

# Assessment of Potential for Radon Exposure at ANL-E

White Paper

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National Institute for Occupational  
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## INTRODUCTION

During the June 27, 2018, Advisory Board on Radiation and Worker Health (ABRWH) Argonne National Laboratory-East (ANL-E) work group teleconference meeting regarding the current status of Sanford Cohen & Associates (SC&A) findings and observations against the ANL-E Site Profile [ORAUT 2006a to 2006e; SC&A 2009], the ABRWH requested additional information on radon sources and the potential for radon exposure at ANL-E (affiliated with Observation/Secondary Issue 4 in SC&A [2009]).

Observation 4 is entitled “Internal Dose to Workers from Radon Exposures is Not Considered” [SC&A 2009] and states since ANL-E handled  $^{226}\text{Ra}$ ,  $^{227}\text{Ac}$ , and  $^{230}\text{Th}$  as part of the research and development, as well as other programs, there is a potential for exposure to radon ( $^{222}\text{Rn}$ ), actinon ( $^{219}\text{Rn}$ ), and thoron ( $^{220}\text{Rn}$ ), which is not thoroughly discussed in the Site Profile.

Past discussions between SC&A and the National Institute for Occupational Safety and Health (NIOSH), documented in the Board Review System and in SC&A memos, include more specific information regarding Observation 4. A summary of this dialogue is provided here:

*NIOSH has not found any indication of routine worker monitoring for radon. Given that there are not large quantities of uranium or radium ore stored or handled at ANL-E, any radon exposure likely would have been minimal and secondary to other nuclide exposures (i.e., radium, actinium, and thorium). SC&A finds that there were opportunities for exposure to Ra-226, Ac-227, and thorium, according to the Site Profile, for ANL-E, therefore further investigation into occupational exposures to radon, thoron, and actinon are necessary. NIOSH has found indications that protective measures (e.g., radon traps in areas of radium work, use of ventilated hoods) and area air monitoring were implemented, even in early operations that had the potential for high radon exposures. SC&A asked about whether there is ANL-E bioassay data for radium and its decay products, which would indicate the potential exposure to radon. NIOSH found over 800 bioassay samples for radium between 1949 and 1979, but the documentation is insufficient to know the process and workplace specific information (e.g., chemical and physical form, quantity) about what radium handling was done for each sample to know whether radon exposures would have been significant. But given the ANL-E policy of performing radiation work in fumehoods, glove boxes, and ventilated enclosures,<sup>1</sup> NIOSH assumes that the occupational internal exposure from radon, actinon, and thoron (and their progeny) due to handling and use of these sources was minimal.*

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<sup>1</sup> Policy of air filtration where alpha contamination was possible is outlined in Fleury [1946]; Wing M of building 200 required ventilation and use of hoods and gloveboxes [DOE 1982, PDF p. 32]; other routine reports in tables below verify implementation of these policies.

This white paper will further explore the assumption that occupational exposure to radon, actinon, and thoron was minimal by reviewing the potential sources of radon occupational exposure at ANL-E and the resulting exposure potential.

NOTE: This paper uses the term “radon” to discuss any isotope of radon and uses the specific isotopes for discussing specific exposures.

### **CURRENT ANL-E SITE PROFILE DISCUSSION OF RADON EXPOSURE AND ASSIGNMENT OF RADON DOSES**

No radon ( $^{220}\text{Rn}$ ,  $^{219}\text{Rn}$ , or  $^{222}\text{Rn}$ ) doses are assigned in the ANL-E Occupational Internal Dose technical basis document (TBD) [ORAUT 2006a]. This approach is based on the assumption that the work with radium, actinium, and thorium was performed in laboratories with engineered controls, like ventilation, gloveboxes, etc., and was a part of a routine workplace monitoring program that would have prevented routine occupational exposures to the gaseous daughter products. Due to the demonstrated storage and working approaches (i.e., known sources of radon were encapsulated and stored in unoccupied areas, work was performed in fumehoods), radon exposure would only have been feasible during transfer of material to laboratory locations. Records demonstrate control of and protection from exposure were accomplished by wrapping or sealing source materials to limit radon emanation and by use of respiratory protection by workers, minimizing such exposures [Rose 1949, PDF pp. 5, 17, 19; ANL 1953, PDF p. 10; Lonergan and Novak 1959, PDF p. 24; Lonergan and Novak 1963, PDF p. 168].

Radon ( $^{220}\text{Rn}$ ) releases and emissions are noted in environmental monitoring at ANL-E starting in 1984 from specific buildings where specific operations were performed. Doses from these releases are assigned via the Occupational Environmental Dose TBD [ORAUT 2006b] as an environmental internal dose to workers in the 200 Area facilities at ANL-E and to workers whose work locations cannot be identified, as a claimant favorable assumption.

These environmental radon releases are associated with a specific program in which spent fuel rods with high quantities of  $^{232}\text{Th}$  were sheared, analyzed, and prepared for disposal in the M Wing of Building 200 [ORAUT 2006b]. This work was done in controlled-ventilated areas (i.e., hot cells, radiochemistry labs with fume hoods) [DOE 1982]. Due to the engineered controls and established routine monitoring, occupational exposure from the source material was controlled and off-normal events would have been detected. Exposure to radon ( $^{220}\text{Rn}$ ) due to the emanation from this source material would have been controlled by the workplace controls in place for the source material. These activities, however, did result in  $^{220}\text{Rn}$  environmental releases through the high-efficiency particulate air (HEPA)-filtered exhaust of the ventilation system from the M Wing of Building 200. These releases resulted in potential environmental exposures to individuals in nearby buildings in the 200 Area. Since this exposure pathway is already discussed in the Occupational Environmental Dose TBD, further assessment of exposures from this program will not be described in this document.

## HISTORICAL BACKGROUND

The Metallurgical Laboratory (also called Met Lab) in Chicago was the direct predecessor to ANL-E, which began operations on July 1, 1946 [Hilberry 1949]. Although Met Lab facilities and operations are not covered by the ANL-E TBD [ORAUT 2006a], evaluation of records of Met Lab operations indicate familiarity with radon and radon progeny as a health risk, an interference with air and surface monitoring performed for other hazardous radionuclides (e.g., uranium and plutonium), and as an indicator to measure radium content in the body.

The Met Lab was involved in early biomedical research on human exposure to radium, including radium watch dial painters and patients treated with radium, because it was "...concerned especially with protecting workers exposed to radiation..." [Holl et al. 1997, PDF p. 64]. Because this research was performed under laboratory conditions to evaluate radium in patients who typically received the radium intakes from their workplace (i.e., radium watch dial painters) or medical procedures, it did not involve "free" quantities of radium large enough to represent a radon risk to researchers. The exposure route to researchers for this work would have been via the exhalation of radon from the study subjects. Rowland [1994] presented results for 2,403 individuals for whom radium body burdens were determined. Of these results, 144 results (6%) were 1  $\mu\text{Ci}$  or greater and the largest derived body burden listed was 25  $\mu\text{Ci}$ . From ORAUT-OTIB-0025, *Estimation of Radium-226 Activity in the Body from Breath Radon-222 Measurements* [ORAUT 2005], the expected  $^{222}\text{Rn}$  concentration in breath from an individual with a body burden of 25  $\mu\text{Ci}$   $^{226}\text{Ra}$  is 100 pCi/l. This is coincidentally the concentration of  $^{222}\text{Rn}$  considered equivalent to one working level (WL) of exposure for continuous exposure conditions. However, the concentration the worker would have been exposed to was reduced due to mixing with room air, and the total exposure time would have been short (on the order of hours or days), rather than continuous. Therefore, even assuming the highest potential exposure situation (i.e., working with the experimental subject with the highest body burden), there was no significant exposure to exhaled radon for laboratory personnel.

This early Met Lab research in radiation effects is noted here to illustrate the early development of techniques (e.g., radon breath analysis) for determining radium body burdens, which used radon measurement technologies. It is therefore reasonable to assume the relationship between radium, radon, and radon progeny was well understood and that monitoring techniques had been developed to detect and distinguish radon at low concentrations.

Other activities are described in the SEC-00135 Evaluation Report for the Met Lab, where radium research was performed in the Ryerson Physical Laboratory, Eckhart Hall, the Kent Chemical Laboratory, and the Jones Chemical Laboratory [Huntsinger et al. 1952; NIOSH 2008, PDF p. 10]. These were radiochemistry research facilities operated by the University of Chicago where work with radium was performed in hoods and ventilated areas; therefore, significant radon quantities would not have been encountered by workers on a routine basis due to the engineered controls. This would also have been the case at Site B, a complex used by the Biology Division and which included "an animal facility where research involving the use of radionuclides, including transuranic nuclides, was conducted" [Huntsinger et al. 1952, PDF p. 290]. A 1946 memo describing waste disposal at the Chicago facilities also lists a potential

source of contaminated waste as animal carcasses “injected with radium and other radioactive materials” [Fleury 1946, PDF p. 3]. Animal carcasses would not have been a significant source of radon because the only significant pathway for radon emanation is exhalation (as described above for human exposures), which would have been nonexistent in an animal carcass. Since the radium intake would have been largely concentrated in bone tissues, radon created in that tissue would not have had time to migrate through intervening tissue before decay, and permeation through skin would also have been diminished in an animal carcass. Furthermore, worker contact times with animal carcasses in unventilated conditions would have been minimal due to aesthetic and hygienic considerations.

Available records consistently indicate that work with radium at the Met Lab was conducted in laboratory settings. Radium sources were sealed or in sealed containers to prevent radon emanation [Rose 1949, PDF p. 17] and kept in non-occupied storage locations when not in use. In addition, charcoal filters (called radon traps) were used to absorb any radon emanated from sealed sources and containers [Rose 1948, 1949]. While detailed descriptions of radon traps have not been located, it should be noted that ANL-E was involved in research of solid absorbents for radon [ANL 1979a, PDF p. 91]. Room-filtered air ventilation and hoods were in place [Fleury 1946, PDF p. 3; Huntsinger et al. 1952, PDF pp. 193–196].

This historical perspective, including information about the Met Lab, is included to demonstrate that the potential for exposure to radon was recognized and controlled, even early in the operations of Met Lab, the direct predecessor of ANL-E.

### **ANL-E OPERATIONS INVOLVING RADIUM**

Radium use at ANL-E involved small (i.e., gram quantity) radium sources or radium compounds, as well as other special uses, such as dosimeter and instrument calibrations [ANL 1979a, PDF p. 246; Strom 1982, PDF pp. 12, 79, 268, 317–318] and accelerator studies [Holl et al. 1997, PDF p. 176; Lonergan and Novak 1962, PDF p. 41]. These operations were consistently described in the records as being conducted in laboratory settings, which involved the use of hoods, gloveboxes, filtered building ventilation, and routine health physics coverage, including routine air monitoring. No records have been found indicating that large-scale processing or handling of radium occurred.

### **Chemistry Division**

With the move from Met Lab to ANL-E, chemistry activities were relocated to Site D (Building 40, East Area and Building 200, 200 Area) [ANL 1955b, PDF p. 5] at the new ANL-E site. Records indicate that precautions and protective measures initiated during Met Lab operations were carried over to ANL-E facilities at Site D when operations were transferred, including sealing of radium sources, storage in non-occupied areas, design of facilities to ensure ventilation by filtered air, and work in hoods [Rose 1949, PDF p. 3; ANL 1955b, PDF pp. 5, 19].

As noted in the site profile [ORAUT 2006c], work with potential sources of radon included use of <sup>226</sup>Ra in Buildings 203 and 211 as part of the accelerator program.

As stated in the introduction, it has been pointed out that the existence of a radium ( $^{226}\text{Ra}$ ) in vitro bioassay program could indicate a potential for exposure to radon ( $^{222}\text{Rn}$ ). Although many bioassay results in the records are not identified by work location, those that can be identified are consistent with the locations identified above as using radium (e.g., D-200, D-203), and many of the results were for follow-up samples for six individuals involved in the June 1952 incident [ANL 1960a], an individual who received a skin puncture wound involving  $^{223}\text{Ra}$  contaminated forceps [ANL 1951–1960], and two individuals involved with a liquid sample spill of 300 microcuries of  $^{226}\text{Ra}$  in a fume hood [Fairman 1974, PDF p. 9]. Table 1 below contains descriptions of representative work activities or incidents involving radium from various safety reports throughout the years.

Typical numbers of radium bioassay samples were less than 10 per month or around 1% to 2% of the total number of bioassay samples processed for ANL-E, indicating that radium was not involved in most radiological work. Consequently, the existence of the radium bioassay program, as described in records, is consistent with the processes described in this document and does not suggest additional large-scale radium operations existed that would have generated significant radon quantities.

### **Biology Division**

The Biology Division (in Building 202, the Biology Building) continued research on the radium dial painters, transferred from Site B by the end of 1952 [Novak 1952, PDF p. 69], as well as biological studies on the effects of radiation on animals and plants. Numerous reports reviewed from between 1952 and 1963 describe animal injections with various radionuclides identified. Radium injections were occasionally reported in microcurie amounts [Lonergan and Novak 1959, 1961, 1962, 1963; Novak 1952, 1953, 1957b; Okolowitz and Novak 1956; ANL 1955b, 1955c, 1958, 1960b]. These reports consistently describe this work as being performed in laboratory environments with associated radiological monitoring. Potential radiological exposures were identified in monthly safety reports (typically none or minor spills). Radon exposures would not have been likely due to the small amounts of material involved, the matrix of the material, and the containers of the source material (liquid solutions in syringes).



**Table 1. Incidents or Documented Work with Radium**

Reference	Date	Location	Source	Description
Rose 1949	02/21/1949	Site A, Room A-10	Leaking 0.5 g Ra-Be source	Air counts in adjacent rooms to where the source was located were twice background levels. Source was placed in room D-6 storage area. Air counts in storage area on 02/28/1949 were 5.3 times the permissible level for radon at 1 hour and slightly more than permissible at 5 hours (radon permissible level given as $10^{-11}$ Ci/L). Source moved to ventilated area at New Chem (University of Chicago site). Storage room was tested again on 03/11/1949 after having been closed and it showed air concentrations 5x permissible level (PDF p. 22).
Rose 1949	03/14/1949	Site A, Room A-18	1 g radium source (RaBr <sub>2</sub> ) ruptured	Source discovered ruptured when opened. Accounts for high radon levels in storage area. Decontamination of Room A-18, where the source was opened, continued through 03/23/1949. Three named individuals involved in clean-up wore respirators, protective clothing, and two of the three provided follow-up bioassay for 'Ra and Rn'. Bioassay results were negative. Radon leakage from container gave surface contamination readings of "20M" (or 20,000 cpm/100 cm <sup>2</sup> ; "M" = 1,000 dpm/100 cm <sup>2</sup> ). Eventually, this source was sent back to New York on 04/04/1949 (PDF pp. 19, 20).
Rose 1949	04/12/1949	New Chem (University of Chicago site)	Leaking 500 mg radium source soldered	Individual worked in a Pb-shielded steel dry box in the hot lab. External doses were measured at various locations and personnel dosimetry (wrist, head, and special body badges) were worn and measured (PDF p. 17). Presumably, this is the same leaking source as 02/21/1949.
ANL 1979a	06/13/1952	Building 203	50 mg radium sulfate powder	PDF p.246, release of radium sulfate powder during calibration of pocket dosimeters.
ANL 1953	11/02/1953	D-202 (Room E-101)	Monitoring service provided while rats were injected with radium	Fifty rats injected with radium (total activity 800 $\mu$ Ci). Surveys were done pre and post job. Post job survey found 2M alpha contamination in on 2'x2' area on the benchtop. Contamination was cleaned with acetone. Air monitoring was taken (filter queen) during the job and showed 1 dpm/m <sup>3</sup> when decayed to 16 hours (PDF p. 7).

Reference	Date	Location	Source	Description
ANL 1953	07/07/1953	D-202 (Room B-222)	Radon leak from low level radium waste container	Radon leak from corroded cap, initial air sample result of 5,696 dpm/M <sup>3</sup> (26% of radon MPL); result of 6 dpm/ M <sup>3</sup> after 24-hour decay (PDF p. 10). Cap was replaced preventing further leaks
ANL 1954–1955	1954	D-200	Ra-223	Ra-223 incident bioassay results (see ANL [1951–1960] below)
ANL 1951–1960	1954	D-200	Ra-223	Ra-223 intake due to skin puncture with contaminated forceps (PDF p. 3); associated bioassay results
ANL 1955c	01/1955	Building 203 (various electrical substations in waist [sic] area)	Contamination from 06/13/1952 incident	Contamination from the 06/13/1952 radium spill discovered during survey; decontaminated to a fixed level of 1M to 10M from initial 54M (PDF p. 120)
ANL 1955c	12/12/1955	Building 203 (A fuse and breaker switch housing at sub-station B)	Contamination from 06/13/1952 incident	Contamination from the 06/13/1952 radium spill decontaminated from 40M to fixed levels of 1 to 5M (PDF p. 6)
Okolowitz and Novak 1956	1956	Site A Building 40 Chemistry	Radium	Site A Building 40 Chemistry - radium spill near hood in room E134 was deconned 06/27/1956 (PDF p. 4); 01/06/1956, transfer of sample to cave in hood in Special Materials (PDF p. 50)
ANL 1958	1958	Building 203	Contamination from 06/13/1952 incident	Further decontamination of electrical substations contaminated by 1952 spill (described in ANL 1955c) with no resulting personnel, equipment, floor, or airborne contamination (PDF p. 78,); residual contamination from same spill found in sump pump pit and partially deconned, with remainder fixed in place (PDF p. 181)
Lonergan and Novak 1959	1959	200 Area	Radium	Release of radium from flask in hood room F114 contaminated only hood, response in Scott Air Paks (PDF p. 24)
Lonergan and Novak 1961	02/1961	Building 203 (air supply duct in G049 and adjacent storage cage)	Contamination from 06/13/1952 incident	Surveys of 40” section cut out and to be replaced by outside contractors showed activity of 1 to 3M direct and 0.2 to 2M loose contamination from the June 13, 1952, radium spill. Reclamation personnel cleaned the duct to permissible levels before the sheet metal men were allowed to proceed (PDF p. 226).



Reference	Date	Location	Source	Description
Lonergan and Novak 1961	02/14/1961	Building 203 (G097 electronic stockroom wall)	Ra-226	Monitoring provided to contract personnel during removal the section of the wall. Survey of the wall showed spots of 1 to 5M direct and less than 0.2M smears. Only one individual involved and wore coveralls and respirator. Air sample taken during operation showed 86% MPC (MPC for 168-hour week = 22 dpm/m <sup>3</sup> ) for Ra-226 after 24 hours decay (PDF p. 226).
Lonergan and Novak 1961	08/24/1961	Building 350 (Fabrication and Technical Area room exhaust)	Radon, Thoron and daughter products detected on prefilters	Forty-eight prefilters were changed by Reclamation Services. Filters read 0-1,000 dpm/62cm <sup>2</sup> alpha and less than 0.1/1" H&S (hard and soft) beta-gamma. Contamination is due to radon and thoron and their daughter products (PDF p. 105).
Lonergan and Novak 1962	1962	Building 316	Radon source	Zero Gradient Synchrotron (ZGS) - ring coil magnet alignment in D369 (proton area) using a "radon source" (PDF p. 41), previously a radium source used.
Lonergan and Novak 1962	1962	Building 316	Radon source Radium	Spill in Building 200 room E134 (material not listed) resulted in hand/foot/floor radon contamination, spill of radium in room R066 Special Materials (PDF p. 68).
Lonergan and Novak 1963	1963	Building 200	Rn-222 Ra-226	Building 200 Chemistry, room F110 - preparing Rn-222 generator from Ra-226 sample, flask arm broke releasing 1 mCi of Rn-222, all work and recovery performed in hoods with no airborne release to laboratory (PDF p. 69).
Lonergan and Novak 1963	01/30/1963	Building 200	Radium source	Special Materials workroom routine survey found radon progeny contamination on floor, traced to leaking radium source moved to hood, work performed with assault masks (PDF p. 168-169).
Lonergan and Novak 1963	02/13/1963	D-369	Radon source	ZGS - ring coil magnet alignment of accelerator in the D369 Building using radon source, area off limits to personnel during exposure (PDF p. 157-158).
Sedlet 1972-1974	10/05/1972	Building 203	Radium	Bioassay records of all involved in 06/13/1952 radium release (described above in ANL [1979a, PDF p. 246]) were reviewed and the decision was made to recheck urine samples from involved individuals that were still employed at ANL-E (PDF p. 26).

Reference	Date	Location	Source	Description
Sedlet 1972–1974	01/29/1973	No information	Broken tube containing Rn-222	PDF p. 152: Tube containing Rn-222 broken during a chemical reaction experiment. The two occupants (one CHM, one visitor) in the area were counted in the BRC (body radioactivity counter) with small amounts of Bi-214 and Pb-214 detected. Evaluation pending per the January 1973 report.
Fairman 1974	11/05/1974	Building 203	Radium (Ra-226) chloride	A 1 milliliter, 300 $\mu$ Ci solution of radium (Ra-226) chloride being evaporated on a hot plate in a hood splattered off planchet and contaminated the immediate surroundings. Follow-up radium bioassay performed for two involved individuals.
ANL 1979a	1979	Building 200 Wing M	Radium Radon generator	1979 - Wing M work described, only work with radium is radon generator for developing solid absorbents for radon (PDF p. 91); accident history outlining 06/13/1952 radium release (PDF p. 246)

## ANL-E OPERATIONS INVOLVING THORIUM

No records have been found indicating that large-scale processing or handling of thorium occurred, except for the operations with spent fuel currently described in the ANL-E Occupational Environmental Dose TBD (ORAUT 2006b).

### Chemistry Division

As noted in the site profile [ORAUT 2006c], work with potential sources of radon included production of  $^{220}\text{Rn}$  in Building 200, handling of natural thorium in the East Area, and handling of Th-228/Th-230 in Building 211. Table 2 gives examples of thorium work discussed in various safety reports throughout the years at ANL-E. As demonstrated in the information in Table 2, planned work with thorium typically involved air and contamination monitoring, as well as use of respiratory protection when there was a potential for elevated air concentrations. Thorium incident follow-up was similar to that seen for radium incidents (i.e., use of respiratory protection for re-entry/clean-up efforts and special bioassay for individuals present at the time of incidents).

Note that because the thorium isotope is not identified in several of the records shown in Table 2, the associated radon isotope [i.e.,  $^{220}\text{Rn}$  ( $^{232}\text{Th}$  decay series) versus  $^{222}\text{Rn}$  ( $^{230}\text{Th}$  and  $^{238}\text{U}$  decay series)] cannot be determined with certainty. It is assumed that the term “thorium” would typically be natural thorium ( $^{232}\text{Th}$ ); therefore, the references are included in this section.

**Table 2. Incidents or Documented Work with Thorium**

Reference	Date	Location	Source	Description
Novak 1952	04/10/1952	D-16	Thorium, uranium	Thorium, uranium, and beryllium compound removed from Site B Special Materials vault and packed in secondary containers, relocated to D-16; protective clothing and assault masks worn at all times in the vault.
ANL 1955c	02/28/1955	D-203	Thorium	Small bottle of thorium surveyed that had been ground into fine powder outside of hood in Room G-126 with no monitoring requested and no air sampling performed; urine and fecal samples requested and recommendation made that similar work be performed in hood; fecal sample results in Robinson [1955–1956, PDF p. 36]; no urine results found, but this document only reports positive results, so any urine sample was likely 0 dpm/1,500 cc.
Novak 1956	08/25/1955	D-301	Thorium, uranium	Airborne release of uranium and thorium oxide in Cave #2.
Okolowitz and Novak 1956	01/01/1956	D-200	Thorium	Series of high air samples determined to be due to thorium crucibles stored in cabinet in lab (lab not identified).
Okolowitz and Novak 1956	06/01/1956	D-22	Thorium	Air samples exceeded [thorium] MPLs during operations using powdered thorium oxide; operations suspended 3 weeks to develop methods to address; pressing of Th-232 pellets begun 06/29/1956, air sampling performed.
Novak 1957a	12/18/1956	N/A	Thorium, uranium	Twenty-two urinalyses for thorium and uranium to be sent to A.P. Greene company.
Novak 1957a	01/16/1957	D-23	Thorium	Permission given to use thorium in Building 23 as long as monitoring performed.
Novak 1957b	10/15/1957	D-316	Thorium	Spill of 1-2 grams of 96% thorium oxide/4% enriched uranium oxide from ZPR-VII fuel tube; urine samples requested, no activity above control values.
ANL 1958	12/15/1958	D-301	Thorium, uranium	Cell B; fuel assembly (uranium-thorium) cut in isolation area, personnel in adjoining area evacuated, air sample showed no significant alpha after 21 hours.
Lonergan and Novak 1959	01/01/1959	Buildings 25, 200, 301	Thorium	Various thorium samples, fuel pellets in use.
ANL 1960b	03/31/1960	D-205	Thorium	F111; rad safety monitoring of thorium solidification run, no incidents.
ANL 1960b	09/01/1960	D-208	Thorium	Evaporating thorium on gold foils in cold trap equipped vacuum; no long-lived activity on air samples.
ANL 1960b	09/15/1960	D-205	Thorium, uranium	Spill of 6-8 liters of uranium-thorium nitrate solution; air samples below 10% maximum permissible concentration (MPC) for uranium, urine samples negative.

Reference	Date	Location	Source	Description
ANL 1960b	10/04/1960	D-16	Thorium	F area; machining of thorium rods with air monitoring; "Various radioactive materials are handled in this room including uranium of various enrichments with associated radon-thoron daughter products. Therefore, a more realistic decay time to determine the presence of long-lived activity was used. Based on the thorium B half-life of 10.6 hr. and 8 half-lives, approximately 3.5 days was used. Under these assumptions no significant amount of thorium was detected." (PDF p. 40)
ANL 1960b	12/01/1960	D-205	Thorium	Decon of furnace wearing assault masks, air samples below thorium MPC (168-hour week) of 22 dpm/ m <sup>3</sup> .
ANL 1960b	12/21/1960	D-200	Thorium	Minor thorium compound spill in Hood #4 room F102; no air or personnel contamination.
ANL 1960b	12/28/1960	D-205	Thorium	B133; spill of thorium compound (100 ml of liquid), no personnel or air contamination.
Lonergan and Novak 1961	07/09/1961	D-22	Thorium	Dismantling of thoria spray booth wearing assault masks, shoe covers, and protective clothing.
Lonergan and Novak 1961	08/08/1961	D-40	Thorium	Decontamination of glovebox where experiments with Th-228/-229/-232 were conducted; assault masks worn; air sample 12% MPC (22.2 dpm/ m <sup>3</sup> , 168-hour week) after 5 days.
Lonergan and Novak 1961	08/24/1961	Building 350 (Fabrication and Technical Area room exhaust)	Radon, thoron	Radon, thoron, and daughter products detected on room exhaust system prefilters; 48 prefilters were changed by Reclamation Services. Filters read 0-1,000 dpm/62cm <sup>2</sup> alpha and less than 0.1/1" H&S beta-gamma. Contamination is due to radon and thoron and their daughter products (PDF p. 105).
Lonergan and Novak 1962	11/07/1962	D-37 vaults	Thorium	Entry to area where kilogram quantities of thorium stored wearing half-face respirators; air samples at 25.7 and 4.0 MPC after 18 and 22 hours; both samples below 2% MPC after 5 days; Th-nat MPC (168 hours/week exposure) = 44 dpm/m <sup>3</sup> ; Pa-234 detected in sample.
Lonergan and Novak 1963	04/01/1963	D-205	Thorium, uranium	Arc-melting of uranium and thorium samples in room H125 in furnace, no room, personnel, or air contamination resulted (PDF p. 103).
Lonergan and Novak 1963	04/26/1963	D-17	Thorium	Machining of thorium rods without prior notification of Radiation Safety; hand/clothing contamination of 1,000 dpm/61 cm <sup>2</sup> resulted, which was successfully deconned; urine sample result negative (PDF p. 101).
Lonergan and Novak 1963	05/23/1963	D-17	Thorium	Machining of 1 kg thorium rod without incident (PDF p. 83).
Golchert and Kolzow 1998	1997	N/A	Rn-220	Rn-220 release, maximum perimeter dose of 0.24 millirem/year (PDF p. 29).

## ANL-E OPERATIONS INVOLVING ACTINIUM

No records have been found indicating that large-scale processing or handling of actinium occurred.

### Chemistry Division

As noted in the site profile [ORAUT 2006c], work with potential sources of radon included handling of  $^{227}\text{Ac}$  in Building 200.

With the move from Met Lab to ANL-E, chemistry activities were relocated to Site D (Building 40, East Area and Building 200, 200 Area) [ANL 1955b, PDF p. 5] at the new ANL-E site. Records indicate that precautions and protective measures in use at the Met Lab facilities were carried over to ANL-E facilities at Site D when operations were transferred, including sealing of radium sources, storage in non-occupied areas, design of facilities to ensure ventilation by filtered air, and work in hoods [Rose 1949, PDF p. 3; ANL 1955b, PDF pp. 5, 19].

During work with  $^{227}\text{Ac}$  in Building 200, which occurred between 1952 and 1958, knowledge of the potential for release of  $^{219}\text{Rn}$  (actinon) was evident in handling the materials. Samples were used in shielded gloveboxes and enclosed ventilation systems, or stored in Special Materials vaults, and were sealed in plastic during transfer, with air sampling performed [ANL 1955c, PDF pp. 107, 118]. Record of an incident involving disconnecting a glovebox from the ventilation system resulted in high concentrations of  $^{219}\text{Rn}$  measured in room air; Scott Air Paks [i.e., self-contained breathing apparatus (SCBAs) with supplied air] were worn during incident recovery operations [ANL 1958, PDF p. 179]. Table 3 gives examples of actinium work discussed in various documents and safety reports throughout the years at ANL-E.

**Table 3. Incidents or Documented Work with Actinium**

Reference	Date	Location	Source	Description
Staniforth 1950	12/20/1950	N/A	Actinium	Indicates 'possible' decision by ANL-E to allow Mound to operate a cave for actinium extraction at ANL-E, with 7 Ci available in May and June 1952 and Jan and Feb 1953.
Novak 1952	10/22/1952	D-200	Ac-227	F-110; 3 mg Ac-227 samples opened in a hood; air counts, dosimeters, and contamination surveys showed no exposures approaching permissible levels occurred.
Novak 1953	12/25/1952	D-200	Ac-227	Room F-110; accidental hood opening containing 7 mg of Ac-227 and other contaminants; air sample for short lived activity 'somewhat higher than normal'; urine samples requested (results in Sedlet [1954, PDF pp. 106–107]).

Reference	Date	Location	Source	Description
Novak 1953	01/05/1953	D-200	Ac-227	Room F-118, hood #1, removed Ac-227 contaminated equipment and waste from hood, air sample at 12.3x MPL for Pu-239; surface contamination on horizontal surfaces from 'actinium emanations'; equipment and waste sealed and removed on 01/06/1953 wearing assault masks; room deconned on 01/07 and 01/08/1953; no contamination of adjoining rooms.
Huntsinger 1953; Novak 1953	04/08/1953	D-200 (Room F-118)	Ac-227 spill	A 5-page memo detailing spill in F-118 that was spread to rooms F-110, F-122, and adjacent corridors and involved shoe and clothing contamination of a number of workers. Contamination was of a 'spot' nature, with the highest activity 12,800 M (M = 1,000 dpm/100 cm <sup>2</sup> ) recorded in Room F-118. Air samples were background except for sample in F-118, which was 135% MPL of 70 dpm/M <sup>3</sup> after 30 minutes, and 8% of MPL at 21 hours. Bioassay samples were requested from several individuals, with corresponding results in Sedlet [1954, PDF pp. 69–77]. Additional incident description in Novak [1953, PDF pp. 24–26].
ANL 1953	08/24– 08/25/1953	D-200 (Room F-102)	Elevated air samples while cleaning known Ac-227 "Berkely Box"	General cleaning of equipment and materials in the "Berkely Box" resulted in 2 air samples above the MPL of 70 dpm/m <sup>3</sup> . Box had been used for high level Ac-227 samples. Coveralls, gloves, and shoe covers worn at all times by the two workers doing the clean-up. Scott Air Packs were issued on August 25. Urine and fecal samples were requested from the two workers, as well as the Industrial Hygiene Section support person (PDF p. 2); gross alpha results for follow-up urine fecal samples for involved workers listed in Sedlet [1954, PDF pp. 21–29].
ANL 1953	10/22/1953	D-200 (Room F-110)	Removal of hood #6 and section of exhaust duct	Ac-227 in an adjacent hood attached to the same exhaust as hood #6 had 3mg of Ac-227. Rather than move source, ventilation (large elephant hose attached to glove port) at 800 lineal feet per minute was exhausted to a fume hood on a separate system. Assault masks and protective clothing worn by personnel and air samples taken during the entire operation. Air samples showed less than 10% MPL after 4-day decay. Post job survey showed no new contamination.



Reference	Date	Location	Source	Description
ANL 1953	11/27/1953 and 11/30/1952	D-200 (Room F-110)	Monitoring service provided during unpackaging of Ac-227	Monitoring service provided during unpackaging of two high-level Ac-227 samples (99 mc and 200 mc). Unpackaging was performed in a Lucite box in glove paneled Blickman hood. Protective clothing including leaded gloves, as well as personnel monitoring devices, were worn by workers. Air samples taken during the operation indicated no activity above normal room background levels.
ANL 1954– 1955	1954	D-200	Ra-223	Ra-223 incident bioassay results (see ANL [1951–1960] below).
ANL 1951– 1960	1954	D-200	Ra-223	Ra-223 intake due to skin puncture with contaminated forceps (PDF p. 3); associated bioassay results.
ANL 1955c	01/06/1955	D-200	Ac-227	F-118; transfer, plating, and preparation of Ac-227 samples in shielded glovebox; hoods and gloveboxes taped prior to electrical shutdown, air sampling showed no elevated levels; constant alpha air monitor installed in F-118 on 01/25/1955.
ANL 1955c	02/02/1955	D-200	Ac-227	F-118; eight 0.3 mg electrodes removed from shielded glove box and relocated to spectroscopy laboratory (K-127); sealed in plastic pouches before transfer; samples mounted and arced in enclosed ventilation system, then resealed in plastic pouches and transferred to special materials vault for storage.
ANL 1955c	03/08/1955	D-200	Actinium powder	F-118; Removal of northwest hood used mainly for dry actinium powder work, subsequent cleaning and decon; contamination levels exceeding 7,000M (M=1,000dpm/100cm <sup>2</sup> ); operation monitored and conducted in Scott Air Pak without incident.
ANL 1955c	06/27/1955	D-200	Ac-227	F-118; Removal, preparation, and plating, of an Ac-227 sample; air sampling and frequent surveys of hands, clothing, and floor conducted with no contamination found during operation.
ANL 1955c	07/07/1955	D-200	Ac-227	F-118; Several Ac-227 samples moved from a hood to a small cave without incident.

Reference	Date	Location	Source	Description
ANL 1958	01/15/1958	D-200	Ac-227	Room F-118; release of Ac-227 during removal of glovebox containing ~2 mg; after disconnection from ventilation system, extremely high concentrations of Rn-219 detected in room air; room sealed, no activity in adjoining rooms or corridor; glovebox wrapped in poly sheeting, placed in plywood container, and moved to isolation room of F-118, where it was determined no radioactive gas was escaping; box sent to 317 for disposal; air sample indicated "4.7x OPL" (assumed to have been a typographical error, intending to stand for maximum permissible level, or "MPL") for Ac-227 after 8 days decay; Scott Air Paks worn, room reopened 01/28/1958.

### INCIDENTS OR DOCUMENTED WORK INVOLVING RADIUM, THORIUM, OR ACTINIUM

A number of incidents resulting in radon releases or potential exposures have been located in ANL-E records, as noted in the tables above. While this is not an exhaustive list, it gives a good idea of the types of incidents and planned unusual work (with health physics support) that occurred at ANL-E, as well as any follow-up conducted as a result of it. Most of this information is coming from searches of monthly Radiation Safety, Radiological Physics, or Industrial Hygiene reports. These reports typically provide names of ANL-E employees that were involved, the survey results, as well as bioassay monitoring that was completed, but not necessarily bioassay results, as these reports would have been completed at a later time after investigations and follow-up. A number of characteristics are common to these incidents and the responses to them.

First, none of the incidents represented long-term, unidentified exposures to personnel. While some releases were known immediately, others were identified by routine surveys. Prompt corrective actions were taken, using protective clothing and respiratory protection for more significant releases.

The incidents also did not represent large potential source terms for radon progeny exposure. Although the quantities of radium were large enough to trigger the use of radiation protection measures because of the potential for internal and external exposure, respiratory protection measures in response to the immediate recovery activities [e.g., use of assault masks (similar to half- or full-face respirators) and Scott Air Paks, relocation of source to hoods] would have also protected against associated short-term radon progeny concentrations that might have occurred during initial response.

After incident response, removal of the released material (in the case of a spill) may have resulted in residual surface contamination. The most significant incident noted in the records was a release involving approximately 50 milligrams of radium sulfate powder on June 13, 1952, in

Building 203 [ANL 1979a, PDF pp. 246–247], which contaminated much of the building. Decontamination efforts were extensive and residual surface contamination remained for several years. Typical surface contamination levels measured on the interior surfaces of ductwork, a fuse and breaker switch housing, and electrical substations ranged from 1,000 to 40,000 dpm/100cm<sup>2</sup> after initial decontamination, which correspond to microcurie levels of activity per square meter of surface. These levels do not represent potential sources of significant airborne radon activities, especially on the interior of enclosed structures. [ANL 1955c, PDF pp. 6, 120; ANL 1958, PDF pp. 78, 181; Lonergan and Novak 1961, PDF p. 226].

Despite the activities discussed above, a 1982 Environmental Report describing the primary functions of Wing M of Building 200 makes no mention of radium use [DOE 1982, PDF p. 31], which is consistent with the infrequent use and small quantities described in the documents above. Additionally, in a 1981 Dosimetry Records and Radiation Hazards Questionnaire sponsored by the U.S. Department of Energy (DOE), “radon & daughters” is the only specifically *excluded* selection by ANL-E under the question regarding “types of internal exposures that may have occurred at your facility” [Dolecek 1981, PDF p. 15]; all other internal exposure types were included as applicable to ANL-E (these were fission products, tritium, iodine, activation products, uranium, plutonium, other transuranics, other beta-gamma emitters, and other alpha emitters). The conclusion that direct personnel monitoring was not necessary for exposure to radon would be consistent with this information.

### AIR SAMPLING AT ANL-E

In addition, air samples were routinely collected and analyzed to detect elevated concentrations of radioactive materials, primarily uranium and plutonium (see Table 4). Evaluation of the records in Table 4 indicates that general area air sampling supporting worker activities was performed on a daily basis for either the entire day or for the period during which the covered activity was being performed. Samples specifically for <sup>222</sup>Rn associated with the A5 stack in Building D-200 were noted in ANL [1967]; these samples were collected in unoccupied areas and were of shorter duration (an hour or less during vented releases) than those supporting worker activities. These samples provided information relevant to offsite releases reported in ANL-E environmental reports and are accounted for in the assigned occupational environmental doses [ORAUT 2006b].

Air samples were typically analyzed for alpha and beta activity. Samples would have been promptly counted after collection, then counted at later times to determine long-lived radionuclide activity following decay of radon progeny. Radon progeny could have been from natural sources or from occupational sources, depending on the location of the air sampler and the work activities being monitored. Because radiation protection personnel were familiar with typical results for ambient (non-occupational) radon interference, initial count values exceeding normal levels allowed identification of elevated (higher than ambient) radon levels due to occupational sources. Evaluation of ANL-E air sampling records indicated that initial counts of samples intended to monitor long-lived alpha emitting radionuclides without potential for radon exposure were typically on the order of tens to hundreds of dpm/m<sup>3</sup> with subsequent counts lower (indicating the presence of non-occupational ambient radon progeny interference in the

initial count). Samples in areas with identified radon sources were on the order of hundreds to several thousand dpm/m<sup>3</sup> and would have been compared with the maximum permissible level (MPL) for radon (1E-11 Ci/L or 10 pCi/L); this MPL was established for radon as early as 1949 [Rose 1949, PDF p. 22]. As seen in the incidents discovered by elevated radon air concentrations in areas of known radon sources (discussed in the tables above), ANL-E follow-up typically involved isolating the source of the radon and clean-up of the areas, which would have been performed in respiratory protection.

Available records indicate air sampling and site-specific analysis of results (i.e., knowledge of typical ambient radon levels) were used on multiple occasions to identify and isolate radon leaks from radium sources in unoccupied storage locations [Rose 1949, PDF pp. 19, 22, Site A; ANL 1953, PDF p. 10, Site D; Lonergan and Novak 1963, PDF pp. 168–169, Site D, Building 200].

**Table 4. Air Sampling Locations.**

Reference	Date of Sampling	Description
ANL 1946	1946	Site B (Chicago)
ANL 1947–1948	1947–48	Site A
ANL 1950	1950	Unknown, one page
ANL 1952	1952	D-300, 301, 310, 340
ANL 1953–1954	1953–54	Site A; West Stands; D-306 and 317
ANL 1954a	1954	D-202
ANL 1954b	1954	D-301; D-306; D-310
ANL 1954c	1954	West Stands; results in %MPL only
ANL 1955a	1955	D-301
ANL 1955–1956	1955–56	D-306
ANL 1956a	1956	D-202
ANL 1956b	1956	Site A
ANL 1956c	1956	D-200
ANL 1956d	1956	D-205
ANL 1956e	1956	D-202, D-310
ANL 1967	1967	Various 200, 300, East Area locations; “Stack Monitor Data Sheet: Radon Monitor Channel 1” (PDF pp. 504–508) (some illegible); Rn-222 air sample result (PDF p. 416), (after release from Cell A-5) (PDF pp. 484, 492, 509)
ANL 1968	1968	D-350, New Brunswick Laboratory (NBL)
ANL 1972	1972	D-350
ANL 1973	1973	D-350, NBL
ANL 1974	1974	D-350, NBL
ANL 1975	1975	D-350, NBL
ANL 1976	1976	D-350, NBL

Reference	Date of Sampling	Description
ANL 1977	1977	Various locations
ANL 1978a	1978	Various locations
ANL 1978b	1978	Various locations
ANL 1979b	1979	Various locations
ANL 1979c	1979	D-350

### **OTHER DOE RADON REFERENCES**

A DOE study performed in 1989 to 1990 and summarized in the report “Results of the U.S. Department of Energy Indoor Radon Study” [DOE 1990], evaluated radon levels throughout the DOE complex. During this study, radon concentrations were measured at 138 locations at the ANL-E site, 4 of which exceeded the action level of 4 pCi/L. These buildings and their associated values were:

14.3 pCi/L - Building 576 (office, 500 area electrical/water distribution buildings)

8.3 pCi/L - Building 212 (Alpha Gamma Hot Cell Facility, HVEM Tandem Accelerator, room F2)

6.5 pCi/L - Building 25 (Argonaut demonstration reactor, room A-110)

4.3 pCi/L - Building 33 (East area surplus facilities, room 108)

None of these buildings are associated with radium operations or processes anticipated to release radon; consequently, these results are considered to be due to environmental rather than occupational sources of radon. The Indoor Radon Study report further notes that the expected probability of any particular building exceeding the 4 pCi/L action level was 7% based on the Nero distribution [Nero, et. al. 1984]; the sitewide value was less than 3% by comparison.

An additional DOE report [Strom and Reif 1996] identified ANL-E as one of 12 DOE facilities and projects having elevated radon levels in a DOE report for year 1991 compliance with National Emissions Standards for Hazardous Air Pollutants (NESHAPs). The NESHAPs compliance report [DOE 1992] references a release of 2900 Ci of <sup>222</sup>Rn during calendar year 1991, projected to result in a maximum offsite dose of 0.29 mrem/yr. These values were obtained from the ANL-E site environmental report [Golchert et al. 1992] and are attributed to environmental releases from the proof of breeding program. Exposures from these releases are discussed and doses assigned in the ANL-E Occupational Environmental Dose TBD [ORAUT 2006b].

### **CONCLUSIONS**

There is no large source term for radon, thoron, or actinon identified at ANL-E; no bulk materials, such as ores, were identified. ANL-E did, however, use millicurie or milligram

amounts of the radium, thorium, and actinium (i.e., primary source material), as demonstrated in the records listed above. These records consistently demonstrate that ANL-E used these materials in controlled laboratory environments (i.e., filtered air flow for building ventilation and work performed in hoods). Measures to control internal exposures to the primary source materials reduced the potential for radon progeny exposures as well, even though radon exposure was not typically the primary emphasis of monitoring and protection measures. Records also demonstrate that during interruptions of ventilation air flow, laboratories would have been sealed and recovery operations would have involved respiratory protection measures for workers. Surface contamination, laboratory sample quantities, and radium sources in sealed containers in unoccupied storage areas would not have provided a significant source of airborne radon. It is further evident that ANL-E was aware of radon as a potential for exposure from the beginning of operation and did not identify it as a significant internal dose pathway.

In response to Observation 4 in SC&A [2009], that there is a potential for exposure to radon ( $^{222}\text{Rn}$ ), actinon ( $^{219}\text{Rn}$ ), and thoron ( $^{220}\text{Rn}$ ), which is not thoroughly discussed in the Site Profile, NIOSH attests that routine exposures to radon from the presence of source materials would not have resulted in significant exposures because:

- engineered measures to control exposure to long-lived radionuclides also prevented exposure to radon,
- incidents where significant exposure was possible were readily identified by routine monitoring and mitigated, and
- response to identified incidents were conducted using worker protective equipment, restricted access, and isolation of sources.

To address the observation, additional exposure assessment explanation based on the information in this document will be added to ANL-E Occupational Internal Dose TBD [ORAUT 2006a] in order to provide additional background information and justification for the current dose reconstruction methodology.

It should be noted, as is the case for the current dose reconstruction process, claims received for individuals involved in known incidents involving elevated radon levels, such as those discussed above, will be reviewed on a case-by-case basis to determine appropriate assignment of dose.



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