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under Contract DE-AC05-76RL01830

PNNL-22342-1

Department of Energy – Office of Science
**Sequim Site Radionuclide
Air Emissions Report for
Calendar Year 2012**

SF Snyder
JM Barnett
TL Gervais

April 2013



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Pacific Northwest National Laboratory
Richland, Washington 99352

Summary

The U.S. Department of Energy Office of Science (DOE-SC) Marine Sciences Laboratory (MSL) located on the Sequim Site has two facilities with the potential to emit low levels of radioactive materials. This is the first Radioactive Air Emissions Report for the Sequim Site since DOE-SC contracted for exclusive use of its radiological operations effective October 1, 2012. The operations, which remain unchanged, were regulated previously as a private rather than DOE radiological air emissions facility.

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." This report meets the calendar year 2012 Sequim Site annual reporting requirement for its operations as a privately owned facility as well as its federally contracted status that began in October 2012.

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 contains only sources classified as fugitive emissions. 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 Sequim Site MEI due to routine operations in 2012 was $9\text{E-}06$ mrem ($9\text{E-}08$ mSv). No non-routine emissions occurred in 2012. The MSL is in compliance with the federal and state 10 mrem/yr standard.

For further information concerning this report, you may contact Theresa L. Aldridge, U.S. Department of Energy, Pacific Northwest Site Office, by telephone at (509) 372 4508 or by e-mail at Theresa.Aldridge@pnso.science.doe.gov.

CERTIFICATION OF PNNL-22342-1

DOE-SC Sequim Site Radionuclide Air Emissions Report Calendar Year 2012

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

5/13/13

Date

Acronyms and Abbreviations

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 effective dose equivalent, 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
Minor	a radioactive point source having a radiological dose potential of less than or equal to 0.1 mrem/yr effective dose equivalent, 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]
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

Pacific Northwest National Laboratory's (PNNL) Sequim Marine Research Operations (Sequim Site) was transitioned in October 2012 from private operation under Battelle Memorial Institute to an exclusive use contract with the U.S. Department of Energy, Office of Science, Pacific Northwest Site Office (DOE-SC PNSO). The Marine Sciences Laboratory (MSL) is a sub-region of the Sequim Site, which is located on the coast of Washington State's Olympic Peninsula (**Figure 1.1**).

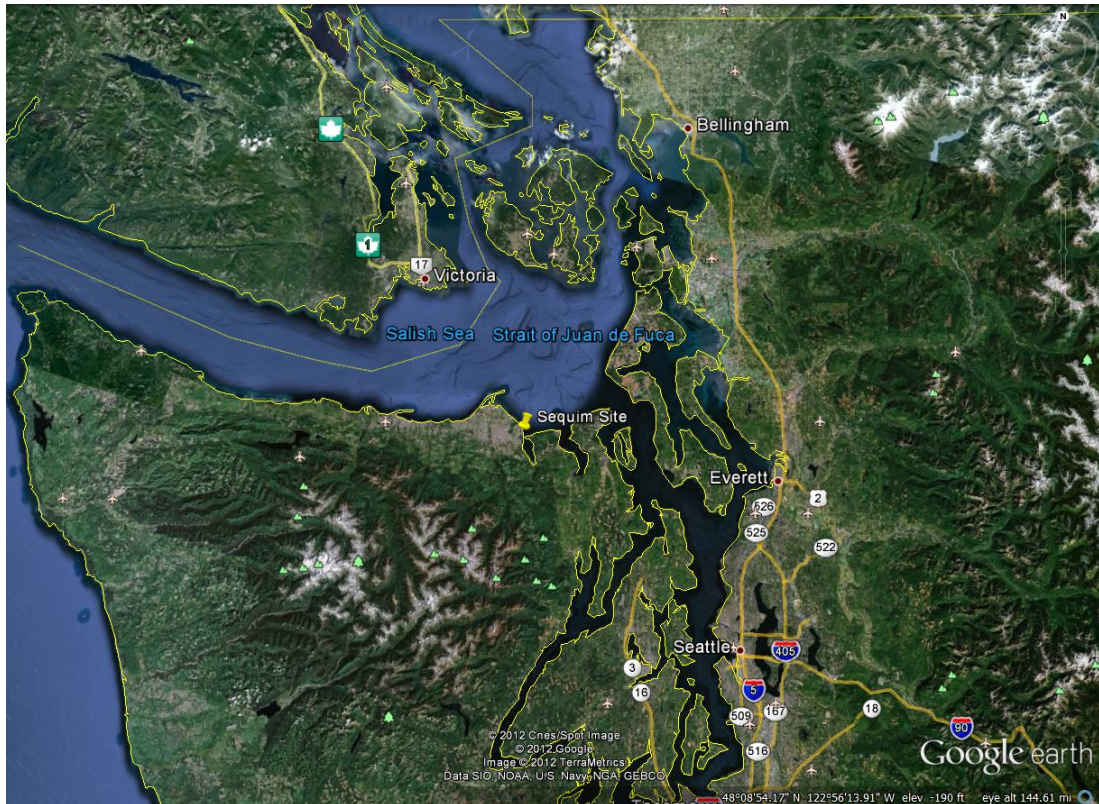


Figure 1.1. Sequim Site in Northwestern Washington State

Per prior discussions with the Washington Department of Health (WDOH), this radiological air emissions report meets the requirements for radiological National Emission Standards for Hazardous Air Pollutants (NESHAP) compliance reporting for both the private operations period and DOE exclusive use contract period for calendar year (CY) 2012.

1.1 Sequim Site Description

The Sequim Site on Washington State's Olympic Peninsula is 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 the Sequim Site. The nearest sea border with Canada is about 17 mi from the Sequim Site in the Salish Sea; the nearest Canadian land border is about 25 mi NW from the Sequim Site.

The Sequim Site (**Figure 1.2**) encompasses 150 acres of uplands and lowlands about 7.5 acres of which has been developed for research operations. The research operations occur at several laboratories and other facilities in an area of the Sequim Site commonly referred to as the MSL, which includes analytical and general purpose laboratories and wet or support laboratories supplied with heated and cooled freshwater and seawater. There are two facilities at the MSL with the potential to emit low levels of radioactive material. In addition, the 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.

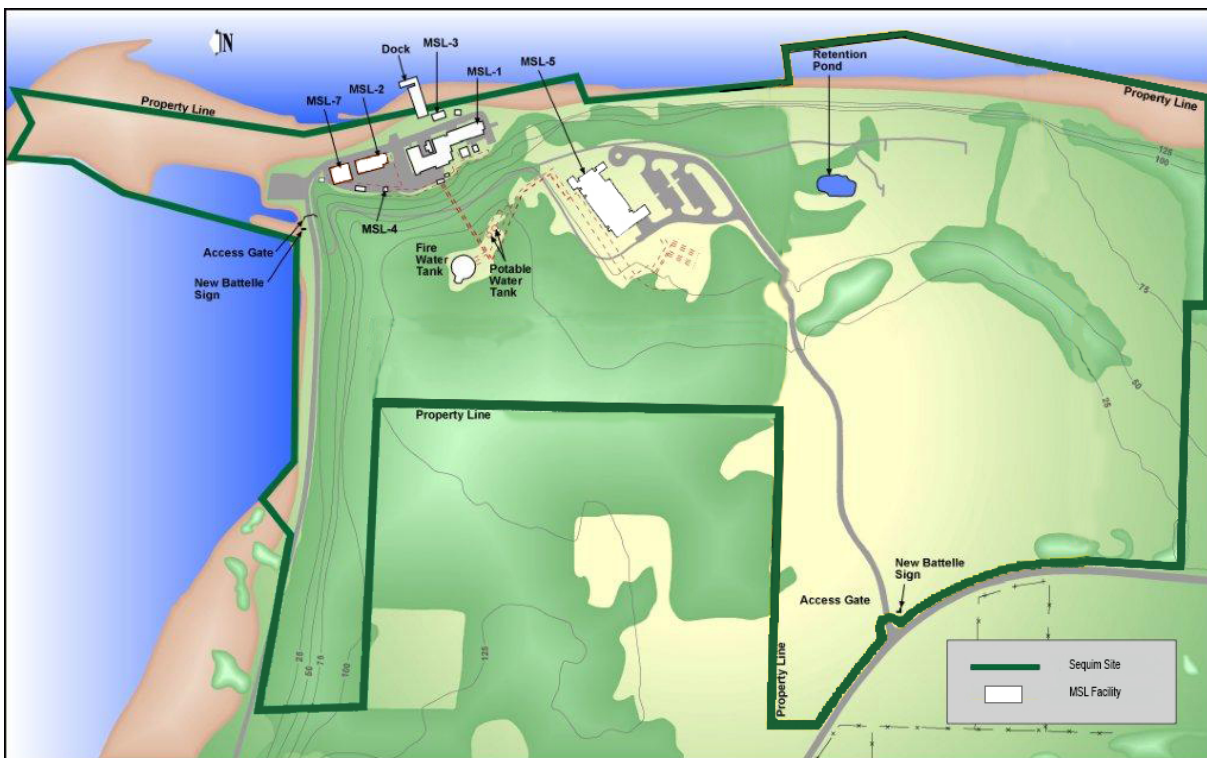


Figure 1.2. Marine Sciences Laboratory, Sequim Site

2.0 Radionuclide Air Emissions

The two registered emission units at the Sequim Site are described and emissions estimates for operations during CY 2012 presented.

2.1 Major, Minor, and Fugitive Emissions Points

The Sequim Site's MSL has two nonpoint source minor emission units associated with buildings MSL-1 and MSL-5 that are registered with the state of Washington under the Radioactive Air Emissions License (RAEL) - 014. These emission units are unchanged from when MSL was licensed as a private facility. 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 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 under consideration for MSL 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 2011). Emissions from each emission unit are identified as < 0.1 mrem/yr EDE and the associated registration PTEs indicate emission unit characteristic will primarily be particulates with building PTEs < 5 E-04 mrem/yr EDE.

Radiological operations at MSL emit very low levels of radioactive materials. [Appendix B](#) contains the full list of radionuclides that may be handled at the Sequim Site. The 2012 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. 2012 MSL Inventory and Emissions Estimates

Nuclide	Emission Type	Site Inventory (Ci)	MSL-1 2012 Release ^(a) (Ci)	MSL-5 2012 Release ^(a) (Ci)
H-3	beta/gamma	1.37E-06	-	1.37E-09
C-14	beta/gamma	6.41E-07	-	6.41E-10
K-40	beta/gamma	4.78E-09	-	4.78E-12
Fe-55	beta/gamma	3.45E-11	-	3.45E-14
Co-57	beta/gamma	9.46E-12	-	9.46E-15
Co-60	beta/gamma	1.75E-11	-	1.75E-14
Sr-90	beta/gamma	8.32E-04	-	8.32E-13
Tc-99	beta/gamma	1.70E-07	-	1.70E-13
Ru-106	beta/gamma	4.05E-10	-	4.05E-13
Sb-125	beta/gamma	5.32E-10	-	5.32E-13
I-129	beta/gamma	1.15E-14	-	1.15E-17
Cs-134	beta/gamma	3.14E-09	-	3.14E-12
Cs-137	beta/gamma	1.35E-06	-	3.72E-11
Eu-152	beta/gamma	6.18E-11	-	6.18E-14
Eu-154	beta/gamma	1.68E-11	-	1.68E-14
Eu-155	beta/gamma	1.77E-11	-	1.77E-14
Pb-210	alpha	1.28E-10	-	1.28E-13
Po-208	alpha	6.96E-07	-	6.96E-10
Ra-226	alpha	2.98E-10	-	2.98E-13
Ra-228	alpha	4.96E-11	-	4.96E-14
Th-228	alpha	2.60E-10	-	2.60E-13
Th-230	alpha	1.53E-07	-	1.53E-13
Th-232	alpha	2.56E-10	-	2.56E-13
U-234	alpha	1.20E-09	8.33E-13	3.71E-13
U-235	alpha	5.58E-11	3.81E-14	1.77E-14
U-238	alpha	1.18E-09	8.28E-13	3.52E-13
Pu-238	alpha	8.16E-11	-	8.16E-14
Pu-239	alpha	7.48E-09	-	3.75E-13
Pu-240	alpha	3.75E-10	-	3.75E-13
Am-241	alpha	4.34E-10	-	4.34E-13
TOTAL beta/gamma			0.00E+00	2.06E-09
TOTAL alpha			1.70E-12	6.99E-10

(a) Emissions estimated using 40 CFR 61, Appendix D methods.

3.0 Dose Assessment

The potential impact of radiological air emissions is described in this section. Dose is calculated in a similar manner to that done for prior years when the MSL was not a DOE facility. Radiological operations at the facility have not changed. 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 no wind rose 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 MEI Locations

Direction from MSL-1 or MSL-5	Smallest distance to MSL boundary	Smallest distance to Sequim Site boundary	Smallest distance to a receptor outside of Sequim Site 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	180 m	-	4,470 m res
S	170 m	570 m	640 m res/farm
SSW	190 m	630 m	820 m res; 290 m farm
SW	170 m	360 m ^(a)	420 m res ^(a)
WSW	140 m	230 m	290 m res
W	130 m	220 m	270 m res
WNW	140 m	230 m	270 m res
NW	170 m	280 m	520 m res
NNW	240 m	-	1,000 m res/farm
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 EDE at MEI location per Ci/yr released). Unit dose factors for a number of nuclides are indicated in [Appendix A](#). The ²⁴¹Am unit dose factor was applied to all alpha-emitters and the ¹³⁷Cs unit dose factor was applied to all beta/gamma emitters, as a conservative measure. For CY2012, the Sequim Site MEI location was assumed to be 0.190 km (0.12 mi) from the emission point. The EDE to the MEI from routine and non-routine point source emissions was 9.2E-06 mrem (9.2E-08 mSv). Table 3.3 shows the relative contributions of each nuclide and facility to the MEI dose. In 2011, the MEI estimate was 1.2E-9 mrem/yr EDE.¹ Although both the 2012 and 2011 dose estimates are far below the dose standard, the primary reason for the increase in the 2012 dose estimate is that no credit was taken for HEPA filtration in the 2012 emissions estimates as per the new DOE-SC radioactive air emissions license.

Table 3.3. Sequim Site 2012 Radionuclide Emissions and MEI Dose

	MSL-1	MSL-5	Total
RELEASES (Ci)			
Beta/gamma	0	2.06E-09	2.06E-09
Alpha	1.70E-12	6.99E-10	7.01E-10
MEI EDE (mrem)			
Beta/gamma ^(a)	0	9.7E-07	9.7E-07
Alpha ^(b)	2.0E-08	8.2E-06	8.2E-06
Total (mrem)	2.0E-08	9.1E-06	9.2E-06
DOSE CONTRIBUTION (%)			
Beta/gamma	0%	11%	11%
Alpha	100%	89%	89%
(a) Unit dose factor for ¹³⁷ Cs applied to estimate dose.			
(b) Unit dose factor for ²⁴¹ Am applied to estimate dose.			

¹ Memo from M. Sula (Sequim Site, Sequim, WA) to M. Barnett (PNNL, Richland, WA). 2012. "2011 NESHAP Airborne Radionuclide Emission Assessment" 27FEB2012, Battelle-Marine Sciences Laboratory, Sequim, WA.

4.0 Supplemental Information

This section provides supplemental information related to Sequim Site radionuclide air emissions in 2012. Supplemental information was requested as part of a Memorandum of Understanding between DOE and EPA (DOE 1995).

4.1 Population Dose Estimate

An estimated 132,000 people (on the U.S. side of the border) live within 30 mi of Sequim, WA; another estimated 1.45 million (U.S.) reside 30–50 mi from Sequim. 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 the Sequim Site. 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 the Sequim Site.

Table 4.1. Major U.S. Cities within 50 mi of the Sequim Site

Distance (mi)	Major Cities
0–10	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 population dose is simply estimated. The MEI dose multiplied by the 30-mi U.S. population results in a population dose of $1.2\text{E-}3$ person-rem. Applying this same method to the Victoria metropolitan area, all of which is 20–30 mi distant, would result in an additional $3.3\text{E-}3$ person-rem.

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

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

4.3 Other Supplemental Information

- Radionuclide emission estimates and periodic confirmatory measurement information related to Notices of Construction (NOCs) is not used.
- 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. No air sampling is conducted at the Sequim Site; therefore, the QA program compliance status with 40 CFR 61, Appendix B, Method 114 does not apply.

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), Subpart H, Appendix B to Part 61, “Test Methods.”
- 40 CFR 61, as amended. *National Emission Standards for Hazardous Air Pollutants* (NESHAP), Subpart H, Appendix D to Part 61, “Methods for Estimating Radionuclide Emissions.”
- 40 CFR 61, as amended. *National Emission Standards for Hazardous Air Pollutants* (NESHAP), Subpart H, Appendix E to Part 61, “Compliance Procedures Methods for Determining Compliance with Subpart I.”
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- EPA—U.S. Environmental Protection Agency. 1989. *User’s Guide for the COMPLY Code*. EPA 520/1-89-003, U.S. Environmental Protection Agency, Office of Radiation and Indoor Air, Washington, D.C.

WAC—Washington Administrative Code. 2011. *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 to apply was based on those indicated in DOE 2010, and for ^{14}C the COMPLY default classification was applied as the only option (EPA 1989). UDFs for radionuclides either in current inventory or previously used at the Sequim Site are presented.

Table A.1. Sequim Site 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
^3H ^(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 the Sequim Site in 2012

Appendix B List of Radioactive Materials Handled or Potentially Handled, or Authorized for Use at the Sequim Site in 2012

Table B.1. List of Radioactive Materials Handled or Potentially Handled, or Authorized for Use at the Sequim Site in 2012

Ac-225	C-14	Eu-152	K-42	Pb-210	Rb-86	Sn-126	Tl-209
Ac-227	C-15	Eu-152m	Kr-81	Kr-83m	Rb-87	Sr-85	Tm-170
Ac-228	Ca-41	Eu-154	Kr-81m	Kr-85	Rb-88	Sr-87m	Tm-171
Ag-108	Ca-45	Eu-155	Kr-90	Kr-85m	Rb-89	Sr-89	U-232
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