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Pacific Northwest National Laboratory
Operated by Battelle for the U.S. Department of Energy

Assessment of Unabated Facility Emission Potentials for Evaluating Airborne Radionuclide Monitoring Requirements at Pacific Northwest National Laboratory - 2007

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January 2008

Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

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Abstract

Assessments were performed to evaluate compliance with the airborne radionuclide emission monitoring requirements in the National Emission Standards for Hazardous Air Pollutants (NESHAP – U.S. Code of Federal Regulations, Title 40, Part 61, Subpart H) and Washington Administrative Code (WAC) 246-247: Radiation Protection – Air Emissions. In these NESHAP assessments, potential unabated offsite doses were evaluated for emission locations at buildings that are part of the consolidated laboratory campus of the Pacific Northwest National Laboratory. This report describes the inventory-based methods and provides the results for the NESHAP assessment performed in 2007.

Summary

Assessments were performed to evaluate compliance with the airborne radionuclide emission monitoring requirements in the National Emission Standards for Hazardous Air Pollutants (NESHAP—U.S. Code of Federal Regulations, Title 40 Part 61, Subpart H). In these NESHAP assessments, potential unabated offsite doses were evaluated for emission locations at buildings that are part of the consolidated laboratory campus of Pacific Northwest National Laboratory (PNNL) operated by Battelle for the U.S. Department of Energy. Two of the buildings evaluated, the 325 Building Radiochemical Processing Laboratory and the 331 Building Life Sciences Laboratory I, met state and federal criteria for continuous sampling of airborne radionuclide emissions. The NESHAP assessments were performed using building radionuclide inventory data obtained in 2007. The buildings that were evaluated are listed below.

DOE Owned Buildings

200E Prototype Surface Barrier Storage
318 Radiological Calibrations Laboratory
320 Analytical and Nuclear Research Laboratory
325 Radiochemical Processing Laboratory
326 Materials Sciences Laboratory
329 Chemical Sciences Laboratory
331 Life Sciences Laboratory I
331-G Interim Tissue Repository
331-H Aerosol Wind Tunnel Research Facility
338 Prototype Engineering Laboratory
361 Modular Equipment Shelter (National Nuclear Security Administration)
747A Whole Body Counter
3020 Environmental Molecular Sciences Laboratory

Privately Owned Buildings Located in Richland, Washington

2400 Stevens Office Building
APEL Applied Process Engineering Laboratory
RTL Research Technology Laboratory
Sigma-5 Office Building
PSL Physical Sciences Laboratory
LSL-II Life Sciences Laboratory II

Glossary

AMD	Aerodynamic Mean Diameter
ANSI	American National Standards Institute
APEL	Applied Process Engineering Laboratory
CAP88-PC	Clean Air Act Assessment Package -1988 for Personal Computers
CFR	Code of Federal Regulations
DOE	Department of Energy
DOT	Department of Transportation
EM	Effluent Management
EMSL	Environmental Molecular Sciences Laboratory
EPA	U.S. Environmental Protection Agency
LMF	Location Modification Factor
LSL-II	Life Sciences Laboratory II
MPR	Maximum Public Receptor
NDA	Nondestructive Assay
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
RIDS	Records Inventory and Disposition Schedule/File Index
RMT	Radioactive Material Tracking System
RSR	Routine Radioactive Shipment Record
RTL	Research Technology Laboratory
TID	Tamper Indicating Devices
WAC	Washington Administrative Code

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Tables

1.0 Introduction

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

- U.S. Code of Federal Regulations (CFR), Title 40, Subpart H: National Emission Standards for Emissions of Radionuclides Other than Radon from Department of Energy Facilities (2002) (40 CFR 61)
- Washington Administrative Code (WAC) 246-247: "Radiation Protection - Air Emissions" (WAC 2005)
- U.S. Department of Energy, DOE/EH-0173T, Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance (DOE 1991).

These documents require the performance of continuous sampling at emission points that have a potential-to-emit (PTE)[(a)] $\geq$ 0.1 mrem/yr offsite maximum receptor dose if routine emissions were not mitigated by engineered pollution control systems.

In response to these requirements, the potential unmitigated offsite receptor dose from the buildings at Pacific Northwest National Laboratory[(b)] (PNNL) that contain radioactive materials or sources is evaluated annually. These evaluations were performed initially in 1991 for the PNNL facilities on the Hanford Site. Based on the initial assessments, four PNNL buildings were identified as containing a sufficient inventory of radioactive material that unmitigated emissions could potentially result in an annual offsite maximum receptor dose of $\geq$ 0.1 mrem. These buildings were the 324 Waste Technology Engineering Laboratory, the 325 Radiochemical Processing Laboratory, the 327 Postirradiation Testing Laboratory, and the 3720 Environmental Sciences Laboratory. In accordance with the National Emission Standard for Hazardous Air Pollutants (NESHAP), qualifying emission points from these buildings were sampled continuously.

The original radionuclide assessments were updated annually. The number and status of buildings evaluated has changed as buildings were transitioned to other contractors, new buildings were added, or laboratory missions were changed. The NESHAP assessments include all PNNL buildings with radioactive materials. This document describes the methodology used and reports summary results for those PNNL buildings. Results of the 1992–1993 assessment were documented by Sula and Jette (1994) with updates to this document in 1995, 1999, 2001, and 2003. This report describes the current methodology for preparing the annual NESAP assessment for 2007.

(a) Potential-to-emit 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.

(b) Pacific Northwest National Laboratory is operated by Battelle for the U.S. Department of Energy under Contract DE-AC05-76RL01830.

2.0 Assessment Methodology

This section describes the methods used by PNNL to determine the potential emissions of radioactive materials from buildings operated by Battelle.

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[(a)] 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 from year-to-year and where historical sampling data may not be a reliable predictor of future emissions.

Since 2006, PNNL has maintained radioactive source and material information using the Radioactive Material Tracking System (RMT). This system is a web-based software tool that supports real-time tracking of radioactive materials. The RMT system provides a process to comply with numerous regulatory requirements pertaining to radioactive material management. Appendix A provides a summary of the database features that apply to radioactive air emissions. The RMT system is updated by radioactive material custodians as well as other users with unescorted 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. The RMT also has extensive search and report capabilities.

(a) This method is described in WAC 246-247-030 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 100ºC or more, boils at a temperature of 100ºC or less, or is intentionally dispersed into the environment.

The PNNL Effluent Management (EM) Group annually requests that RMT custodians and cognizant space managers review and verify current radioactive inventory as well as throughput and proposed radioactive additions in the upcoming year. This request is made through a central point of contact for each of the PNNL divisions, who then requests inventory information from custodians in his/her organization.

The radioactive material custodians and cognizant space managers verify the RMT inventory for material under their scope of responsibility. The inventory information is downloaded to a Microsoft Access database to generate the final reports. The Effluent Management Group also incorporates radioactive materials that have been removed from a building in the past year and also emissions reported from the Radioactive Air Gas Inventory Database.

Radionuclides meeting any 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 EM Records Inventory and Disposition Schedule/File Index (RIDS) assessment records.

Potential release fractions for radionuclides are based on the physical form of the radionuclide as shown in Table 2.1. Radionuclides present as sealed sources or in sealed, unvented Department of Transportation (DOT) shipping containers are assumed to be unavailable for release under normal circumstances.

2.2 Maximum Public Receptor Unit Dose Calculation

For unit dose calculations, the offsite maximum public receptor (MPR) is defined as an individual whose residence location, work location, and lifestyle maximize the dose from airborne pathways. All potential environmental transport pathways associated with an airborne radionuclide release were included (that is, air inhalation, air submersion, exposure to deposited radionuclides, and ingestion).

Unit dose factors for the offsite maximum public receptor were calculated for specific radionuclides using the U.S. Environmental Protection Agency (EPA) compliance code CAP88-PC (Chaki and Parks 2000). Radionuclides that were not represented in CAP88-PC were conservatively assigned default values, usually equal to that of 241Am for alpha emitters or ^{137}Cs for non-alpha emitters. The decay of the daughter products was also considered in assigning default values for short half-life radionuclides. The unit dose release factors were calculated for the Hanford Site and documented (Diediker et al. 2006). This document describes the methods and assumptions used and provides unit dose factors for the 300 Area.

Table 2.1. Physical Forms and Potential Annual Release Fractions for Radionuclides

Form	Description	Potential Release Fraction[(a)]
Gas	Nuclide in a gaseous form.	1
Gas (Unopened Cylinder)	Nuclides in gaseous form in unopened commercial gas cylinders.	10^{-3}
Liquid (Heated > B.P.)	Nuclide will exceed its boiling point when uncontained.	1
Liquid	Nuclide will exceed its melting point (liquid form) or be present in particulate form (Aerodynamic Mean Diameter [AMD] <100 microns) when uncontained; exception—liquid and powders in unopened containers may be listed as solid(s).	10^{-3}
Liquid (Unopened Container)	Nuclides not meeting conditions for the more dispersible classes.	10^{-6}
Powder	Nuclide will be present in particulate form (AMD <100 microns).	10^{-3}
Solid (Heated > M.P.)	Nuclide will exceed its melting point.	10^{-3}
Solid	Nuclides not meeting conditions for the more dispersible classes.	10^{-6}
Powder (Unopened Container)	Nuclides not meeting conditions for the more dispersible classes.	10^{-6}
Solid (Unopened Container)	Nuclides not meeting conditions for the more dispersible classes, including material in sealed, DOT containers.	0
Sealed Source: Any Type	Sealed sources.	0
(a) Based on Table 1 of ANSI/HPS N13.1-1999 (ANSI/HPS 1999).		

For the Environmental Molecular Sciences Laboratory (EMSL), dose assessments were performed by applying a *location modification factor* to the 300 Area unit dose factor to correct for varying source-receptor distances and directions. The location modification factor was calculated by dividing the atmospheric dispersion values (Chi/Q) for this building by the atmospheric dispersion values for the 300 Area. The compliance code CAP88-PC was used to calculate these dispersion values. Similar calculations were performed to obtain location modification factors for other Battelle-operated DOE buildings outside the 300 Area.

2.3 Potential Emission Dose Assessment

Doses from projected radionuclide emissions were calculated by multiplying the quantity of each radionuclide present in the building by its associated potential release fraction, the 300 Area unit dose

release factor, and, if necessary, the location modification factor. Doses from individual radionuclides were summed to derive the total potential unabated annual emission dose for each building.

The facility radionuclide NESHAP assessments were prepared for each building and contained the raw inventory information, any communications clarifying or correcting the inventory information, summarized inventory information, and a cover sheet showing the resulting dose and approval signatures. The assessments were independently reviewed and subsequently approved by the author, technical reviewer, divisional points of contact for the inventory custodians, and the applicable building manager. After approval, assessments are maintained as records in the Effluent Management Group RIDS.

3.0 Reports

Three reports are generated using the database for the NESHAP assessment for inclusion in the final packet:

- A report of the raw data provided by the RMT system, proposed data and throughput data.
- A summary page listing the potential dose (mrem/yr) for the building inventory with sign-off blocks.
- A complete listing of each radioisotope present in the building, associated dose contribution (mrem), and the percent of the total dose for the building.

A summary of the assessment results for 2007 is provided in Table 3.1. The table also identifies the radionuclides in inventory that contribute 10% or more of the potential dose for buildings where sampling is required.

Table 3.1. Emission System Potential Dose Assessment Summary

Emission System	Emission Type[a]	System Description	Emission Measurement Required	Potential Offsite Dose (mrem/yr)	Comment
Systems Located on the Hanford Site					
318	Point	Radiological Calibrations Laboratory	Periodic	4.29E-06	Primarily sealed and check sources
320	Point	Analytical and Nuclear Research Laboratory	Periodic	1.01E-03	
323	Point	Mechanical Properties Laboratory	Periodic	1.17E-07	
325	Point	Radiochemical Processing Laboratory	Continuous	3.74E+01	
326	Point	Materials Sciences Laboratory	Periodic	4.85E-03	
329	Point	Chemical Sciences Laboratory	Periodic	1.59E-02	
331	Point	Life Sciences Laboratory I	Continuous	1.13E-01	
331G	Point	Interim Tissue Repository	None	0	Sealed Sources
331H	Point	Aerosol Wind Tunnel Research Facility	None	0	Sealed Sources
338	Fugitive	Prototype Engineering Laboratory	None	0	Sealed Sources
361	Fugitive	Modular Equipment Shelter	None	1.82E-13	
3730	Point	Gamma Irradiation Facility	Periodic	2.34E-04	
200E Storage	Fugitive	Prototype Surface Barrier Storage	None	0	Sealed Sources
Systems Located on the PNNL Site					
3020 (EMSL)	Point	Environmental Molecular Sciences Laboratory	None	0	Sealed Sources
Systems Located in Richland					
747A	Fugitive	Whole Body Counter	None	0	Sealed Sources
2400 Stevens	Fugitive	2400 Stevens Office Building	None	0	Sealed Sources
APEL	Fugitive	Applied Process Engineering Laboratory	None	0	Sealed Sources
RTL	Point	Research Technology Laboratory	Periodic	5.07E-3	
Sigma-5	Point	Sigma 5 Office Building	None	0	Sealed Sources
PSL	Point	Physical Sciences Laboratory	None	0	Sealed Sources
LSL-II	Point	Life Sciences Laboratory II	Periodic	1.71E-6	

(a)"Fugitive emissions" are radioactive air emissions that do not and could not reasonably pass through a stack, vent, or other functionally equivalent structure and which are not feasible to directly measure and quantify.
"Point source" is a discrete, well-defined location from which radioactive air emissions originate, such as a stack, vent, or other functionally equivalent structure (WAC 2005).

4.0 References

40 CFR 61, Subpart H. 2002. "National Emission Standards for Emissions of Radionuclides Other Than Radon From Department of Energy Facilities." *U.S. Code of Federal Regulations*, U.S. Environmental Protection Agency.

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Washington Administrative Code (WAC). 2005. "Radiation Protection - Air Emissions." WAC 246-247, Olympia, Washington.

Appendix A
Radioactive Material Tracking Database Features

The Radioactive Material Tracking (RMT) system was developed and maintained to be a web-based, real-time-management software system. RMT users update or propose 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 NESHAP assessment process includes downloading this information and compiling it into a final report. The inventory information obtained and reported is described in Section 2.1.

Database Population

New or updated inventory information is obtained from individual research personnel who act as custodians or users of the material. Inventory information is required to be updated during any significant changes of the material or when the material changes location. The 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.

The 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
- Name of the research or support division of the custodian
- Material form—gas, liquid, powder, solid, sealed source
- Inventory basis—analysis, book value, estimate (routine radioactive shipment record [RSR]/transportation records), estimate (other), estimate (process knowledge), Hanford Tanks, nondestructive assay (NDA), piece count (tamper indicating devices [TID]), spent fuel
- Nuclides
- Inventory in activity or concentration for each nuclide
- Building and room in which material is stored or used
- User-defined identification—numbers previously assigned by the custodian or by other databases (sealed source or material balance area identification numbers are often used)
- Specific item description and any additional comments related to the material (for example, reference numbers on the material, whether or not the material is considered throughput, and a description of the material)
- RMT ID number is assigned by the database for each entry.

RMT Reference Table Information

Potential dose calculations are made possible through reference table data that are run in an Oracle 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, Rev. 3). Any changes to these reference tables or processes require rigorous testing, peer review, and documentation before implementation.

Dose-per-Unit Release Factors Table—Factors for the dose per curie (mrem/Ci) for different isotopes are listed in DOE/RL-2006-29, Revision 0 and are entered as a separate table in the database.

Release Fraction Table—The release fractions for material forms (such as, solids, liquids, or powders).

Location Modification Factors Table—Location Modification Factor 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). Location modification factors (LMFs) modify these doses for other facilities based on a ratio of dispersion factors.

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, grams, mCi, μCi, mg, μg) to curie (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 the dose-per-unit release factor (mrem/Ci) for the specific nuclide, the location modification factor, and the release fraction to determine the potential dose for that nuclide inventory.

Example: Calculate the potential-to-emit to the offsite maximum public receptor for 20 g of ^{238}U powder in the 325 Building

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

where
- 20 g = quantity of material
- 3.36E-7 Ci/g = specific activity of ^{238}U
- 1E-03 = release fraction for powders 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 = location modification factor (with relation to 300 Area).

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

Reports

Three reports are generated using the database for the NESHAP assessment for inclusion in the final packet:

- A report of the raw data provided by the RMT system, proposed data, and throughput data.

- A summary page listing the potential dose (mrem/yr) for the building inventory with sign-off blocks for the preparer, reviewer, building safety representative, divisional representative(s), and the building manager.

- A complete listing of each radioisotope present in the building, associated dose contribution (mrem), and the percent of the total dose for the building.

Appendix B

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 PNNL Radiological Air Task Documents

Recommendations for Periodic Confirmatory Measurements (PCMs)	• **PNNL-10855, Rev. 4, *Assessment of Unabated Facility Emission Potentials for Evaluating Airborne Radionuclide Monitoring Requirements at Pacific Northwest National Laboratory – 2007*** • ***PNNL Potential Impact Categories, June 2006, Rev. 1*** • **EM-QA-01, Rev. 5, *Effluent Management Quality Assurance Plan*** • **PNNL-15992, *300 Area 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.	The PNNL Potential Impact Categories (PIC) document specifies the basis for minor source categories and identifies a PIC for each PNNL minor source.
(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 NESHAP assessment. The annual assessments use current radionuclide inventory for each building.
(3) SUPPORTIVE DATA: • Meteorological • Release Fractions • Materials volatilization temperatures • MEI selection method	• The Hanford Meteorological Station provides meteorological measurements. Dose modeling meteorological data are published in the appendix of the annual air emission report for the Hanford Site (i.e., DOE/RL-2007-01). • Release fractions described in PNNL-10855. • Materials volatilization temperatures considered in potential release-fraction determinations, PNNL-10855 • MEI selection for the Hanford Site emissions described in the *Calculating Potential-to-Emit Radiological Release and Doses* (DOE/RL-2006-29).
(4) DISPERSION/DOSE MODEL USED: The reason for using any code other than CAP-88 version 3 should be explained.	CAP88-PC used for dispersion modeling as described in PNNL-10855.

Recommendations for Periodic Confirmatory Measurements (PCMs)	• **PNNL-10855, Rev. 4,** ***Assessment of Unabated Facility Emission Potentials for Evaluating Airborne Radionuclide Monitoring Requirements at Pacific Northwest National Laboratory – 2007*** • ***PNNL Potential Impact Categories, June 2006, Rev. 1*** • **EM-QA-01, Rev. 5,** ***Effluent Management Quality Assurance Plan*** • **PNNL-15992,** ***300 Area Pacific Northwest National Laboratory Facility Radionuclide Emission Points and Sampling Systems***
(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.
(6) FREQUENCY OF CONFIRMATION: The frequency that source emissions will be confirmed by sampling or other means.	PNNL-10855, describes the annual NESHAP assessment methodology. The Washington Department of Health permits specify monitoring requirements and sampling frequencies. Current sampling frequencies are maintained in the 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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