PNNL-22342-3

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Department of Energy – Office of Science
Pacific Northwest National Laboratory

Marine Sciences Laboratory Radionuclide Air Emissions Report for Calendar Year 2014

SF Snyder
JM Barnett

May 2015

U.S. DEPARTMENT OF ENERGY

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

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Pacific Northwest National Laboratory
Richland, Washington 99352

Summary

The U.S. Department of Energy Office of Science (DOE-SC) Pacific Northwest Site Office (PNSO) has oversight and stewardship duties associated with the Pacific Northwest National Laboratory (PNNL) Marine Sciences Laboratory (MSL) located on Battelle Land – Sequim. The facility has two buildings with the potential to emit low levels of radioactive materials. This is the third Radioactive Air Emissions Report for MSL since DOE-SC contracted for exclusive use of its radiological operations effective October 1, 2012. MSL operations remain unchanged from the previous year.

This report is prepared to document compliance with the Code of Federal Regulations (CFR), Title 40, Protection of the Environment, Part 61, National Emission Standards for Hazardous Air Pollutants (NESHAP), Subpart H, "National Emission Standards for Emissions of Radionuclides Other than Radon from Department of Energy Facilities" and Washington Administrative Code (WAC) Chapter 246-247, "Radiation Protection–Air Emissions." Compliance is indicated by comparing the estimated effective dose equivalent (EDE) to the maximally exposed individual (MEI) with the 10 millirem per year (mrem/yr) U.S. Environmental Protection Agency (EPA) standard. The MSL has only fugitive emissions sources. Despite the fact that the regulations are intended for application to point source emissions, fugitive emissions are included with regard to complying with the EPA standard.

The EDE to the MSL MEI due to routine operations in 2014 was 9E-05 mrem (9E-07 mSv). No non-routine emissions occurred in 2014. The MSL is in compliance with the federal and state 10 mrem/yr standard.

For further information concerning this report, you may contact Thomas M. McDermott, U.S. Department of Energy, Pacific Northwest Site Office, by telephone at (509) 372 4675 or by e-mail at tom.mcdermott@science.doe.gov.

CERTIFICATION OF PNNL-22342-3

DOE-SC
Pacific Northwest National Laboratory
Marine Sciences Laboratory
Radionuclide Air Emissions Report
Calendar Year 2014

I certify under penalty of law that I have personally examined and am familiar with the information submitted herein and, based on my inquiry of those individuals immediately responsible for obtaining the information, I believe that the submitted information is true, accurate, and complete. I am aware that there are significant penalties for submitting false information, including the possibility of fine and imprisonment. See, 18 U.S.C. 1001. [verbatim from 40 CFR 61, Subpart H, 61.94(b)(9)]

Roger E. Snyder, Manager
U.S. Department of Energy
Pacific Northwest Site Office

5/15/15
Date

Acronyms and Abbreviations

BL-S	Battelle Land - Sequim
CFR	Code of Federal Regulations
Ci	curie
CY	calendar year
DOE	U.S. Department of Energy
DOE-SC	U.S. Department of Energy, Office of Science
EDE	effective dose equivalent
EPA	U.S. Environmental Protection Agency
HEPA	high efficiency particulate air (filter)
km	kilometer
Major	a radioactive point source having a radiological dose potential of greater than 0.1 mrem/yr EDE, based on emissions that would result if all pollution-control equipment did not exist but facility operations were otherwise normal
MEI	maximally exposed individual
mi	mile(s)
Minor	a radioactive point source having a radiological dose potential of less than or equal to 0.1 mrem/yr EDE, based on emissions that would result if all pollution-control equipment did not exist but facility operations were otherwise normal
mrem	millirem [i.e., 1×10^{-3} rem]
MSL	Pacific Northwest National Laboratory Marine Sciences Laboratory
mSv	millisievert
NA	not applicable
NESHAP	National Emission Standards for Hazardous Air Pollutants
NOC	Notice of Construction
PCM	periodic confirmatory measurement
PNNL	Pacific Northwest National Laboratory
PNSO	Pacific Northwest Site Office
PTE	potential-to-emit
QA	quality assurance
RAEL	Radioactive Air Emissions License
rem	roentgen equivalent man
SD	standard deviation
Sv	sievert
UDF	unit-release dose factor
WAC	Washington Administrative Code
WDOH	Washington State Department of Health
yr	year

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1.0 Introduction

The Pacific Northwest National Laboratory (PNNL) Marine Sciences Laboratory (MSL) is located on Battelle Land-Sequim (PNSO 2013). The U.S. Department of Energy, Office of Science, Pacific Northwest Site Office (DOE-SC PNSO), oversees MSL activities through an exclusive use contract with Battelle Memorial Institute. MSL, DOE's only marine research laboratory, is located on the coast of Washington State's Olympic Peninsula (Figure 1.1).

This radiological air emissions report meets the Washington Department of Health (WDOH) requirements for radiological National Emission Standards for Hazardous Air Pollutants (NESHAP) compliance reporting for the activities at MSL for calendar year (CY) 2014.

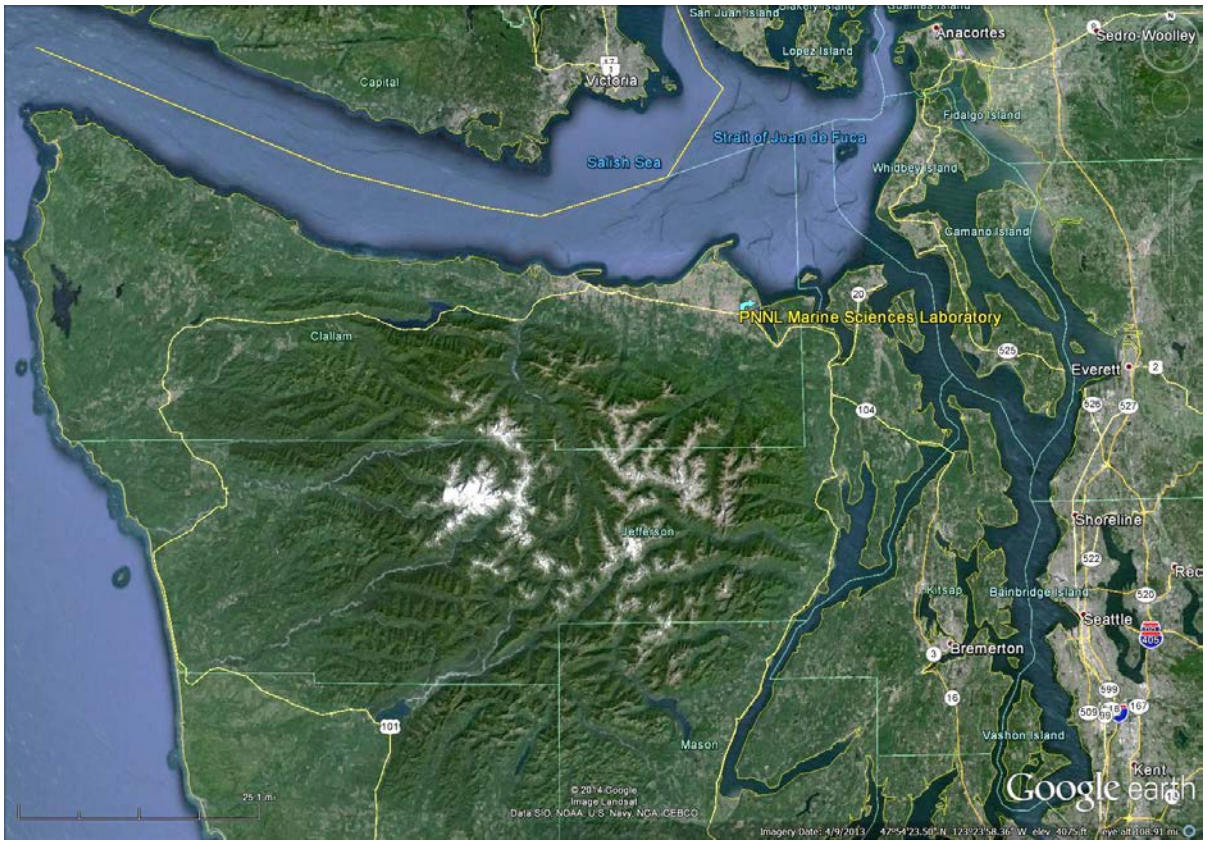

Figure 1.1. MSL in Northwestern Washington State

1.1 Battelle Land – Sequim and MSL Description

Battelle Land-Sequim (Figure 1.2) encompasses 150 acres of uplands and tidelands of which about 7.5 acres has been developed for research operations. The research operations occur at several laboratories and other facilities in an area referred to as MSL, which includes analytical and general purpose laboratories and wet or support laboratories supplied with heated and cooled freshwater and seawater. There are two emission units at MSL with the potential to emit low levels of radioactive material. In addition, MSL has a state-of-the-art waste seawater treatment system and a dock facility for a 28-foot research vessel and a specialized scientific diving boat.

Battelle Land-Sequim on Washington State's Olympic Peninsula is the site of DOE's only marine research laboratory. It lies on the shores of the Strait of Juan de Fuca and is in the rain shadow of the Olympic Mountains in Clallam County at approximate coordinates 48°04'40" N, 123°02'55" W. Despite its coastal location, it receives less than 15 inches of rainfall on average annually. Average monthly temperatures range from 31°F to 70°F. Nearby cities are Sequim (population 6,600), Port Angeles (population 19,000), and Port Townsend (population 9,100) (DOC 2011). Seattle is approximately 50 miles (mi) from MSL. The nearest sea border with Canada is about 17 mi from MSL in the Salish Sea; the nearest Canadian land border is about 25 mi northwest from MSL.

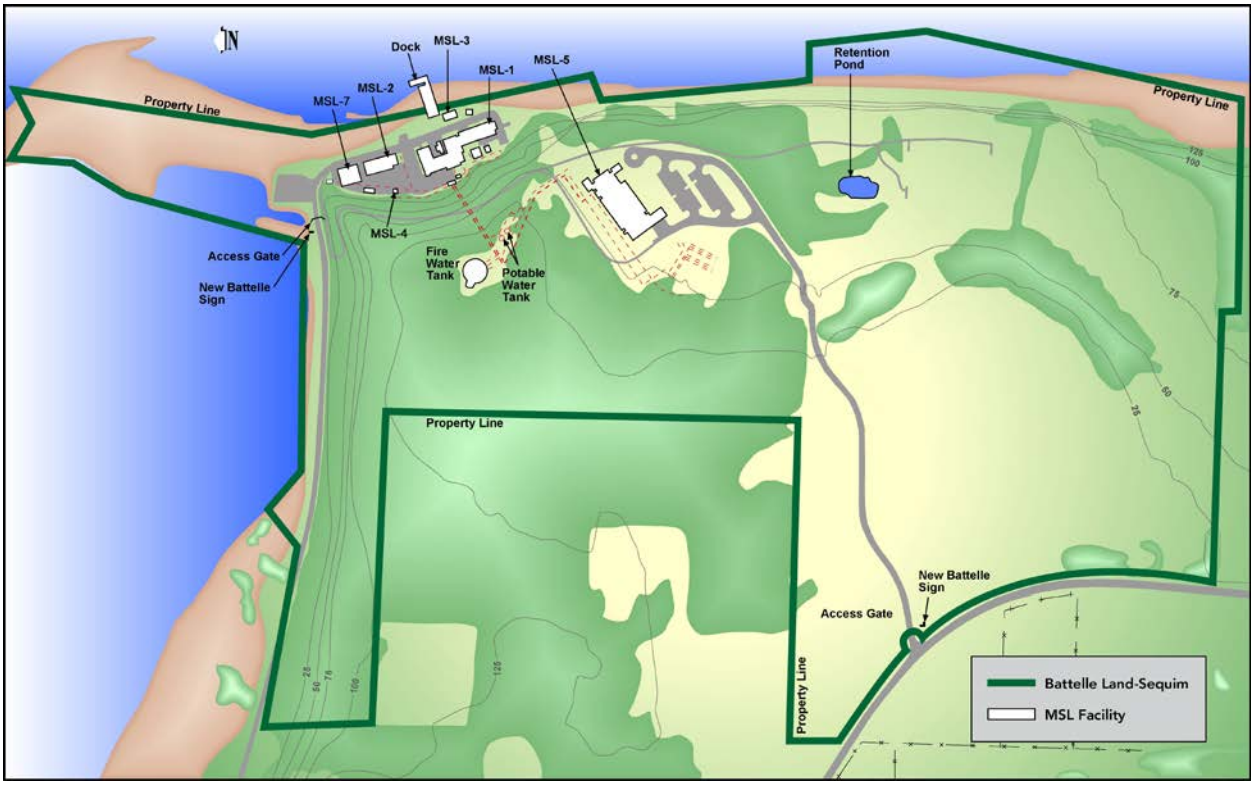

Figure 1.2. Battelle Land-Sequim and Marine Sciences Laboratory

2.0 Radionuclide Air Emissions

The two registered MSL emission units are described and emissions estimates for operations during CY 2014 presented.

2.1 Major, Minor, and Fugitive Emissions Points

Two nonpoint source minor emission units associated with buildings MSL-1 and MSL-5 are registered with the state of Washington under the Radioactive Air Emissions License (RAEL)-014. Radioactive air emissions continue to be well below the criteria for classification as a minor emission unit (i.e., potential-to-emit [PTE] contribution is < 0.1 millirem per year (mrem/yr) effective dose equivalent [EDE] to the MEI). Information regarding the radionuclides-of-concern, emission rates, and emission unit physical characteristics are described below.

The emission units include EP-MSL-1 and EP-MSL-5 (Figure 1.2). EP-MSL-1 is located on the tidelands, and EP-MSL-5 is located on the upland. The emission unit characteristics are the same for both MSL-1 and MSL-5. These buildings have several locations where radioactive air emissions may originate and exit the building. While they are not fugitive by definition, emissions are fugitive in nature; however, because emissions can come from several points within each building, the emission unit is characterized as a nonpoint source (WAC 2014). Emissions from each minor emission unit have an associated PTE of less than 5E-04 mrem/yr EDE that is primarily made up of particulates.

Radiological operations at MSL emit very low levels of radioactive materials. Appendix B contains the full list of radionuclides that may be handled at MSL. The 2014 radioactive material emissions to the air are indicated in Table 2.1. The 40 CFR 61, Appendix D method of determining unabated emissions was used. No credit was taken for abatement controls (e.g., HEPA filtration) at MSL-1 or MSL-5.

Table 2.1. 2014 MSL Inventory and Emissions Estimates

Nuclide	Emission Type	2014 EP-MSL-1 [(a)] (Ci)	2014 EP-MSL-5 [(a)] (Ci)
H-3	beta/gamma	-	1.37E-09
C-14	beta/gamma	-	6.41E-10
K-40	beta/gamma	-	4.78E-12
Fe-55	beta/gamma	-	3.45E-14
Co-57	beta/gamma	-	9.46E-15
Co-60	beta/gamma	-	1.75E-14
Sr-90	beta/gamma	-	8.32E-13
Tc-99	beta/gamma	-	1.70E-10
Ru-106	beta/gamma	-	4.05E-13
Sb-125	beta/gamma	-	5.32E-13
I-129	beta/gamma	-	1.15E-17
Cs-134	beta/gamma	-	3.14E-12
Cs-137	beta/gamma	-	3.72E-11
Eu-152	beta/gamma	-	6.18E-14
Eu-154	beta/gamma	-	1.68E-14
Eu-155	beta/gamma	-	1.77E-14
Pb-210	alpha[b]	-	1.28E-13
Po-208	alpha	-	6.96E-10
Po-209	alpha		1.40E-11
Ra-226	alpha	-	2.98E-13
Ra-228	alpha[b]	-	4.96E-14
Th-228	alpha	-	2.60E-13
Th-230	alpha	-	1.53E-13
Th-232	alpha	-	1.35E-11
U-234	alpha	-	3.33E-11
U-235	alpha	-	1.53E-12
U-238	alpha	-	3.31E-11
U-natural	alpha	3.40E-09	3.40E-09
Pu-238	alpha	-	8.16E-14
Pu-239	alpha	-	3.75E-13
Pu-240	alpha	-	3.75E-13
Am-241	alpha	-	4.34E-13
TOTAL beta/gamma (Ci)		**0.00E+00**	**2.23E-09**
TOTAL alpha (Ci)		**3.40E-09**	**4.19E-09**

(a) Emissions based on 40 CFR 61, Appendix D methods.
(b) Although Pb-210 and Ra-228 are beta-emitters, their decay products include alpha-emitters, therefore they are considered alpha-emitters for dose determination.

3.0 Dose Assessment

The potential impact of MSL radiological air emissions is described in this section. Radiological operations at MSL have not changed from the prior year. A review of radiological assessment needs was published in the Data Quality Objects report (Barnett et al. 2012).

3.1 Dose Model and Potential Receptors

The COMPLY Code version 1.6 (Level 4) was used for estimating dose for comparison to the EPA standard of 10 mrem/yr EDE to any member of the public (40 CFR 61, Subpart H and WAC 246-247). This code is approved for use for compliance determination (40 CFR 61, Appendix E). Input parameters, originally reported in Barnett et al. (2012), were not changed (Table 3.1).

Table 3.1. COMPLY Input Parameters

Parameter	MSL Value (Level 4)
Nuclide names	<varies by year>
Concentrations (Ci/m^3)	NA
Annual possession amount (Ci)	NA
Release rates (Ci/yr *or* Ci/s)	<varies by year>
Release height (m)	8 m
Building height (m)	8 m
Stack or vent diameter (m)	NA
Volumetric flow rate (m^3/s)	NA
Distance from source-to-receptor (m)	190 m[(a)]
Building width (m)	30 m
Wind speed (m/s)	2 m/s
Distances to sources of food production (m)	190 m[(a)]
Stack temperature (°F)	NA
Ambient air temperature (°F)	NA
Wind rose	NA(nwr)[(b)]
Building length	NA(nwr)[(b)]

NA = not applicable
(a) Smallest receptor distance either MSL-1 or MSL-5 applied to both emission units.
(b) NA(nwr) = not applicable because **n**o **w**ind **r**ose data is used.

Potential receptor locations for 16 compass directions are provided in Table 3.2, as reported in Barnett et al. (2012), which concluded that continuation of the 190-m source-to-receptor distance used in prior evaluations would result in an over-estimate of any expected receptor impacts but would continue to be used. The nearest location where a member of the public would actually reside or abide (e.g., dwelling, business, school, office) relative to the MSL-1 or MSL-5 emissions locations was determined to be 270 m W or WNW. Given that winds blow predominantly toward the east (see Table 4.3 of Barnett et al. 2012), away from either of these 270 m receptors, an additional level of conservatism is included.

Table 3.2. Potential MSL MEI Locations

Direction from MSL-1 or MSL-5	Smallest distance to BL-S boundary	Smallest distance to a receptor outside BL-S boundary
N	-	1,790 m res[(a)]
NNE	-	39,700 m res[(a)]
NE	-	9,630 m res[(a)]
ENE	-	2,000 m res[(a)]
E	-	1,900 m res[(a)]
ESE	-	2,620 m res
SE	-	3,930 m res
SSE	-	4,470 m res
S	570 m	640 m res/farm
SSW	630 m	820 m res; 290 m farm
SW	360 m[(a)]	420 m res[(a)]
WSW	230 m	290 m res
W	220 m	270 m res
WNW	230 m	270 m res
NW	280 m	520 m res
NNW	-	1,000 m res/farm

BL-S = Battelle Land-Sequim (see Figure 1.2)
A dash (-) = a shoreline location where no potential receptor could reside or abide.
res = residence site
(a) Distance from MSL-1 applied; all others from MSL-5.

3.2 Compliance Assessment

The dose standard in 40 CFR 61, Subpart H, applies to radionuclide air emissions, other than radon, from DOE facilities. Dose is estimated as the product of the emission rate (Ci/yr) and unit dose factor (mrem/yr at MEI location per Ci/yr released). Unit dose factors for a number of nuclides are indicated in Appendix A. The ^{241}Am unit dose factor was applied to all alpha-emitters and the ^{137}Cs unit dose factor was applied to all beta/gamma emitters, as a conservative measure, except for ^{129}I which used the nuclide-specific dose factor. For CY2014, the MSL MEI location was assumed to be 190 m (0.12 mi) from the emission point. The EDE to the 2014 MEI from routine and non-routine point source emissions was 9E-05 mrem (9E-07 mSv). Table 3.3 shows the relative contributions of each nuclide and facility to the MEI dose. The 2013 MEI estimate was 5E-5 mrem/yr (5E-07 mSv/yr) EDE.

Table 3.3. MSL 2014 Radionuclide Emissions and MEI Dose

	MSL-1	MSL-5	Total
RELEASES (Ci)			
Beta/gamma	0	2.23E-09	**2.23E-09**
Alpha	3.40E-09	4.19E-09	**7.59E-09**
MEI EDE (mrem)			
Beta/gamma[(a)]	0	1.0E-06	**1.0E-06**
Alpha[(b)]	4.0E-05	4.9E-05	**8.9E-05**
Total (mrem)	**4.0E-05**	**5.0E-05**	**9.0E-05**
DOSE CONTRIBUTION (%)			
Beta/gamma	-	2%	**1%**
Alpha	100%	98%	**99%**

(a) Unit dose factor for Cs-137 applied to estimate dose for all nuclide emissions except ^{129}I.
(b) Unit dose factor for Am-241 applied to estimate dose.

Comparing the MSL 2014 MEI dose to average U.S. background radiation (NCRP 2009):

- Annual natural background radiation 310.0 mrem/yr
- Daily natural background radiation 0.85 mrem/d
- Hourly natural background radiation 0.035 mrem/hr
- Per minute natural background radiation 0.00059 mrem/min
- **MSL 2014 MEI dose** **0.000090** **mrem/yr**
- Per second natural background radiation 0.0000098 mrem/sec

4.0 Supplemental Information

This section provides supplemental information related to MSL radionuclide air emissions in 2014. Supplemental information is provided as part of a Memorandum of Understanding between DOE and EPA (DOE 1995). Collective dose information is reported under DOE O 458.1 requirements (DOE 2011).

4.1 Collective Dose Estimate

An estimated 132,000 people (on the U.S. side of the border) live within 30 mi of MSL; another estimated 1.45 million (U.S.) reside 30–50 mi from MSL. The major cities at various distances are indicated in Table 4.1. Victoria, British Columbia is the only major Canadian city within 50 mi of MSL. The Victoria metropolitan area (20–30 mi distant) has an estimated population of 358,000, almost three times the entire U.S. population within 30 mi of MSL.

Table 4.1. Major U.S. Cities within 50 mi of MSL

Distance (mi)	Major Cities
0–10	City of Sequim
10–20	Port Angeles (portion), Port Townsend
20–30	Port Angeles (portion), Oak Harbor
30–40	Anacortes, Bremerton (portion), Edmonds, Mukilteo, Poulsbo, Silverdale, Stanwood
40–50	Arlington, Bainbridge Island, Bothell, Bremerton (portion), Burlington, Edmonds, Everett, Kenmore, Kirkland, Lake Stevens, Lynnwood, Marysville, Mount Vernon, Mountlake Terrace, Port Orchard, Seattle (large portion), Snohomish

The collective dose is simply estimated in a manner that greatly overestimates a more precisely calculated value. The MEI dose multiplied by the 30-mi U.S. population results in a collective dose of 1.2E-2 person-rem. Applying this same method to the Victoria metropolitan area, Canada, all of which is 20–30 mi distant, would result in an additional 3.2E-2 person-rem. The Canadian collective dose is even more greatly overestimated than the US collective dose estimate.

4.2 Compliance Status with Subparts Q and T of 40 CFR 61

- No storage or disposal of radium bearing materials occurs at MSL; therefore, 40 CFR 61, Subpart Q does not apply to MSL operations.
- No uranium mill tailings or ore disposal activities have been conducted at MSL; therefore, 40 CFR 61, Subpart T does not apply to MSL operations.

4.3 Other Supplemental Information

- Periodic confirmatory measurement information is not required by the Notices of Construction (NOCs).
- The PNNL Radioactive Material Tracking system is used to manage potential emissions below permit thresholds resulting in overall confirmation of inventory limits and emissions estimates to respective NOCs.
- Quality assurance (QA) program status of compliance with 40 CFR 61, Appendix B, Method 114 does not apply because no air sampling is conducted at MSL.
- There were no radon emissions in 2014.

5.0 References

40 CFR 61, as amended. *National Emission Standards for Hazardous Air Pollutants* (NESHAP), Subpart H, "National Emission Standards for Emissions of Radionuclides Other than Radon from Department of Energy Facilities."

40 CFR 61, as amended. *National Emission Standards for Hazardous Air Pollutants* (NESHAP), Appendix B, "Test Methods."

40 CFR 61, as amended. *National Emission Standards for Hazardous Air Pollutants* (NESHAP), Appendix D, "Methods for Estimating Radionuclide Emissions."

40 CFR 61, as amended. *National Emission Standards for Hazardous Air Pollutants* (NESHAP), Appendix E, "Compliance Procedures Methods for Determining Compliance with Subpart I."

40 CFR 61, as amended. *National Emission Standards for Hazardous Air Pollutants* (NESHAP), Subpart Q, "National Emission Standards for Radon Emissions from Department of Energy Facilities."

40 CFR 61, as amended. *National Emission Standards for Hazardous Air Pollutants* (NESHAP), Subpart T, "National Emission Standards for Radon Emissions from the Disposal of Uranium Mill Tailings."

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NCRP—National Council on Radiation Protection and Measurements. 2009. *Ionizing Radiation Exposure of the Population of the United States*. NCRP, Bethesda, MD.

PNSO—Pacific Northwest Site Office. 2013. *PNNL Terminology Reference Document*. PNSO-REFR-05, U.S. Department of Energy, PNSO, Richland, WA.

WAC—Washington Administrative Code. 2014. *Radiation Protection – Air Emissions*. WAC-246-247, Statutory Law Committee, Olympia, WA.

Appendix A

COMPLY Unit Dose Factors

Appendix A COMPLY Unit Dose Factors

As originally reported in Barnett et al. 2012, COMPLY v1.6 was used to determine unit-release dose factors (UDFs), which represent impacts to a hypothetical receptor 190 m from the emission unit with an assumed 2 m/s wind speed and wind blowing toward the receptor 25 percent of the time. These assumptions are based on calculations of COMPLY v1.6 at Level 4 with no wind rose used. The appropriate solubility class (DOE 2010) was applied, replacing the DOE 2010 solubility classifications (F,M,S) with the analogous solubility classifications available in COMPLY (D,W, Y, respectively). Several nuclides (^{133}Ba, ^{22}Na, ^{210}Pb, ^{3}H, and ^{14}C) are footnoted to indicate that only one option was available (EPA 1989). Additionally, the more conservative (overestimating) classification was applied to uranium. UDFs for radionuclides either in current inventory or previously used at MSL are presented.

Table A.1. MSL Unit Dose Factors

Nuclide	COMPLY Solubility Class	Unit Dose Factor (mrem EDE per Ci/yr released)
^{241}Am[a]	W	11700
^{133}Ba[b]	D	135
^{14}C[c]	"1"	1.5
^{109}Cd	W	5.5
^{57}Co	W	4.8
^{60}Co	W	426
^{137}Cs[a]	D	469
^{154}Eu	W	345
^{155}Eu	W	13.3
^{3}H[b]	V	0.004
^{125}I	D	84.5
^{129}I	D	1250
^{54}Mn	W	27.2
^{22}Na[b]	D	234
^{63}Ni	W	0.3
^{210}Pb[b]	D	1100
^{238}Pu	W	10300
^{239}Pu	W	11200
^{106}Ru	W	13.9
^{90}Sr[d]	Y	211
^{99}Tc	W	32.7
^{234}U	Y	3450
^{235}U	Y	3470
^{238}U	Y	3110
Natural U[e]	Y	3290

Bold font = Alpha-emitting nuclides. All others are beta/gamma emitters.
(a) ^{241}Am is the surrogate alpha emitter for those not specifically listed; ^{137}Cs is the surrogate beta-emitter for those not specifically listed.
(b) The solubility class listed is the only option available in COMPLY v1.6.
(c) Default class of COMPLY v1.6 used.
(d) Solubility class W is preferred, but not an option. Class Y was used as an over-estimating assumption.
(e) Determined from natural uranium mass fractions: 0.000055 ^{234}U; 0.0072 ^{235}U; 0.9928 ^{238}U (DOE 2009).

Appendix B

List of Radioactive Materials Handled or Potentially Handled, or Authorized for Use at MSL in 2014

Appendix B List of Radioactive Materials Handled or Potentially Handled, or Authorized for Use at MSL in 2014

Table B.2. List of Radioactive Materials Handled or Potentially Handled, or Authorized for Use at MSL in 2014

Ac-225	Bk-249	Cs-134m	Ho-166m	Mo-93	Pm-143	Rh-103m	Ta-182m	U-234
Ac-227	Bk-250	Cs-135	I-122	Mo-99	Pm-144	Rh-104	Ta-183	U-235
Ac-228	Br-82	Cs-136	I-123	Mo-103	Pm-145	Rh-105	Tb-157	U-235m
Ag-108	Br-82m	Cs-137	I-125	Mo-104	Pm-146	Rh-105m	Tb-158	U-236
Ag-108m	Br-83	Cs-138	I-126	Mo-105	Pm-147	Rh-106	Tb-160	U-237
Ag-109m	Br-84	Cs-139	I-128	N-13	Pm-148	Rn-219	Tb-161	U-238
Ag-110	Br-84m	Cs-140	I-129	Na-22	Pm-148m	Rn-220	Tc-95	U-239
Ag-110m	Br-85	Cs-141	I-130	Na-24	Pm-149	Rn-222	Tc-95m	U-240
Ag-111	C-11	Cu-64	I-130m	Na-24m	Pm-151	Rn-224	Tc-97	V-48
Al-26	C-14	Cu-66	I-131	Nb-91	Po-208	Ru-97	Tc-97m	V-49
Al-28	C-15	Cu-67	I-132	Nb-91m	Po-209	Ru-103	Tc-98	W-181
Am-240	Ca-41	Dy-159	I-132m	Nb-92	Po-210	Ru-105	Tc-99	W-185
Am-241	Ca-45	Dy-165	I-133	Nb-93m	Po-211	Ru-106	Tc-99m	W-187
Am-242	Ca-47	Dy-169	I-133m	Nb-94	Po-212	S-35	Tc-101	W-188
Am-242m	Cd-107	Er-169	I-134	Nb-95	Po-213	Sb-122	Tc-103	Xe-122
Am-243	Cd-109	Er-171	I-134m	Nb-95m	Po-214	Sb-124	Tc-106	Xe-123
Am-245	Cd-111m	Es-254	I-135	Nb-97	Po-215	Sb-125	Te-121	Xe-125
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