	0.8	9.1	9.3	9.3	9.7	8.9	8.7	7.0	1.7	<0.1	<0.2	0.3	0.2	0.3	0.4	0.4
NNE	1.0	10.8	11.0	11.0	11.5	10.5	10.3	8.3	2.0	<0.1	<0.2	0.3	0.3	0.3	0.5	0.5

The primary external dose at the main ANL-E site is from ^{41}Ar from CP-5. Onsite 24-hr doses near the reactor could be as much as 50 mrem/yr, which could be measured on TLDs near the reactor. A waste storage yard there would have masked that signal. Workers in nearby buildings would have received the doses in Table 4-8 assuming that the buildings provided no shielding and caused no interference with air movement (not likely). If a building is not listed in Table 4-8, dose reconstructors should use a nearby building. In later years, ^{11}C and ^{41}Ar releases from IPNS are the dominant contributors and primarily affect nearby buildings, but not greater than 1 mrem/yr, so dose reconstructors can ignore them. It is likely but unknown that the ZGS had comparable releases as listed in Table 4-6 and would have caused similar doses that can also be neglected.

Table 4-8. External doses (mrem) at Site D Buildings (Column 2)..

From CP-5	Affected building	Direction	Distance (m)	1946-1953	1954-1962	1963-1973	1974	1975	1976	1977	1978	1979	1980-2004
	350	SE	337	0.4	4.5	4.6	4.6	4.8	4.4	4.3	3.5	0.8	<0.1
	362	E	868	0.1	1.5	1.5	1.5	1.6	1.4	1.4	1.1	0.3	<0.1
	331	E	479	0.4	4.1	4.2	4.2	4.4	4.0	3.9	3.2	0.8	<0.1
	315	S	247	0.2	2.2	2.2	2.2	2.3	2.1	2.1	1.7	0.4	<0.1
	205	N	632	0.3	3.6	3.7	3.7	3.9	3.5	3.5	2.8	0.7	<0.1
	213	NNE	879	0.2	2.0	2.0	2.0	2.1	1.9	1.9	1.5	0.4	<0.1
	203	N	1,084	0.1	1.5	1.5	1.5	1.6	1.4	1.4	1.1	0.3	<0.1

4.4 INTERNAL DOSE FROM RADIONUCLIDE RELEASES

Only tritium and ^{220}Rn contribute to the internal dose. The tritium dose is effectively a whole-body dose from the water in the body and has an annual effect less than 0.05 mrem effective dose at 150 m in all directions per CAP-88 so it can be neglected.

The ^{220}Rn release is attributed to the "proof of breeding" program for the Navy. The Light Water Breeder Reactor at Shippingport, PA was operated about 5 years ending in late 1982. This program for production of ^{233}U from ^{232}Th was not mentioned in the 1982 EA so likely began in 1984 (Baker et.al.1996, DOE 1982). The fuel rods (~20) arrived at ANL in seven shipments between August 13, 1984 and September 23, 1985 (Graczyk et al. 1987). The fuel was sheared into sections, the sections were dissolved, analytical operations were performed, and most of the waste was processed for disposal. Reports were written on each fuel rod and are dated from October 16, 1984 to November 11, 1985. The ^{220}Rn releases would begin only after the rods were sheared, but would continue until D&D of the facility was completed. The releases are first mentioned in the 1984 environmental report, entirely consistent with the above history.

The thorium daughter ^{220}Rn has a 55 s half life and decays to 0.15 s ^{216}Po. The ^{216}Po decays to ^{212}Pb (10.64 hr) which beta decays to ^{212}Bi (1.01 hr). Detailed decay calculations shown in Figure 4-2 show that the activity of the ^{216}Po increases within 1 s to nearly the Rn-220 activity and then follows it. The ^{212}Pb activity reaches a maximum activity after about 100 s, stays about constant until 3 hr and then decays. The ^{212}Bi activity increases to a maximum value after about 3 hr and then decays somewhat after the ^{212}Pb. Since the affected buildings are nearby, the plume transit time is in the 25 to 100 sec range so the thoron daughters will be building in.

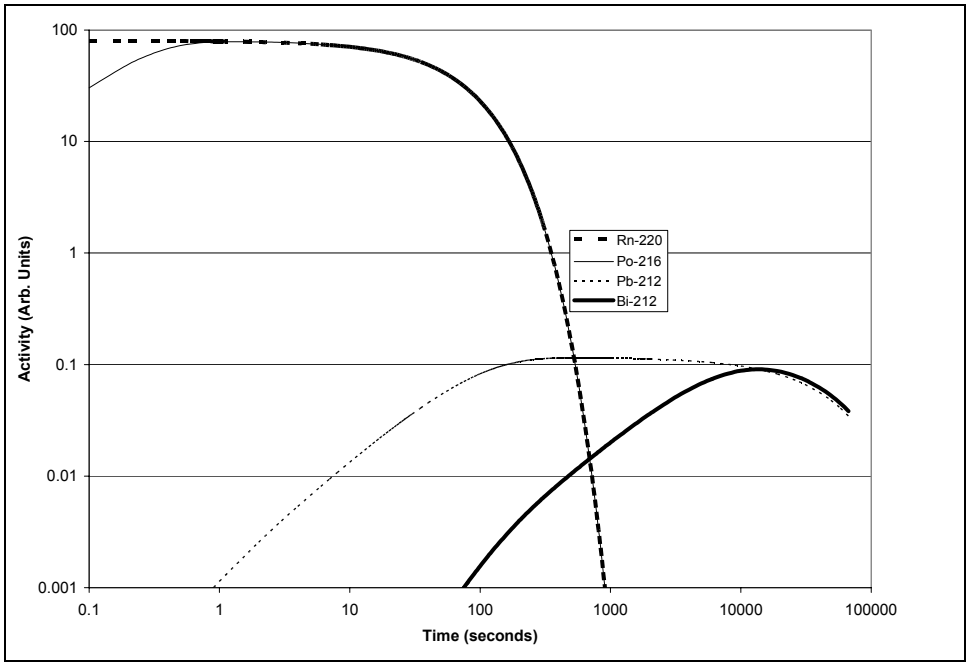

Figure 4-2. Rn-220 and daughters. .

Using CAP-88 and the distances to the nearby buildings from the M-Wing of Building 200 the intakes of ^{220}Rn for an individual breathing 2,400 m^3 of air in a work week were calculated as shown in Table 4-9. The intakes of ^{216}Po are essentially identical.

To include the longer lived radioactive daughters of 100 Ci ^{220}Rn (55 s), detailed decay calculations shown in Figure 4-2 were performed. From the weather file used as input to CAP-88, the time to travel the distance to the various buildings were calculated for each stability class based on the arithmetic average of the wind speeds for each direction. The time averaged over the stability classes was used as the time for each building. The calculated activities of ^{212}Pb and ^{212}Bi relative to the released activity of ^{220}Rn were combined with the concentration of ^{85}Kr (a long-lived surrogate to account for the dispersion but not the decay of ^{220}Rn) calculated by CAP-88 at the buildings to get the concentrations of ^{212}Pb and ^{212}Bi. Working level was calculated using the definition of working level from NCRP 97 (1988)

$$WL = 0.122\,(B) + 0.0116\,(C)$$

where B, C = concentrations of ^{212}Pb and ^{212}Bi in pCi/l.

Multiplying by 12 gives the WLM for the buildings as listed in Table 4-10.

Table 4-9. Intakes (kBq/yr) of ^{220}Rn and ^{216}Po by year from M Wing of Building 200.

Building	Dir.	Dist.(m)	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995
206	N	137	24	59	54	31	16	22	25	25	17	15	9
208	NNE	147	27	68	62	36	18	25	29	29	20	17	10
203	NE	237	18	45	41	24	12	17	19	19	13	11	7
201	E	600	3	8	7	4	2	3	3	3	2	2	1
205	SE	353	7	18	16	10	5	7	8	8	5	5	3
213	E	495	5	12	11	7	3	5	5	5	4	3	2

Table 4-10. Milli Working level months for thoron daughters from M-Wing.

Building	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
206	28	69	63	37	18	26	29	30	20	17	10	4	3	2	2
208	25	63	57	34	17	23	26	27	18	16	9	3	3	2	2
203	30	75	69	40	20	28	32	32	22	19	11	4	3	3	2
201	18	46	42	25	12	17	20	20	13	12	7	3	2	2	1
205	24	61	56	33	16	23	26	26	18	15	9	3	3	2	2
213	22	54	49	29	14	20	23	23	16	14	8	3	2	2	1

The potential for environmental dose resulting from inhalation of airborne particulate radionuclides at ANL-E was investigated (Attachment A). Information reviewed included reports of ground-level and aerial radiological surveys for Site A and Plot M, annual environmental monitoring reports, an aerial survey, onsite emission measurements for ventilation systems, and an environmental assessment (for Site D) (DOE 1997). These reports indicate that airborne particulate radionuclides were not likely to lead to a significant environmental inhalation dose to workers during operations at the Site A, Plot M, and Site D locations. The continuous monitoring and emissions monitoring did not identify significant sources of airborne radionuclides at the site. Thus, inhalation intake of particulate radionuclides is insignificant for potential environmental occupational dose at the ANL-E site.

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ATTACHMENT A
INTAKE OF PARTICULATES

Site A

At the Site A/Plot M location in the Palos Park Forest, there is no record of an organized environmental monitoring program for radioactivity during the years of operation. Any monitoring that occurred was with portable survey meters, particularly at the time of burial of material at Plot M. From 1948 to 1958, a few environmental samples were collected, mostly plants and animals. An environmental monitoring program for Plot M was established in 1954, consisting of sampling of stream water and sediment, surface soil, and soil cores. This was on an approximately every-other-year basis until 1973, when elevated tritium concentrations were detected in wells north of Plot M (Golchert and Sedlet 1977). There are no direct measurements of air concentrations of radioactive particulates at Site A or Plot M. Therefore, estimated intakes during the operational period of Site A (1946 – 1954), when workers were on the site for a significant period, are based on other environmental measurements made since the site closed. Use of Plot M was discontinued in 1949, and operation of the CP-3' reactor was discontinued in 1954.

In 1963, a few surface soil samples 50 ft north of Plot M were found to contain abnormally high alpha and beta activities, which on analysis was determined to be associated with uranium contamination. Extensive sampling followed in 1964, 1965, and 1969 to delineate the region of contamination. Because core samples did not indicate elevated levels, it was speculated that the activity was accidentally spread on the surface during burial or storage bin removal operations, which occurred prior to July 1949 (Golchert and Sedlet 1977). Analyses in 1973 and 1975 indicated one sample with elevated concentrations of plutonium (0.22 pCi/g versus expected fallout levels of 0.05 pCi/g) in the same area of contamination. The uranium concentration in this sample, however, was indistinguishable from background. Thorium was at background levels in all samples. Average air concentrations of uranium and plutonium (no isotopes specified) of 1.5×10^{-5} Bq/m^3 (0.4 fCi/m^3) and 3.0×10^{-7} Bq/m^3 (8 aCi/m^3), respectively, were estimated (Golchert and Sedlet 1977). These estimates were based on resuspension factors measured for vegetation-covered soil. Assuming an inhalation rate of 2,400 m^3/yr, inhalation dose coefficients can be applied to estimate corresponding organ doses, assuming all uranium is ^{234}U and all plutonium is ^{239}Pu. These isotopes have the highest organ dose coefficients associated with the respective elements. Using ICRP Publication 68 (ICRP 2001) organ dose coefficients of 7.5×10^{-5} Sv/Bq for ^{234}U (for extrathoracic airways) and 1.0×10^{-3} Sv/Bq for ^{239}Pu (for bone surfaces), estimated inhalation doses due to resuspension of the contaminated soil in Plot M are on the order of 3×10^{-6} Sv/yr (0.3 mrem/yr) for inhaled ^{234}U, and 7×10^{-7} Sv/yr (0.07 mrem/yr) for inhaled ^{239}Pu.

A 1976 aerial gamma radiological survey of Site A and Plot M used sodium iodide detectors mounted in a helicopter (Jobst, Houk, and Mohr 1978). The only indications of above-normal concentrations were a few isolated areas within the Site A boundaries, indicating ^{137}Cs contamination from past operations at the site. The results were indeterminate for Plot M. Soil analysis confirmed the contamination in the Site A area with the highest detected ^{137}Cs, which was approximately three times the concentration attributable to fallout. A gamma-ray survey using a portable sodium iodide detector of Plot M in 1976 by the ANL-E Radiological Resurvey Team did not detect radioactivity in surface soil attributable to buried waste materials (Golchert and Sedlet 1977).

A more extensive evaluation of surface and subsurface soil at Site A and Plot M occurred in 1976 (Golchert and Sedlet 1977), which was 22 years after operations ceased at these locations. The subsurface study took soil cores near the buried CP-3' containment shell at Site A and at various locations around Plot M, and analyzed them for tritiated water and gamma-emitting radionuclides. Selected samples were analyzed for nonvolatile alpha and beta activity, ^{90}Sr, uranium, and plutonium

(Golchert and Sedlet 1977). The results of the subsurface study indicated that all analyte concentrations were in the range consistent with that of natural background and fallout, with the exception of tritiated water for Site A and Plot M. Thus, the ^{137}Cs detected by the aerial gamma survey at Site A did not appear to arise from the contaminated reactor shell location, which was buried under 7 m of building rubble and soil (Section 2.2.7.1 of ORAU 2006).

A surface soil study was not conducted for Plot M in this 1976 survey; however, the subsurface study indicated tritiated water as a contaminant attributable to the presence of buried waste at this location. For Site A, surface soil (to 6-in. depths) was sampled at 25-ft intervals in the center of the site (near the buried reactor shell location), 50-ft intervals outside this area, and 100-ft intervals at the Site A perimeter. Of the 105 samples taken, one indicated the presence of ^{125}Sb above its detection limit and had the highest ^{137}Cs concentration (5.5 pCi/g) of all samples. Estimated fallout concentrations for ^{137}Cs at the time average 0.5 pCi/g, but range up to 1.0 pCi/g. Thus, the highest detected cesium concentration in soil appeared to be about 10 times the average fallout concentration. Three surface samples from Site A had ^{60}Co in detectable concentrations. The two samples with the highest ^{137}Cs concentrations were also analyzed for ^{90}Sr. The ^{90}Sr concentrations in these samples exceeded the fallout concentrations of this isotope (0.1 to 0.2 pCi/g) by approximately a factor of 5 to 10, which is comparable to the ^{137}Cs numbers. The results of these studies indicated that surface contamination existed at random locations but, on average, at a very low level. The majority of the samples showing ^{137}Cs concentrations in excess of 1.0 pCi/g were only slightly in excess. The average excess ^{137}Cs concentration (i.e., above 0.5 pCi/g) in the 105 samples was about 0.2 pCi/g. Correcting for decay back to 1946, the average excess concentration would probably be at most a factor of two higher than the 1976 value [reasonably assuming negligible downward migration due to the normally high sorptive behavior of cesium on soil (Sheppard and Thibault 1990)], or 0.4 pCi/g. Adding this corrected excess to the average fallout concentration of 0.5 pCi/g gives an average concentration of 0.9 pCi/g for 105 samples, which is still within the range for ^{137}Cs fallout concentrations.

Based on the evidence collected since operations ended at Site A and Plot M, it is reasonable to assume that environmental inhalation exposures to particulates during operations which ended in 1954 were negligible. The only surface contamination identified over the years at the Plot M location involved minor amounts of uranium and plutonium. Estimated inhalation doses are less than 1 mrem/yr. The surface contamination identified at Site A was, on average, very low-level and not readily distinguishable from fallout concentrations.

Site D

For Site D, monthly concentrations of long-lived alpha and beta activities between 1953 and 1973 have been reported for air filter samples collected at ANL-E (ANL 1965, 1968; Sedlet 1955, 1957, 1958a,b, 1959, 1961; Sedlet and Iwami 1963, 1965a,b; Sedlet and Stehney, 1954; Sedlet, Golchert, and Duffy 1973, 1974). Beginning in 1960, concentrations of principal gamma-ray emitters, as determined by spectroscopy, were reported. Annual reports describing monthly concentrations were not formally produced between 1964 and 1971, but informal reports contain monthly concentrations for the first half of 1965 and for 1968 (ANL 1965, 1968). The reported monthly concentrations represent continuous monitoring, and are reported as a site average (not by location). In March 1972, sampling for radiochemical analysis of isotopes of plutonium, thorium, and uranium in airborne particulates began in the northeast area of the site (Sedlet, Golchert, and Duffy 1973). Isotopes of strontium were added to these analyses in 1973 (Sedlet, Golchert, and Duffy 1974).

In 1953, continuous air monitors were placed at five onsite locations – one on the site periphery near East Area, one in the 200 Area, one between the Experimental Boiling Water Reactor (EBWR) and CP-5 reactors in the 300 Area, one in the 800 Area, and one on the southwest corner of the site

(Sedlet 1958b). Three offsite sampling locations were added in mid-1957 – 6 miles northwest, 10 miles southwest, and 13 miles southeast of the site (Sedlet 1958b), which increased to five in 1962. The number of continuously monitored onsite locations expanded to six in 1959 (Sedlet 1961) and to nine by 1972 (Sedlet, Golchert, and Duffy 1973). Figure A-1 shows the locations of air samplers in 1968. Additional airborne particulate monitoring occurred when specific operations could increase

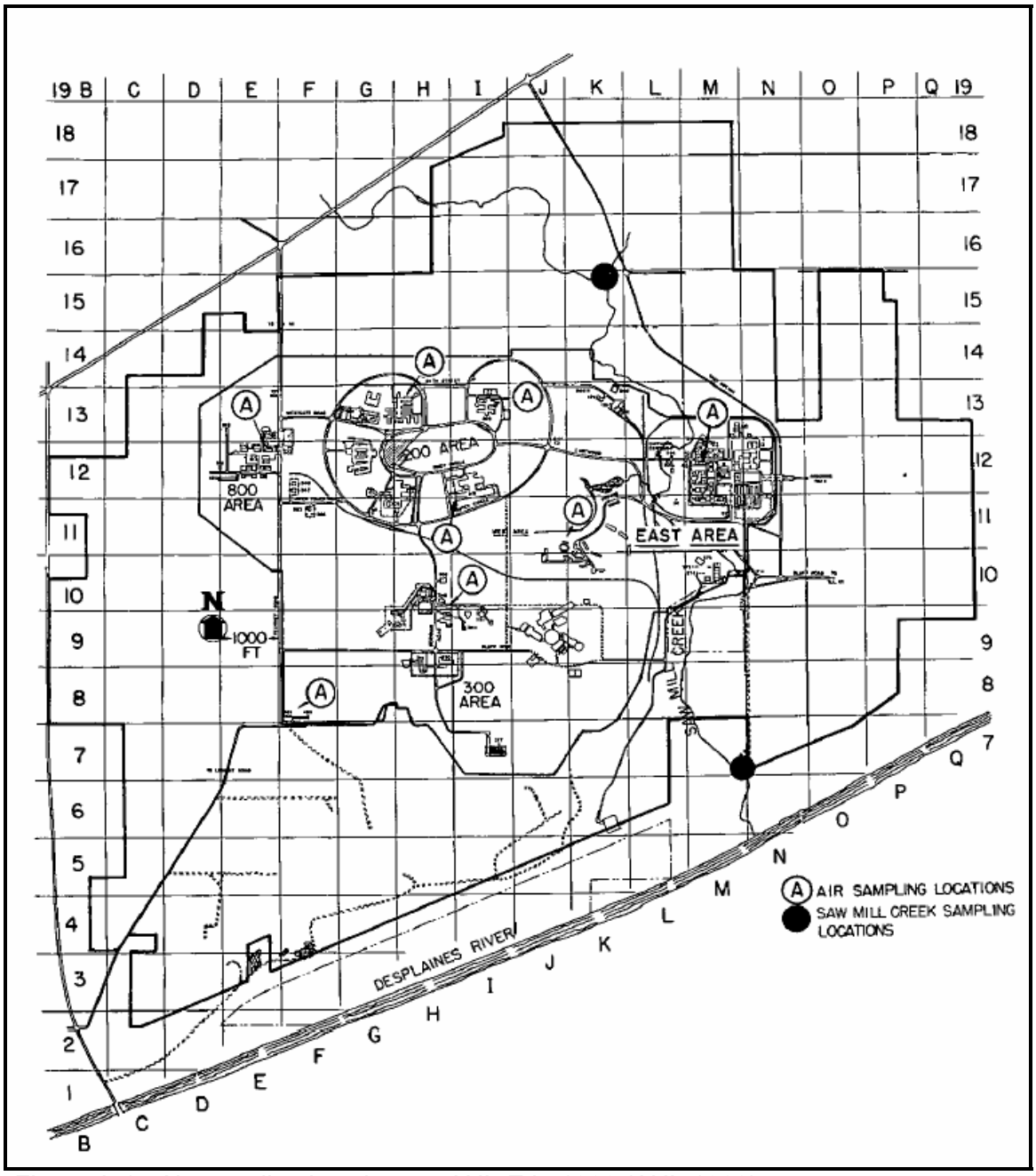

Figure A-1. Air sampling locations at ANL-E in 1968 (ANL 1968).

localized airborne radioactive particulates. For example, in 1958 the EBWR operated with a simulated defective fuel element, and additional air samplers were placed in and near the EBWR building to detect activity leaving the reactor stack. In this particular case, no activity attributable to the EBWR was detected in air samples collected outside the building (Sedlet 1959). Similarly,

beginning in October 1972, samples for radiochemical analyses for plutonium, thorium, and uranium in the 300 Area, close to the center of plutonium usage on the site, were collected (Sedlet, Golchert, and Duffy 1973).

Table A-1 lists the average of measured gross onsite concentrations (not corrected for fallout contributions) of long-lived alpha- and beta-emitting particulates between 1953 and 1973, along with the average of measured offsite concentrations of these particulates during this period. The table indicates that average onsite particulate concentrations were not significantly different from offsite concentrations. A similar finding was made for the principal gamma emitters between 1960 and 1974. One minor exception occurred when detection limits for ^{140}Ba and ^{140}La (from decay of ^{140}Xe) became low enough to distinguish onsite concentrations from nondetectable offsite concentrations of these isotopes in 1973 (Sedlet, Golchert, and Duffy 1974). However, the measured annual average concentrations were on the order of 4.4×10^{-5} Bq/m^3 (1.2×10^{-9} µCi/m^3), which corresponds to an estimated annual inhalation dose less than 0.001 mrem for occupational exposure over the year. Thus, the onsite outdoor environment does not appear to have contributed a significant exposure to radionuclides via inhalation of particulates between 1953 and 1973, assuming that exposures for years when measurements were not made or reported were similar to those for other years.

Table A-1. Radioactivity in airborne particulates on and off ANL-E site before 1974.

	Average onsite concentration (Bq/m^3)		Average offsite concentration (Bq/m^3)		
	Site D				
Year	LL alpha[a]	LL beta[b]	LL alpha[a]	LL beta[b]	Reference
1948–1952					Not available
1953	3.7E-04	4.4E-02	Not reported	Not reported	Sedlet 1958a
1954	3.7E-04	1.5E-02	Not reported	Not reported	Sedlet 1958a
1955	2.6E-04	3.0E-02	Not reported	Not reported	Sedlet 1958a
1956	3.7E-04	4.8E-02	Not reported	Not reported	Sedlet 1958a
1957	3.0E-04	7.8E-02	2.6E-04	7.8E-02	Sedlet 1958b
1958	2.6E-04	1.0E-01	2.6E-04	1.3E-01	Sedlet 1959
1959	2.3E-04	8.1E-02	2.1E-04	8.5E-02	Sedlet 1961
1960	2.6E-04	3.7E-03	2.2E-04	3.3E-03	Sedlet and Iwami 1963
1961	1.5E-04	1.0E-01	1.5E-04	9.3E-02	Sedlet and Iwami 1963
1962	1.9E-04	1.8E-01	1.9E-04	1.8E-01	Sedlet and Iwami 1965a
1963	2.2E-04	2.1E-01	2.2E-04	2.1E-01	Sedlet and Iwami 1965a
1964	1.9E-04	3.7E-02	1.9E-04	3.7E-02	Sedlet and Iwami 1965b
1965	1.4E-04	1.5E-02	1.4E-04	1.5E-02	ANL 1965
1966					Not available
1967					Not available
1968	1.6E-04	6.7E-03	1.8E-04	7.0E-03	ANL 1968
1969					Not available
1970					Not available
1971					Not available
1972	8.9E-05	3.6E-03	9.3E-05	3.6E-03	Sedlet, Golchert, and Duffy 1973
1973	9.3E-05	1.3E-03	8.9E-05	1.3E-03	Sedlet, Golchert, and Duffy 1974

a. Long-lived total alpha concentration.
b. Long-lived total beta concentration.

In 1974, reporting of continuous particulate air monitoring results at onsite locations was discontinued; annual reports provided only perimeter and offsite measurements (Sedlet, Golchert and Duffy 1975; Golchert, Duffy, and Sedlet 1976 – 1986; Golchert and Duffy 1987 – 1990, Golchert, Duffy, and Moos

1991, 1992; Golchert and Kolzow 1993 – 2000, 2002 – 2005; Golchert, Kolzow, and Moos 2001). Figure A-2 shows the locations of the monitoring stations in 1996. The focus of these measurements was to assess the degree to which emissions from ANL-E might affect the offsite environment (or location of closest approach by the public). One of the perimeter monitors (at grid point 7I, Figure A-2) was adjacent to the WSA in Area 317, and one is in the predominant downwind direction from the site (at grid point 12N, Figure A-2). Other monitors are on the perimeter, but close to ANL-E facilities (two monitors at grid point 14H, Figure A-2). Table A-2 lists the average of measured perimeter concentrations of long-lived alpha- and beta-emitting particulates between 1974 and 2004, along with the average of measured offsite concentrations of these particulates during this period.

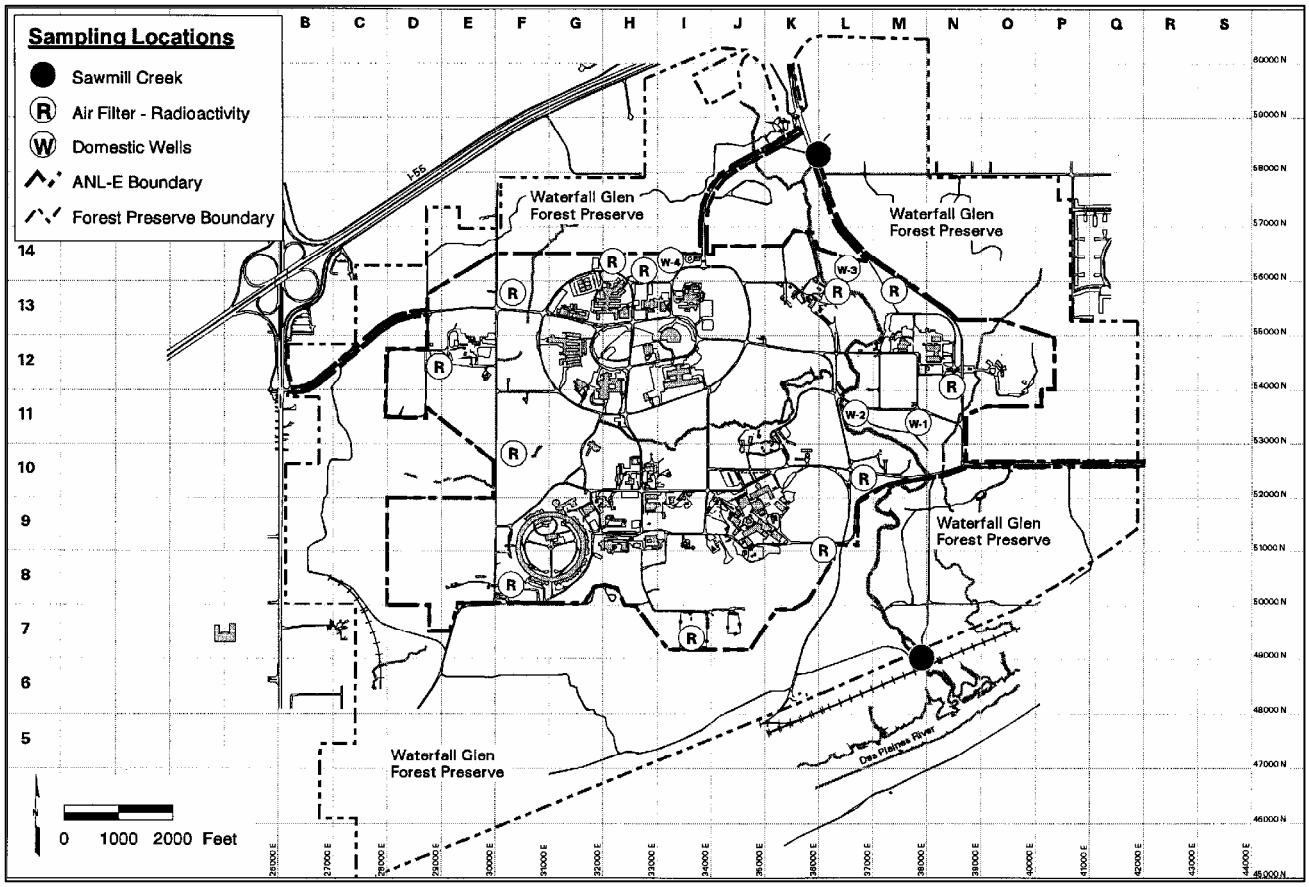

Figure A-2. Sampling locations at ANL-E in 1996 (Golchert and Kolzow 1997).

As a supplement to the environmental program at ANL-E and in addition to perimeter monitoring, an aerial radiological survey of the laboratory and its surroundings was conducted in 1977 (Jobst 1982). An adjunct study to the aerial measurements was the collection of 27 soil samples from randomly selected locations at the laboratory, which were analyzed for radioisotope composition. The most significant anomalies in external dose rates were attributed largely to gamma sources used in research or isotopes in the WSA in Area 317. Other than ^{3}H, only 2 of the 27 soil samples contained small, but measurable, quantities of isotopes that might have been associated with ANL-E operations. In particular, one soil sample contained a small amount of ^{60}Co, and another contained uranium not in equilibrium with its daughters, although the ^{238}U concentration was at a level consistent with that of background. Thus, the aerial survey did not indicate a potential for significant inhalation exposure to particulate radionuclides, other than perhaps in the 317 Area, where a perimeter monitor was located.

For many years, emissions of radioactive air particulates were generally not reported in annual environmental reports, other than a statement that "various [or several other] fission products" were released from reactor stacks in "millicurie or smaller amounts" (Golchert and Duffy 1975; Golchert, Duffy, and Sedlet 1976 – 1980). Between 1980 and 1990, no mention of air particulate emissions was made in these reports (Duffy and Sedlet 1981 – 1986; Golchert and Duffy 1987 – 1990).

Table A-2. Radioactivity in airborne particulates at perimeter and off ANL-E site (after 1974).

Year	Average perimeter concentration (Bq/m3) Site D		Average offsite concentration (Bq/m3)		Reference
	LL alpha[a]	LL beta[b]	LL alpha[a]	LL beta[b]	
1974	8.9E-05	4.8E-03	8.5E-05	4.8E-03	Sedlet, Golchert, & Duffy 1975
1975	8.5E-05	2.6E-03	8.5E-05	2.6E-03	Golchert, Duffy, & Sedlet 1976
1976	9.6E-05	1.7E-03	9.3E-05	1.7E-03	Golchert, Duffy, & Sedlet 1977
1977	8.1E-05	5.7E-03	8.1E-05	5.6E-03	Golchert, Duffy, & Sedlet 1978
1978	8.1E-05	3.6E-03	7.4E-05	3.6E-03	Golchert, Duffy, & Sedlet 1979
1979	7.0E-05	1.3E-03	7.4E-05	1.2E-03	Golchert, Duffy, & Sedlet 1980
1980	6.7E-05	1.1E-03	7.8E-05	1.1E-03	Golchert, Duffy, & Sedlet 1981
1981	7.0E-05	4.4E-03	7.4E-05	4.4E-03	Golchert, Duffy, & Sedlet 1982
1982	6.3E-05	9.6E-04	6.7E-05	9.3E-04	Golchert, Duffy, & Sedlet 1983
1983	9.3E-05	1.0E-03	7.8E-05	9.3E-04	Golchert, Duffy, & Sedlet 1984
1984	8.1E-05	8.9E-04	7.4E-05	8.5E-04	Golchert, Duffy, & Sedlet 1985
1985	7.8E-05	8.9E-04	7.0E-05	7.8E-04	Golchert, Duffy, & Sedlet 1986
1986	7.8E-05	1.5E-03	6.7E-05	1.4E-03	Golchert and Duffy 1987
1987	8.1E-05	9.3E-04	7.4E-05	8.5E-04	Golchert and Duffy 1988
1988	8.5E-05	9.6E-04	7.0E-05	9.3E-04	Golchert and Duffy 1989
1989	8.5E-05	1.0E-03	7.4E-05	9.5E-04	Golchert and Duffy 1990
1990	6.7E-05	1.0E-03	7.4E-05	1.1E-03	Golchert, Duffy, & Moos 1991
1991	6.3E-05	9.3E-04	7.8E-05	1.1E-03	Golchert, Duffy, & Moos 1992
1992	5.9E-05	9.3E-04	7.0E-05	1.0E-03	Golchert and Kolzow 1993
1993	5.6E-05	8.9E-04	6.3E-05	9.5E-04	Golchert and Kolzow 1994
1994	6.3E-05	1.1E-03	7.0E-05	1.0E-03	Golchert and Kolzow 1995
1995	6.3E-05	1.1E-03	5.9E-05	1.0E-03	Golchert and Kolzow 1996
1996	5.6E-05	1.0E-03	6.3E-05	1.0E-03	Golchert and Kolzow 1997
1997	4.8E-05	9.3E-04	5.9E-05	1.1E-03	Golchert and Kolzow 1998
1998	4.1E-05	8.9E-04	4.8E-05	8.7E-04	Golchert and Kolzow 1999
1999	3.3E-05	7.0E-04	4.1E-05	6.9E-04	Golchert and Kolzow 2000
2000	3.3E-05	6.3E-04	4.8E-05	7.4E-04	Golchert, Kolzow, & Moos 2001
2001	3.7E-05	5.6E-04	8.5E-05	8.1E-04	Golchert and Kolzow 2002
2002	4.4E-05	5.9E-04	7.0E-05	5.1E-04	Golchert and Kolzow 2003
2003	5.2E-05	5.2E-04	8.0E-05	7.3E-04	Golchert and Kolzow 2004
2004	4.4E-05	5.6E-04	7.4E-05	7.4E-04	Golchert and Kolzow 2005

a. Long-lived total alpha concentration.
b. Long-lived total beta concentration.

However, in 1990 increased National Emission Standards for Hazardous Air Pollutants requirements for monitoring and reporting resulted in expanded reporting requirements for emission points, including ventilation systems for hot cell facilities, for currently operating and inactive reactors, for particle accelerators, and associated with the DOE operated New Brunswick Laboratory. There was only one report of potential particulate radionuclide emissions exceeding 3.7×10^7 Bq/yr (1 mCi/yr); this was in 1990 for ^{241}Pu (7.0×10^7 Bq/yr, or 1.9 mCi/yr) from the New Brunswick Laboratory (Building 350).

An estimate of air concentration resulting from this 1990 ^{241}Pu release can be made using a simplistic screening-level model from Volume 1 of NCRP (1996). The NCRP model adopted for this calculation is a simple dispersion model that accounts for potential increases in exposure due to building wake

effects for close-in receptors (i.e., those less than 100 m from the source location). Air concentration (C) of ^{241}Pu was calculated from:

$$C = (f\ Q)/(\pi\ u\ h\ K)\ Bq/m^3 \quad \text{(A-1)}$$

where:

C = concentration at receptor (Bq/m^3)
Q = stack or building vent release rate (Bq/s)
f = wind frequency
u = wind speed (m/s)
h = height of effluent release (m)
K = constant (m)

Default values from NCRP (1996) are used for K of 1 m and f of 0.25 (the maximum frequency for any compass point). Wind rose data for ANL-E for 1990 (Golchert, Duffy, and Moos 1991) indicate variable wind speeds over the year. This calculation assumed the annual average wind speed was on the order of 4 m/s, which is in the lower range of wind speeds, and thus tends to overestimate the resulting calculated concentration. The assumed height of effluent release from the ventilation system of Building 350 is 10 m, because the fan loft of this building is above the third story (Wescott and O'Rourke 2001). Assuming a ^{241}Pu release rate of 7×10^7 Bq/yr, or 2.2 Bq/s, the estimated concentration of ^{241}Pu in the vicinity of the release point is 4.4×10^{-3} Bq/m^3. This corresponds to an effective dose of less than 1 mrem, or an organ dose (bone surface) of approximately 20 mrem in 1990 for continuous occupational exposure (i.e., 2,000 hr/yr). Because it is not plausible that an individual would remain outside within 100 m of the building for a major part of the year, this upper limit estimate of dose rate does not suggest that inhalation dose due to this release of ^{241}Pu in 1990 is likely to be significant with respect to environmental dose.

Although some of the remediation activities, particularly at Area 317, and decontamination and decommissioning activities could have been a source of onsite particulate radionuclides, these projects typically required air monitoring, air sampling, and bioassay sampling in addition to personal protective equipment in many cases (Baker et al. 1996). DOE (1997) estimated that noninvolved workers in the 570 Area (Unlined Holding Basin) or the 317/319/ENE Area (Waste Storage/Disposal areas) during remediation activities would either require respiratory protection or restriction from the areas. Assuming the nearest nonprotected workers are 600 m from these areas, the calculated dose to noninvolved workers was 7×10^{-6} mrem (DOE 1997).

The information for Site D (present ANL-E location) can be summarized as follows. Between 1953 and 1974, onsite monitoring for radioactive air particulates indicated that there were no significant differences between on- and offsite air particulates and, thus, ANL-E operations did not contribute significantly to an environmental dose via inhalation of air particulates. Although onsite measurements ended in 1974, the aerial survey and soil sampling done in 1977 suggested that surface contamination by ANL-E airborne particulates was minimal up to that point. Monitoring of emissions for reactor stacks began in the 1970s and expanded to address most ventilation systems in the 1990s and indicated minimal release of particulate radionuclides. Thus, there is no evidence that radioactive air particulates have posed a significant environmental dose to ANL-E workers.