PNNL-10855 Rev. 6

Pacific Northwest NATIONAL LABORATORY

Determining Unabated Airborne Radionuclide Emissions Monitoring Requirements Using Inventory-Based Methods

December 2018

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U.S. DEPARTMENT OF ENERGY

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Abstract

Compliance with the airborne radionuclide emission monitoring requirements in the National Emission Standards for Hazardous Air Pollutants (NESHAP; Title 40 of the U.S. Code of Federal Regulations Part 61, Subpart H) and State requirements in Washington Administrative Code 246–247: Radiation Protection – Air Emissions and 173-480: Ambient Air Quality Standards and Emission Limits for Radionuclides were evaluated for Pacific Northwest National Laboratory (PNNL) operations. Additional guidance may be found in the U.S. Department of Energy Handbook, *Environmental Radiological Effluent Monitoring and Environmental Surveillance*. To meet regulatory requirements, reviews of planned or proposed projects and activities provide the basis for implementing necessary monitoring adjustments or for implementing changes to projects and activities in a timely manner. Potential unabated offsite doses were evaluated for emission locations managed by PNNL and licensed to the Department of Energy. These locations were at facilities in Richland, Washington (i.e., the Hanford Site and PNNL Richland Campus) and in Sequim, Washington, (PNNL Marine Sciences Laboratory). This report describes the inventory-based methods and provides the results for the NESHAP assessment performed in 2017.

Summary

Compliance with the airborne radionuclide emission monitoring requirements in National Emission Standards for Hazardous Air Pollutants (NESHAP; Title 40 of the U.S. Code of Federal Regulations Part 61, Subpart H) and State requirements in Washington Administrative Code 246–247: Radiation Protection – Air Emissions and 173-480 Ambient Air Quality Standards and Emission Limits for Radionuclides were evaluated for Pacific Northwest National Laboratory (PNNL) operations. Potential unabated offsite doses were evaluated for emission locations at buildings that are located on the PNNL Richland Campus and the PNNL Marine Sciences Laboratory operated by Battelle for the U.S. Department of Energy (DOE) and at Hanford Site buildings operated by PNNL. Five PNNL managed buildings have emission units that meet state and federal criteria for continuous sampling of airborne radionuclide emissions:

1. Hanford Site – 325 Building, Radiochemical Processing Laboratory
2. Hanford Site – 331 Building, Life Sciences Laboratory I
3. PNNL Richland Campus – 3410 Building, Materials Science and Technology Laboratory
4. PNNL Richland Campus – 3420 Building, Radiation Detection Laboratory
5. PNNL Richland Campus – 3430 Building, Ultra-Trace Analysis Laboratory.

The NESHAP assessments were performed using building radionuclide inventory data obtained in 2017. Several Hanford Site emission units, evaluated in the prior revision of this document considered for 2010 radionuclide inventories, are no longer relevant because they were vacated or ownership was transferred to another DOE sub-contractor. The buildings evaluated in 2017 included:

- Buildings with one or more emission points; filtered and unfiltered emission pathways are evaluated.
- Buildings that do not currently contain radioactive material but are in PNNL's Radioactive Material Tracking System because they have had or may have radioactive material.

Note that PNNL is not the sole occupant of some locations where PNNL tracks radioactive material; nevertheless, only the PNNL unabated emissions are evaluated against the PNNL compliance limits at such locations. Comprehensive results of these facility locations, footnoted below, may not be included in this document.

Emission units not associated with a building are not included herein. For example, fugitive emissions released under the volumetrically release radioactive material permit may occur anywhere on the PNNL Richland Campus and are not restricted to a single building emission unit location.

- Hanford Site Buildings
  - 200E Prototype Surface Barrier Storage
  - 318 Radiological Calibrations Laboratory
  - 325RPL Radiochemical Processing Laboratory
  - 331 Life Sciences Laboratory I[1]

[1] In addition to the building filtered emission point EP-331-01-V, potential unabated emissions were calculated separately for the minor emission unit EP-331-09-S.

- 361 Modular Equipment Shelter (National Nuclear Security Administration)
  - HAMMER Hazardous Materials Management and Emergency Response Training Center[2]
- PNNL Richland Campus Buildings
  - 2400 Stevens Office Building
  - 3020 Environmental Molecular Sciences Laboratory
  - 3310 Biological Sciences Facility
  - 3410 Materials Science and Technology Laboratory
  - 3420 Radiation Detection Laboratory[3]
  - 3425 Ultra Low Background Counting Laboratory
  - 3430 Ultra-Trace Analysis Laboratory[4]
  - 3440 Large Detector Laboratory
  - 3850 General Purpose Chemistry Laboratory
  - AML Atmospheric Measurements Laboratory
  - EDL Engineering Development Laboratory[5]
  - LSL-II Life Sciences Laboratory II
  - RTL-520 Research Technology Laboratory[6]
  - RTL-530 Research Technology Laboratory Support Building[7]
  - ESB Engineering Support Building[8]
  - PSL Physical Sciences Laboratory[9]
- PNNL Marine Sciences Laboratory DOE-Owned Buildings
  - MSL1 Marine Sciences Laboratory-1
  - MSL5 Marine Sciences Laboratory-5
- Privately Owned Buildings Located in Richland, Washington
  - 3350 LSB Laboratory Support Building[10]

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[2] PNNL tracks radioactive material but is not the sole occupant of the building.
[3] In addition to the EP-3420-01-S filtered exhaust, there is also the EP-3420-02-S unfiltered exhaust.
[4] In addition to the EP-3430-01-S filtered exhaust, there is also the EP-3430-02-S unfiltered exhaust and the five registered unfiltered perchloric acid hood emission units EP-3430-xxxxP-S.
[5] Building had no radioactive material inventory at the time of the 2017 NESHAP assessment but is included here because it is listed in the Radioactive Material Tracking System (RMTS) and has had or may have radioactive material.
[6] Building demolished/removed in 2018.
[7] Building demolished/removed in 2018.
[8] Building had no radioactive material inventory at the time of the 2017 NESHAP assessment but is included here because it is listed in the RMTS and has had or may have radioactive material.
[9] Building had no radioactive material inventory at the time of the 2017 NESHAP assessment but is included here because it is listed in the RMTS and has had or may have radioactive material.
[10] Building had no radioactive material inventory at the time of the 2017 NESHAP assessment but is included here because it is listed in the RMTS and has had or may have radioactive material.

Acronyms and Abbreviations

ANSI	American National Standards Institute
CAP88-PC	Clean Air Act Assessment Package – 1988 for Personal Computers
CFR	U.S. Code of Federal Regulations
Ci	curie
DOE	U.S. Department of Energy
EM	Effluent Management Group
EMSL	Environmental Molecular Sciences Laboratory
EPA	U.S. Environmental Protection Agency
HPS	Health Physics Society
LMF	location modification factor
LSL	Life Sciences Laboratory
MA	maximum air (location)
MPR	maximum public receptor
mrem	millirem
MSL	Marine Sciences Laboratory
NESHAP	National Emission Standards for Hazardous Air Pollutants
PCM	periodic confirmatory measurement
PIC	potential impact category
PNNL	Pacific Northwest National Laboratory
PSL	Physical Sciences Laboratory
PTE	potential-to-emit
RMT	Radioactive Material Tracking (System)
RTL	Research Technology Laboratory
WAC	Washington Administrative Code
WDOH	Washington Department of Health
yr	year

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Tables

1.0 Introduction/Project Description

Radionuclide emission locations at the Pacific Northwest National Laboratory (PNNL) Richland Campus and the PNNL Marine Sciences Laboratory (MSL) are licensed by the State of Washington Department of Health (WDOH) to the U.S. Department of Energy (DOE) Office of Science – Pacific Northwest Site Office. Radionuclide emission locations on the Hanford Site are licensed by WDOH to the DOE, Richland Operations Office.

Requirements for sampling airborne radionuclide emissions are contained in the following regulations and guidelines:

- U.S. Code of Federal Regulations, Title 40, Part 61 (40 CFR 61) Subpart H: National Emission Standards for Emissions of Radionuclides Other than Radon from Department of Energy Facilities (2002)
- Washington Administrative Code (WAC) 246-247: Radiation Protection – Air Emissions (WAC 2018)
- WAC 173-480: Ambient Air Quality Standards and Emission Limits for Radionuclides (WAC 2007)
- DOE-HDBK-1216-2015, *Environmental Radiological Effluent Monitoring and Environmental Surveillance* (DOE 2015).

The first two regulations require the performance of continuous sampling at emission points that have a potential-to-emit (PTE)[11] ≥ 0.1 mrem/yr offsite maximum public receptor (MPR) dose[12] if routine emissions were not mitigated by engineered pollution control systems. The third regulation requires a dose estimate at the offsite Maximum Air (MA) location, where the emissions of radionuclides are limited to a maximum dose of 10 mrem/yr to the whole body to any member of the public and presumes an individual is at that offsite location fulltime. The MA estimates are conducted for emissions at the PNNL[13] Richland Campus and the MSL. The fourth document provides guidance on monitoring and sampling radioactive airborne effluent streams.

In response to and in compliance with the listed requirements, the potential unmitigated offsite MPR dose from the PNNL-operated buildings that contain radioactive materials or sources is evaluated annually in a National Emission Standards for Hazardous Air Pollutants (NESHAP) Assessment. The MA dose is reported in the annual compliance reporting (e.g., Snyder et al. 2018) and generally is equal to or not much larger than the MPR dose.

The radionuclide NESHAP assessment is completed annually. The number and status of buildings evaluated has changed as buildings are transitioned to other contractors, new buildings are added, or laboratory missions change. The NESHAP assessments include all PNNL buildings that house radioactive materials. This document describes the methodology used and reports summary results for those PNNL buildings. A summary of most recent five assessments' references are provided in Table 1.

[11] PTE is defined as the rate of release of radionuclides from an emission unit based on the actual or potential discharge of the effluent stream that would result if all abatement control equipment did not exist, but operations are otherwise normal.
[12] Dose is reported in mrem, rather than millisieverts, to be consistent with the reported NESHAP standard units.
[13] PNNL is operated by Battelle for DOE under Contract DE-AC05-76RL01830.

Table 1. References for Prior Published NESHAP Assessments

Radionuclide Inventory Year	Document
2013	Memo (no number)
2014	Memo (no number)
2015	Memo (no number)
2016	Memo No. EHSS-EPRP-16-026
2017	Memo No. EHSS-EPRP-18-001 Memo No. EHSS-EPRP-18-002[a]

[a] Summary of areas not included in NESHAP assessment.

2.0 Assessment Methodology

This chapter describes the methods used by PNNL to determine the potential emissions of radioactive materials from building operations subcontracted to Battelle (see footnote 13 for contract information).

2.1 Projections of Annual Emission Quantities

Several methods for projecting potential unmitigated annual emission quantities are prescribed in WAC 246-247:

- Apply an annual release fraction to the radionuclide inventory in the building
- Multiply actual measured annual emissions by control system decontamination factors
- Add actual measured annual emission quantities to actual measured quantities retained by control systems
- Measure the annual discharge upstream from all control devices.

The inventory-based assessment method[14] has been used by PNNL since the initial building assessment in 1991. The inventory method yields an assessment based on the current building status (or even the future status if projected future inventory quantities are used in the assessment), while the other prescribed methods yield an assessment based on past building measurements. The inventory method may be more appropriate for use at research and development facilities where types and quantities of radionuclides may change and where historical sampling data may not be a reliable predictor of future emissions.

Since 2006, PNNL has maintained radioactive inventory information using the Radioactive Material Tracking (RMT) System,[15] a web-based software tool that supports real-time tracking of radioactive materials. The RMT System provides a process that is used to achieve compliance with numerous regulatory requirements pertaining to radioactive material management. Appendix A provides a summary of the database features that apply to radioactive air emissions. RMT System is updated by radioactive material custodians as well as other users with authorized access to radioactive material. The software allows the approved user to add, move, modify, and ship radioactive materials in and out of PNNL buildings and verifies that building radioactive material inventory limits have not been exceeded with any inventory update. RMT System also has extensive search and report capabilities. For buildings with multiple emission points, RMT System has the capability to verify compliance with sub-building zones. This capability has not yet been employed for MSL and for some spaces ascertained to have unfiltered emission units in FY 2017 (e.g., 3430 individual perchloric acid hoods). In these cases, calculations were performed in spreadsheets using data downloaded from RMT System and from other sources to estimate emissions, and reports were manually generated outside of RMT System.

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[14] This method is described in WAC 246-247-030 (21.a) as follows: “Multiply the annual possession quantity of each radionuclide by the release fraction for that radionuclide, depending on its physical state. Use the following release fractions: 1) 1 for gases, 2) 10^{-3} for liquids or particulate solids, and 3) 10^{-6} for solids. Determine the physical state for each radionuclide by considering its chemical form and the highest temperature to which it is subjected. Use a release fraction of 1 if the radionuclide is subjected to temperatures at or above its boiling point; use a release fraction of 10^{-3} if the radionuclide is subjected to temperatures at or above its melting point, but below its boiling point. If the chemical form is not known, use a release fraction of 1 for any radionuclide that is heated to a temperature of 100°C or more, boils at a temperature of 100°C or less, or is intentionally dispersed into the environment....”

[15] The RMT System is not fully deployed at the MSL because MSL has a small number of radionuclides with unit doses unique to that site. An annual inventory-based assessment is performed at MSL similar to that described here but with site-specific calculations.

Radioactive inventory as well as throughput and proposed radioactive additions in the upcoming year are evaluated annually (PNNL 2017). The evaluation includes the RMT System Administrator contacting RMT System custodians, contacting Environmental Compliance Representatives, reviewing outstanding radioactive material requests, and proposing nuclear material additions for updates and inputs. The RMT custodians verify the RMT inventory for material under their scope of responsibility. The custodians enter and manage inventory that represents current materials in the building and items on order or expected to be used in a future year (e.g., throughput items). Environmental Compliance Representatives provide information from Electronic Prep and Risk reviews about new projects involving radioactive materials. The RMT System Administrator enters and maintains data representing additional radioactive materials that may be processed in the coming year, throughput for normal operations,[16] anticipated new work, and emission of gases.

Radioactive gas emissions (e.g., Kr-85) are managed separately in the Radioactive Air Gas Inventory Database. Data from the database are used as a resource to populate expected gas emissions for the coming year. The RMT Administrator assigns a radioactive material custodian as appropriate for the additional information, and the custodian reviews and verifies the complete set of data under his/her name. The RMT Administrator generates NESHAP reports for each building that list RMT inventory items, summarizes the total PTE, and identifies the percent of each isotope in the inventory contributing to the PTE. As mentioned previously, some NESHAP reports were generated outside of RMT in 2010 for individual emission units that are a sub-component of total building emissions. Also, several new buildings known collectively as the Physical Sciences Facility were completed in 2010, and radioactive material was being moved into these buildings (i.e., the 3410, 3420, 3425, and 3430 Buildings) at the time of the 2010 assessment. Because these buildings were in transition, RMT data were downloaded and supplemented with additional information on inventory to be transferred or added to the building in the coming year.

Radionuclides meeting either of the following criteria are excluded from the assessments:

- Radionuclides present in commercially available building/construction materials
- Radionuclides that can be purchased or possessed without a special radioactive materials license.

The data are reviewed and revised as needed to eliminate duplicate information and to obtain additional information as necessary. The review process is then documented and filed with the Effluent Management (EM) Group File Plan.

Potential release fractions for radionuclides are based on the physical form of the radionuclide as shown in Table 2. Radionuclides present as sealed sources or in sealed, unvented U.S. Department of Transportation-compliant shipping containers are assumed to be unavailable for release under normal conditions.

[16] A recent update to RMT calculates throughput for the previous year and provides a field for custodians to record items on order and any expected changes in throughput for the future year. This update went into effect just after the 2010 NESHAP assessment.

Table 2. Physical Forms and Potential Annual Release Fractions for Radionuclides

Form	Description	Potential Release Fraction
Gas[a]	Radioactive material in a gaseous or vapor form. This includes solids or liquids heated to a high enough temperature to be in a volatized state or are intentionally dispersed into the environment.	1
Liquid*	Radioactive material is a liquid, solution, or slurry, and its primary container will be opened at some point during the calendar year.	10^{-3}
Particulate solid*	Radioactive material will be present in powder form.	10^{-3}
Non-particulate Solid*	Radioactive material is a monolithic solid or consists of relatively large chunks.	10^{-6}
Non-dispersible Radioactive Material	Radioactive material in a leak-proof rigid container (including waste drums and boxes) that is not opened or is not planned to be opened through the calendar year. Also includes radioactive material determined to be non-dispersible in accordance with Radiation Control Program Procedure 3.1.01.	0
Sealed source: any type	Radioactive source manufactured, obtained, or retained for the purpose of using the emitted radiation. The sealed radioactive source consists of a known or estimated quantity of radioactive material contained within a sealed capsule, sealed between layer(s) of non-radioactive material, or firmly fixed to a non-radioactive surface by electroplating or other means intended to prevent leakage or escape of the radioactive material.	0

[a] Based on Table 1 of American National Standards Institute/Health Physics Society N13.1-1999 (HPS 2011).

2.2 Unit Dose Calculation

For unit dose calculations, the offsite MPR is defined as an individual whose residence location or work location maximize the potential dose from airborne pathways. Models assume the receptor occupies the location 24/7 and acquires food grown at the location. All potential environmental transport pathways associated with an airborne radionuclide release were included (i.e., air inhalation, air submersion, exposure to deposited radionuclides, and food ingestion). This is a prospective dose based on estimated potential emissions from emission units and is used in determining emission monitoring requirements as part of the permitting process.

Unit dose factors for the Richland area MPR were calculated for specific radionuclides using the U.S. Environmental Protection Agency (EPA) compliance code (Clean Air Act Assessment Package – 1988 for Personal Computers [CAP88-PC]; Chaki and Parks 2000; Rosnick 2007; EPA 2015). Unit dose factors for the Sequim area MPR were calculated using the EPA COMPLY code (EPA 2017). Radionuclides that were not represented in COMPLY were conservatively assigned default values, usually equal to that of ^{241}Am for alpha emitters; or the greater of either ^{137}Cs or ^{90}Sr for non-alpha emitters or as indicated in the unit dose factor documentation. Daughter product decay also was considered in assigning default values for short half-life radionuclides. The most recently published unit release dose factors were calculated using CAP88-PC Version 4.0 for the Hanford Site (Snyder and Rokkan 2016) and for the PNNL Site (Snyder and Barnett 2016). Older references for Hanford Site and PNNL Richland Campus dose factors (Rhoads et al. 2010, Rhoads and Barnett 2009, respectively) were calculated using CAP88-PC Version 3 (Rosnick 2007). These documents describe the methods and assumptions used and provide unit dose factors for the PNNL emission units in the Hanford Site 300 Area and PNNL Richland Campus. Unit dose release factors for MSL are indicated in Appendix B of the annual air emissions reports (e.g., Snyder and Barnett 2018).

For the Physical Sciences Facility, dose assessments were performed using unit dose factors for the PNNL Richland Campus. For buildings not on the Hanford or PNNL sites (i.e., located elsewhere in Richland, Washington), dose assessment were performed by applying a location modification factor (LMF) to the unit dose factor to correct for varying source-receptor distances and directions. The LMF was calculated by dividing the atmospheric dispersion value (Chi/Q) for the building by the atmospheric dispersion value for the assumed MPR location when the same meteorology and receptor assumptions are applied. The compliance code CAP88-PC was used to calculate these dispersion values.

2.3 Potential Emission Dose Assessment

Desk instruction DI-AIR-001, Conducting and Documenting the Annual NESHAPS Assessment, describes the process for generating the annual Rad NESHAP reports (PNNL 2017). The RMT System is used to track radioactive material inventory at PNNL and generates NESHAP reports for the annual assessment. The annual NESHAP report, derived from this assessment, is based largely on two reports that are created by and exported from the RMT System. The first report is the “Dose Contribution Report (Cumulative).” It provides, for all PNNL buildings, the PTE dose for each radionuclide that has been in the building during the current reporting year and the contribution of each radionuclide to the annual PTE calculated by the RMT System for that building. The second report is referred to as the “FEMP Data Evaluation Report (Cumulative).” It provides a listing of RMT items that have been in each building throughout the year and the data used to calculate the PTE for the item (including each item’s activity by radionuclide, physical form, release fraction, radionuclide- and location-specific PTE dose conversion factor, and PTE value for each item by radionuclide) (PNNL 2017). Assessment results and reports are maintained as records in the EM Group File Plan.

3.0 Reports

As indicated in Appendix B, two reports are generated from the RMT System for the NESHAP assessment for inclusion in the final packet:

- A report of individual items provided by the RMT System, including current inventory and additional inventory expected to be processed in the coming year or brought in as part of new work. This includes potential gas emissions.
- A complete listing of each radioisotope present in the building, the associated dose contribution (mrem), and the percent of the total dose for the building.

Potential unabated doses are assessed for the MPR for each PNNL registered emission unit. Facilities with only sealed sources in 2017 (no licensed emission units) are listed in Table 3. Assessment results for CY 2017 are provided in Table 4.

Table 3. Exempt[a] Facilities with Sealed Sources Only, 2017

Required Emission Monitoring Method	Building	System Description	2017 Potential Offsite Dose, mrem/yr	Comment
	Systems Located on the Hanford Site			
None	200E Storage	Prototype Surface Barrier Storage	0	Sealed sources
	Systems Located on the PNNL Richland Campus			
None	EMSL (3020)	Environmental Molecular Sciences Laboratory	0	Sealed sources
None	3440	Large Detector Laboratory	0	Sealed sources
	Systems Located Elsewhere in Richland, Washington			
None	2400 Stevens	2400 Stevens Office Building	0	Sealed sources
None	AML	Atmospheric Measurements Laboratory	0	Sealed sources
None	BSF	Biological Sciences Facility	0	Sealed sources
None	EDL	Engineering Development Laboratory	0	Sealed sources
None	ESB	Engineering Support Building	0	Sealed sources
None	LSB	Laboratory Support Building	0	Sealed sources; no radioactive inventory at time of 2017 assessment
None	PSL	Physical Sciences Laboratory	0	Sealed sources; no radioactive inventory at time of 2017 assessment
	Systems Located at the MSL in Sequim, Washington			
		N/A		

(a) Exempt as a registered emission unit because the facility contains only sealed sources; however, for the PNNL Richland Campus Potential Impact Category-5 authorized sources (e.g., Volumetrically Released Radioactive Material, Non-Dispersible Radioactive Material) may be authorized at these facilities.

Table 4. Registered Emission Unit Potential Dose Assessment for Unabated Emissions, 2017

Required Emission Monitoring Method per WDOH License[(a)]	Emission Unit	Emission Type[(b)]	System Description	2017 Assessment Dose, mrem/yr	Comment
	Systems Located on the Hanford Site				
Inventory	J-318	Fugitive	Radiological Calibration Laboratory	3.68E-05	Primarily sealed and check sources
Cont. Monitoring	EP-325-01-S	Point	325-Radiochemical Processing Laboratory	5.00E+01	
Cont. Monitoring	331-01-V	Point	Life Sciences Laboratory I	1.02E-02	
Inventory	331-09-S	Fugitive	Life Sciences Laboratory I	7.89E-06	
Inventory	J-361	Fugitive	Modular Equipment Shelter	2.30E-15	Radioactive gases
	Systems Located on the PNNL Richland Campus				
Cont. Monitoring	EP-3410-01-S	Major Point	Material Science and Technology Laboratory	3.16E-04	
Cont. Monitoring	EP-3420-01-S	Major Point	Radiation Detection Laboratory	6.75E-03	
Inventory	EP-3420-02-S	Minor Point	Radiation Detection Laboratory	1.30E-06	
Inventory	J-3425	Fugitive	Ultra-Low Background Counting Laboratory	1.49E-06	
Cont. Monitoring	EP-3430-01-S	Major Point	Ultra-Trace Laboratory	1.89E-03	
Inventory	EP-3430-02-S	Minor Point	Ultra-Trace Laboratory	1.51E-07	
Inventory	EP-3430-1606P-S	Minor Point	Ultra-Trace Laboratory – Perchloric Hood	4.89E-13	Trace quantities
Inventory	EP-3430-1608P-S	Minor Point	Ultra-Trace Laboratory – Perchloric Hood	4.89E-13	Trace quantities
Inventory	EP-3430-1610P-S	Minor Point	Ultra-Trace Laboratory – Perchloric Hood	4.89E-13	Trace quantities
Inventory	EP-3430-1612P-S	Minor Point	Ultra-Trace Laboratory – Perchloric Hood	4.89E-13	Trace quantities
Inventory	EP-3430-1614P-S	Minor Point	Ultra-Trace Laboratory – Perchloric Hood	4.89E-13	Trace quantities
Inventory	EP-LSLII-01-V EP-LSLII-02-V	Minor Point	Life Sciences Laboratory II	0	Inventory based
Inventory	EP-RTL-520 -10-V EP-RTL-520 -11-V	Fugitive - Transitioned	Research Technology Laboratory (RTL)	0	Transitioned to inventory in August 2014
Inventory	J-RTL-530	Fugitive - Transitioned	RTL Support Building	4.04E-08	Transitioned to inventory in October 2012
Survey	J-RTL-Complex	Fugitive	Research Technology Laboratory Complex	N/A	Demolition. emissions limited to 2.0E-04 mrem/yr

Required Emission Monitoring Method per WDOH License[(a)]	Emission Unit	Emission Type[(b)]	System Description	2017 Assessment Dose, mrem/yr	Comment
	Systems Located Elsewhere in Richland, Washington				
	N/A	N/A	N/A	N/A	
	Systems Located at the MSL in Sequim, Washington				
Inventory	MSL-1	Fugitive	MSL Building 1 (Beach Office)	1.32E-04	Transitioned to J-MSL in 2018.
Inventory	MSL-5	Fugitive	MSL Building 5 (Upland)	4.01E-05	Transitioned to J-MSL in 2018.

(a) **Monitoring Measurement Required**: "*Inventory*" means 40 CFR 61 Appendix D Methods based on radionuclide inventory. *Cont. Monitoring* means continuous measurement (sampling) of stack emissions. "*Periodic*" means periodic (non-continuous) measurement of emissions. "*Survey*" means in field radiological surveys conducted during demolition and excavation.

(b) **Emission Types**: "*Fugitive* emissions" are radioactive air emissions that do not and could not reasonably pass through a stack, vent, or other functionally equivalent structure, and that are not feasible to directly measure and quantify. "*Major Point* source" is a discrete, well-defined location from which radioactive air emissions originate, such as a stack, vent, or other functionally equivalent structure with a PTE equal to or greater than 0.1 mrem/yr and requires continuous sampling and in some cases also continuous monitoring. "*Minor Point* source" is a discrete, well-defined location from which radioactive air emissions originate, such as a stack, vent, or other functionally equivalent structure with a PTE less than 0.1 mrem/yr. (WAC 2018)

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Appendix A

Radioactive Material Tracking Database Features

Appendix A

Radioactive Material Tracking Database Features

The Radioactive Material Tracking (RMT) System is a web-based, real-time-tracking software system. RMT users add, move, modify, or ship out radioactive materials in the system to maintain inventory management and to verify that emissions are below regulatory permitted quantities. The calculated potential-to-emit (PTE) information obtained from the RMT System provides the basis to verify that each building is operating within set threshold limits. The annual Radiation National Emission Standards for Hazardous Air Pollutants assessment process includes verification of this information and compiling it into a final report. The inventory information obtained and reported is described in Section 2.1.

A.1 Database Population

New or updated inventory information is obtained from individual research personnel who act as custodians or users of the material. The PNNL HDI work control Radiological – General, Exhibit – Radioactive Material Tracking (RMT), contains requirements for entering and maintaining inventory data. RMT users have been granted access to the RMT System and are trained by an RMT Administrator. Training is documented and maintained through the PNNL Laboratory Training Database.

Information entered into the RMT System that is pertinent to radiological air emissions includes:

- Name of the staff member acting as custodian of the material
- Material physical form—gas, liquid, particulate solid, non-particulate solid, non-dispersible, or sealed source
- Radionuclides[17]
- Inventory in activity, mass, or concentration for each nuclide
- Building and room in which material is stored or used
- Specific item description and any additional comments related to the material (e.g., reference numbers on the material, whether the material is considered throughput, and a description of the material)
- RMT ID number is assigned by the database for each entry.

Additional information may be entered to assist staff in identification or management of the material.

A.2 RMT Reference Table Information

Potential dose calculations are made possible through reference table data that are run in a software stored procedure. These software stored procedures and reference tables are developed and maintained as safety software under the *RMT System Quality Assurance Plan* (PNNL-FS-RMT-017). Any changes to these reference tables or processes require rigorous testing, peer review, and documentation before implementation.

[17] Appendix B provides support data for radionuclides in uranium and plutonium enrichments.

Dose-per-Unit Release Factors Table – Current factors for the mrem dose per curie released to ambient air (mrem/Ci) for different isotopes are listed in:

- Hanford Site emission facilities: DOE/RL-2006-29 (Revision 1) (Rhoads et al 2010)
- PNNL Richland Campus emission facilities: PNNL-17847 (Revision 1) (Rhoads and Barnett 2009)
- MSL emission facilities: Anderson (2012).

In 2019, the RMT software is expected to be updated to incorporate the most current dose factors. Nevertheless, the existing dose factors remain useful and typically are not very different from the current ones.

- Hanford Site emission facilities: DOE/RL-2006-29 (Revision 2) (Snyder and Rokkan 2016)
- PNNL Campus emission facilities: PNNL-17847 (Revision 4) (Snyder and Barnett 2016)
- MSL emission facilities: PNNL-22342, Appendix B (Snyder et al 2018).

Release Fraction Table – The release fractions for material forms (such as gases, liquids, or particulate solids).

Location Modification Factors Table – LMF for the building from which the potential emission occurs. Unit dose factors are calculated for the worst-case offsite unit dose factor in the 300 Area (offsite MPR <40 m release height, east). LMFs modify these doses for facilities in Richland based on a ratio of dispersion factors.

A.3 Calculations

The database uses queries and macros that are applied to the inventory data to calculate the potential dose for the different PNNL buildings.

Normalizing Inventory Data – The database is designed to convert the reported mass and activity inventory units (such as g, mCi, μCi, mg, μg) to Ci units for use in subsequent calculations.

Potential Dose Calculations – Potential dose calculations are determined on a building-specific basis. The reported inventory is first converted to Ci and then is multiplied by release fraction (based on the physical form of the item), the dose-per-unit release factor (mrem/Ci) for the specific nuclide, and the LMF.

Example: Calculate the PTE to the offsite MPR for 20 g of ^{238}U powder in the 325 Building

$$20 \text{ g} \times 3.36\text{E-7 Ci/g } ^{238}\text{U} \times 1\text{E-03} \times 66 \text{ mrem/Ci/yr} \times 1.0 = 4.4\text{E-07 mrem/yr}$$

where:

20 g	=	quantity of material
3.36E-7 Ci/g	=	specific activity of ^{238}U
1E-03	=	release fraction for particulate solids or liquids
66 mrem/Ci/yr	=	dose-per-unit release factor of ^{238}U for the offsite receptor in the east sector of the 300 Area with a < 40 m release height
1.0	=	LMF (with relation to 300 Area).

The cumulative PTE for the building is determined by summing the potential doses of each inventory entry.

Note that PTE calculations are performed by the RMT software for inventory changes in Hanford Site and PNNL Richland Campus facilities. For MSL, these calculations are performed using an Excel spreadsheet. Spreadsheet updates are prompted by emails generated when a transaction involving MSL is proposed in RMT. Proposed new receipts and moves of radioactive materials to/from MSL also are flagged in the Battelle radioactive material shipping process.

A.4 Radioactive Material Tracking Reports

Reports generated by RMT System for the radioactive air National Emission Standards for Hazardous Air Pollutants assessment include the Throughput Report, the FEMP Data Evaluation Report, and the FEMP Dose Contribution Report. Together these reports provide the itemized inventory data provided by the RMT System, including current inventory plus additional inventory anticipated to be processed in the coming year or brought in for new projects. The reports provide the cumulative inventory that has been in the building during the calendar year, physical forms, release fractions, dose-per-unit-release factor, and PTE values. The reports also provide a complete listing of each radioisotope present in the building, associated dose contribution (mrem), and the percent of the total dose for the building.

Other methods not available in the RMT System are also utilized during the assessment process. These include the Volumetric Released Radioactive Material approvals for the calendar year, the Radiological Air Gas database authorizations for gas emissions during the calendar year, and the Sequim Tracking spreadsheet maintained by the RMT System Administrator. Finally, the Electronic Prep and Risk System is evaluated for upcoming projects with radioactive material.

Appendix B

Common Radionuclide Mixtures

Appendix B

Common Radionuclide Mixtures

For uranium and plutonium inventory items where enrichment is known but data on specific isotopic breakdown are not available, a method was devised to conservatively estimate isotopic composition. Uranium and plutonium blends can be grouped under the categories shown in Table B.1. Each of these categories represents an isotopic blend of uranium or plutonium that may be commonly found on the Hanford Site. The percentages shown are weight percent, and other components that make up the blend are other isotopes of uranium or plutonium. For aged plutonium blends, Am-241 also makes up a significant fraction.

Table B.1. Uranium and Plutonium Blend Information

Material in Inventory	Blend Information	Bin
Depleted Uranium or Uranium ≤0.25% U-235	Depleted Uranium	U(dep)
Natural Uranium or Uranium ≤0.72% U-235	Natural Uranium	U(nat)
Uranium ≤0.83% U-235 (commonly found at Hanford)	Hanford Uranium	U(Hanf)
Uranium Enriched ≤20% U-235	Uranium Enriched <20%	U(20%)
Uranium Enriched ≤90% U-235	Uranium Enriched <90%	U(90%)
Uranium ≥90% U-235	U-235	U-235
Plutonium with ≤6% Pu-240	Pu Blend with 6% Pu-240	Pu (6%)
Plutonium with ≤12% Pu-240	Pu Blend with 12% Pu-240	Pu (12%)
Plutonium with ≥12% Pu-240	Pu Blend with 24% Pu-240	Pu (24%)

Data and calculations for each of the blends are described here. Uranium blend information was obtained from Sula, Carbaugh, and Bihl (1991) and is shown in Table B.2.

Table B.2. Uranium Blend Specific Activities

Uranium Blend	Specific Activity, Ci/g
U(dep)	3.64E-7
U(nat)	6.87E-7
U(Hanf)	9.0E-7
U(20%)	9.36E-6
U(90%)	6.21E-5

Data for depleted uranium, natural uranium, and uranium commonly found at Hanford are from Tables 8.2 and 8.3 of the referenced report. For the 20% and 90% U-235 blends, an equation was used to calculate specific activity. The equation was obtained from Figure 8.1 of Sula, Carbaugh, and Bihl (1991) and is back-referenced to WASH-1251 (Alexander 1974).

$$SA = (0.4 + 0.38E + 0.0034E^2) \times 10^{-6}$$

where: SA = specific activity, Ci/g and
E = weight percent of U-235.

For E = 20 wt% U-235, S = 9.36E-6 Ci/g
For E = 90 wt% U-235, S = 6.21E-5 Ci/g

The uranium isotopes that contribute significantly to the activity are alpha emitters and have approximately the same dose potential per curie. Therefore, the specific activity is used in converting a known mass of uranium blend to activity, and the activity is all attributed to U-235.

The radionuclide and isotopic composition of 6% and 12% plutonium blends was also obtained from Sula, Carbaugh, and Bihl (1991). Data for these Pu mixtures prior to any decay of Pu-241 to Am-241 are shown in Table B.3 and were obtained from Tables 9.1 and 9.2 of the referenced report.

Table B.3. Isotopic Composition and Specific Activity of Pu Blends

		6% Pu mix			**12% Pu mix**		
Isotope	**Specific Activity Ci/g**	**No Decay wt%**	**40-yr Decay wt%**	**Spec Act Mix Ci alpha/g**	**No Decay wt%**	**40-yr Decay wt%**	**Spec Act Mix Ci alpha/g**
Pu-238	17.1	0.05	0.05	8.6E-03	0.1	0.1	1.7E-02
Pu-239	0.0621	93.0	93.0	5.8E-02	84.4	84.4	5.2E-02
Pu-240	0.227	6.1	6.1	1.4E-02	12.4	12.4	2.8E-02
Pu-241	103	0.8	0.1	[a]	3.0	0.45	*
Pu-242	3.92E-03	0.05	0.05	2.0E-06	0.1	0.1	3.9E-06
Am-241	3.43	0.0	0.7	2.3E-02	0.0	2.55	8.7E-02
			Total mix	1.0E-01		Total mix	1.9E-01

[a] Pu-241 is excluded from the total mix calculation because it is a beta-emitter, and the dose is insignificant compared to the dose from the other alpha-emitting radioactive materials.

Plutonium inventory items at Hanford have most likely aged for many years and contain significant amounts of Am-241.[18] A 40-year age is assumed for plutonium blend calculations and the amount of Pu-241 decayed to Am-241 is calculated using the following equation (Shleien, Slaback, and Birky 1998):

$$N/N_0 = e^{(-0.693\, t / T)}$$

where: N/N_0 = the fraction of parent material left,
t = time, and
T = ½ life of the parent material.

Pu-241 has a half-life of 14.4 years (Shleien, Slaback, and Birky 1998, Table 8.13) and decays to about 15% of its original mass after 40 years, according to the above equation. Am-241 has a much longer half-life (432 years), so all of the Pu-241 that is converted to Am-241 is present. Table B.3 shows the resulting weight percent of each isotope after a 40-year decay for each of the plutonium blends.

All of the isotopes in Table B.3 are alpha emitters except for Pu-241 which decays by beta emission and is much less damaging per curie than the others. The dose effect from the Pu-241 contribution is negligible compared to the rest. Thus, Pu-241 is excluded from further calculations, and the specific activity for the mix is calculated in terms of curies of alpha emitter per gram. The specific activity of the mix is determined by summing the contributions from the alpha-emitting nuclides.

The isotopic composition for a 24% Pu blend was obtained from ANSI N317–1980, *Performance Criteria for Instrumentation Used for Inplant Plutonium Monitoring*. This document provides isotopic compositions of plutonium for different reactor types and burnup. The composition that results in the most conservative unit dose is shown in Table B.4 and corresponds to the data for boiling water reactor

[18] Fuel reprocessing at Hanford took place from the mid-1940s to the mid-1980s (Ballinger and Hall 1991).

with 28,000 MWD/T burnup with 40 years decay for Pu-241. The data in Table B.4 were taken directly from ANSI N317 and do not quite add up to 100%, most likely because of the number of significant digits used in the data.

Table B.4. Isotopic Composition and Specific Activity of 24% Pu Blend

		24% Pu mix		
Isotope	**Specific Activity Ci/g**	**No Decay wt%**	**40-yr Decay wt%**	**Spec Act Mix Ci alpha/g**
Pu-238	17.1	1.80	1.8	3.1E-01
Pu-239	0.0621	54.20	54.2	3.4E-02
Pu-240	0.227	23.80	23.8	5.4E-02
Pu-241	103	13.50	2.0	[a]
Pu-242	3.92E-03	6.40	6.4	2.5E-04
Am-241	3.43	0.00	11.5	3.9E-01
			Total mix	7.9E-01

[a] Pu-241 is excluded from the total mix calculation because it is a beta-emitter and the dose is insignificant compared to the dose from the other alpha-emitting radioactive materials.

Appendix C

Comparison of EPA Recommendations with PNNL Radiological Air Task Documents

Appendix C

Comparison of EPA Recommendations with PNNL Radiological Air Task Documents

Comparison of EPA Recommendations for a Uniform Protocol for Periodic Confirmatory Measurements of “Minor” Air Emissions Sources Subject to 40 CFR Part 61, Subpart H (May 9, 2007) with Pacific Northwest National Laboratory (PNNL) Radioactive Air Task Documents.

Recommendations for Periodic Confirmatory Measurements (PCMs)	• **PNNL-10855, Rev 6 (2018), *Determining Unabated Airborne Radionuclide Emissions Monitoring Requirements Using Inventory-Based Methods*** • **PNNL-19904, Rev 5 (2018), *PNNL Potential Impact Categories*** • **EM-QA-01, Rev 10 (2016), *Effluent Management Quality Assurance Plan*** • **PNNL-15992, Rev 4 (2018), *Pacific Northwest National Laboratory Facility Radionuclide Emission Points and Sampling Systems***
(1) GRADED APPROACH TO CLASSIFICATION SYSTEM: Describe how minor sources are subdivided and the basis for each classification.	PNNL-19904, the PNNL potential impact categories (PICs) document, specifies the basis for minor source categories, identifies a PIC for each PNNL minor source, and the use of PIC-5 for Campus-wide radionuclide emissions applications.
(2) METHODS FOR PCM: Methods used to confirm that minor sources are correctly categorized (e.g., emissions measurement, radionuclide inventory).	PNNL-10855 describes the methodology for completing the annual National Emission Standards Hazardous Air Pollutant assessment. The annual assessments use current radionuclide inventory for each building.
(3) SUPPORTIVE DATA: • Meteorological • Release fractions • Materials volatilization temperatures • Maximally exposed individual selection method	• The Hanford Site Meteorological Station provides meteorological measurements for the Hanford Site and PNNL Richland Campus. Dose modeling meteorological data are published in the appendix of the annual radionuclide air emission report for the Hanford Site (i.e., DOE/RL-2018-05). No detailed meteorological data is currently required for MSL. • PNNL-10855 describes release fractions. • PNNL-10855 describes materials volatilization temperatures considered in potential release-fraction determinations. • Maximally exposed individual selection for: – Hanford Site emissions: Described in DOE/RL-2006-29, *Calculating Potential-to-Emit Radiological Release and Doses*. – PNNL Richland Campus emissions: Described in PNNL-17847, *PNNL Campus Dose-per-Unit-Release Factors for Calculating Radionuclide Emissions Potential-to-Emit Doses.* – MSL emissions: Described in annual air emissions reports (i.e., PNNL-22342-6).
(4) DISPERSION/DOSE MODEL USED: The reason for using any code other than CAP-88 version 3 (or newer version) should be explained.	CAP88-PC used for Hanford Site and PNNL Richland Campus dispersion modeling as described in PNNL-10855. COMPLY used for MSL dispersion modeling due to small emissions and limited meteorological data availability.
(5) QUALITY CONTROL ASPECTS: Quality assurance activities performed on a minor source should be consistent with a graded approach.	EM-QA-01 details quality assurance methods in place to validate the data gathering and reporting process. Standard operating procedures are implemented for related work (e.g., sampling activities) and updated biennially.

Recommendations for Periodic Confirmatory Measurements (PCMs)	• **PNNL-10855, Rev 6 (2018),** ***Determining Unabated Airborne Radionuclide Emissions Monitoring Requirements Using Inventory-Based Methods*** • **PNNL-19904, Rev 5 (2018),** ***PNNL Potential Impact Categories*** • **EM-QA-01, Rev 10 (2016),** ***Effluent Management Quality Assurance Plan*** • **PNNL-15992, Rev 4 (2018),** ***Pacific Northwest National Laboratory Facility Radionuclide Emission Points and Sampling Systems***
(6) FREQUENCY OF CONFIRMATION: The frequency that source emissions will be confirmed by sampling or other means.	PNNL-10855 describes the annual National Emission Standards Hazardous Air Pollutant assessment methodology. The Washington Department of Health permits specify monitoring requirements and sampling frequencies. Current sampling frequencies are maintained in a database (e.g., Gaseous Effluent Database) and documented in PNNL-15992. Note: Although the potential-to-emit (PTE) for each emission unit is calculated annually using actual radionuclide inventory, the PTE used for assigning PICs should be the permitted PTE, which is based on maximum estimated inventory and throughput for permitted activities.

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