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# Analysis and Recommendation of Tritium Gas Continuous Air Monitor Alarm Setpoints for the RPL Stack Exhaust

September 2021

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

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# **Abstract**

The Radiochemical Processing Laboratory (RPL) tritium continuous air monitor (CAM) is used for near real-time detection of tritium gas (i.e., elemental tritium [HT] and tritiated water vapor [HTO]) in the stack exhaust. The CAM interfaces with software developed by the Pacific Northwest National Laboratory (PNNL)—called the "PNNL OS3700 Tritium Monitoring Software"—that provides near real-time estimates of tritium gas air concentrations and integrated daily activities that are calculated from measured CAM counts. The OS3700 software implements alarm setpoints to alert the facility of larger tritium gas releases that if allowed to persist, could begin to challenge permitted emission and established dose constraints.

This report performs a detailed review of historical and current tritium alarm setpoints used at RPL, including discussion of the technical basis used in their development, analysis of alarm frequencies using measured historical data, and performs a detailed dose assessment using more realistic release scenarios and meteorology. Based on the results, the tritium air concentration and integrated daily tritium activity alarm setpoints will remain  $2.0 \times 10^{-5} \,\mu\text{Ci/ml}$  and 25 Ci/day, respectively. These setpoints achieve the right operational balance in identifying larger releases from planned tritium work at RPL, without being overly conservative so as to cause nuisance alarming. Furthermore, implied doses associated with these setpoints are well below defined and regulatory limits.

Abstract

# **Acronyms and Abbreviations**

ALARA as low as reasonably achievable

ANSI American National Standards Institute

Bq becquerel (i.e., one nuclear disintegration per second)

CAM continuous air monitor

Ci curie (equals 3.7 x 10<sup>10</sup> Bq)

DOE U.S. Department of Energy

EPA U.S. Environmental Protection Agency

HT elemental tritium
HTO tritiated water vapor
HPS Health Physics Society

ICRP International Commission on Radiological Protection

MEI maximally exposed offsite individual

mrem millirem (i.e., 1 x 10<sup>-3</sup> rem)

NCRP National Council on Radiation Protection and Measurements

NRC U.S. Nuclear Regulatory Commission
PNNL Pacific Northwest National Laboratory

rem unit of dose equivalent (Roentgen equivalent man)

RPL Radiochemical Processing Laboratory

scfm standard cubic feet per minute
WAC Washington Administrative Code

WDOH Washington State Department of Health

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# 1.0 Introduction

Pacific Northwest National Laboratory (PNNL) operates the Radiochemical Processing Laboratory (RPL [325 Building]) for the U.S. Department of Energy at the Hanford Site, Washington. The RPL is a three-story building, with over 13,000 m² of space consisting of offices, laboratories, and specialized areas such as high bays, hot cells, shops, and waste treatment units. The mission of the RPL is to create and implement innovative processes for environmental cleanup and the beneficial use of radioactive materials, including ways to advance the cleanup of radiological and hazardous wastes, processing and disposal of nuclear fuels, and production and delivery of medical isotopes. Projects conducted in the facility frequently change. Consequently, inventories of radioactive material in the building also change, and can include microgram-to-kilogram quantities of fissionable materials and up to megacurie quantities of other radionuclides.

RPL is classified as a major emission point (Barnett and Snyder 2018). Environmental regulations (40 CFR 61, Subpart H [2002]) require the use of continuous air monitors (CAM) for real-time monitoring of radioactive particulates and tritium gas (i.e., elemental tritium [HT] and tritiated water vapor [HTO]) in the exhaust from the main stack. A six-nozzle rake probe that spans the stack diameter is used to obtain a representative exhaust sample at the well-mixed location. The sample probe is connected to a transport line that extends downward and terminates in the facility sample room where it enters an EG&G Berthold LB150D Alpha-Beta CAM (Berthold 1993 [Berthold Technologies U.S.A. LLC, 99 Midway Lane, Oak Ridge, TN 37830]). A portion of that flow (i.e., 235 mL/min) is diverted to an EG&G-Berthold LB110 Tritium CAM (Berthold 2009), where it is first mixed with a P-10 counting gas (i.e., 10 percent methane, 90 percent argon) at a 1:4 ratio to maximize detection sensitivity and minimize interference from other radionuclide decays. The CAM operates in conformance with American National Standards Institute/Health Physics Society (ANSI/HPS) N13.1–2011 (HPS 2011) standard requirements; additional information about the RPL emission unit and CAM system can be found in the "PNNL Facility Radionuclide Emission Points and Sampling Systems" report (Barnett and Snyder 2018).

The LB-110 CAM is a windowless, flow-through proportional detector with pulse rise-time discrimination (Barnett et al. 2004). The discriminator divides individual ionization events into short rise-time events and long rise-time events. The pulse rise-time is dependent on the drift time required by the electrons generated in the primary ionization track to traverse the counting chamber. Drift time depends on the length and course of the primary ionization track, which is a function of the type and energy of the particle causing the ionization. Short rise-time events are calibrated to tritium (Channel A), while long rise-time events are currently calibrated to <sup>85</sup>Kr (Channel B). The detector does not distinguish between the tritium pulse counts of HT and HTO.

The tritium pulse counts are sent to a data acquisition card, which is accessed and analyzed by software developed by PNNL—called the "PNNL OS3700 Tritium Monitoring Software" (Rishel et al. 2019). The OS3700 software has user-defined parameters that allow background radiation levels to be subtracted and detector efficiency and counting periods to be defined. These parameters are used by the software to calculate real-time tritium air concentrations and integrated daily activities that are compared to alarm setpoints. Permit requirements only necessitate setpoints for the tritium emissions, therefore Channel B has no alarm setpoints and is not discussed further herein. The tritium air concentration alarm setpoint is used to provide an early indication of a larger release that if allowed to persist, could begin to approach established

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dose constraints. The integrated daily tritium activity alarm setpoint is used to serve as a timely warning that if occurring routinely, could begin to approach permitted emission limits. The current OS3700 tritium alarm setpoints have been used operationally at RPL since 2006. Because permitted tritium work activities at RPL have changed over time, the alarm setpoints, and their associated technical basis, are being reviewed to verify that they are still appropriate and meet programmatic needs.

This report is organized into five sections, including this introduction. Section 2 documents the historical and current alarm setpoints, including a discussion of the technical basis used in their development. Section 3 evaluates the effectiveness of the current alarm setpoints by examining alarm frequency using 5-years of measured historical data. Section 4 provides a detailed dose assessment to include more realistic release scenarios and meteorology used in the calculation of dose factors. This section also reviews reported annual tritium emissions and doses for RPL from the continuous sampling program to verify the effectiveness of the current tritium alarm setpoints. Section 5 provides a summary and recommendation on appropriate tritium CAM alarm setpoints to use at RPL.

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# 2.0 Tritium CAM Alarm Setpoints

This section documents the original and current RPL tritium stack CAM alarm setpoints as well as the associated technical basis.

# 2.1 Original Tritium Stack CAM Alarm Setpoints

The original RPL tritium stack CAM alarm setpoints are documented in a Facilities and Operations report, "Tritium Monitor Alarm Setpoint Revision Radiochemical Processing Facility Exhaust Stack" (Sula 2000; [Appendix A]). According to the report, the original (1994) tritium concentration alarm setpoint was set to 40  $\mu$ Ci/m³ over a 3-minute running average period, which was based on "as low as reasonably achievable" (ALARA) considerations; this results in a 0.5 Ci release at normal stack flow (i.e., 140,000 scfm).

The tritium alarm setpoints were revised in 2000 to accommodate a significant increase in tritium inventory under the "Tritium Target Qualification Program." Under that program, the RPL tritium inventory increased substantially and alarm setpoints based solely on ALARA considerations no longer met programmatic needs. Thus, new dose-based alarm setpoints that were tempered with ALARA-based considerations were developed.

According to the 2000 Sula report, a hypothetical 20-Ci release at a normal stack flow of 140,000 scfm over a 2-hour period ( $40~\mu\text{Ci/m}^3$  average concentration) could lead to a 2-mrem dose to the hypothetical maximally exposed offsite individual (MEI) using conservative model assumptions. The 2-mrem dose constraint was based on U.S. Department of Energy requirements (DOE 1990) cited in the RPL Facility Effluent Monitoring Plan (Shields 1999); this value is 20 percent of the 10 mrem annual standard (40~CFR 61, Subpart H [2002]; Washington Administration Code Chapter 246-247 [2019]). The report did not state the assumptions used in the dose modeling, but it did note that for such a dose to be realized, the release would have to be HTO and take place during local crop harvest because the primary exposure pathway would be deposition uptake by foodstuffs.

For ALARA considerations, the assumed release was reduced to 10 Ci over a running 2-hour period at normal flow ( $20 \,\mu\text{Ci/m}^3$  average concentration). In addition, a 24-hour (midnight-to-midnight) 25 Ci alarm setpoint would serve as a timely warning, that if occurring routinely over multiple days, could begin to approach the annual emission permit limit. Table 1 summarizes the 1994 and 2000 tritium CAM alarm setpoints and the technical basis.

Table 1. Original Tritium Alarm Setpoints (Sula 2000; see Appendix A)

Year	Alarm Description	Alarm Setpoint	Technical Basis
1994	3-minute running average concentration	40 μCi/m <sup>3</sup>	ALARA
2000	2-hour running average concentration	20 μCi/m <sup>3</sup>	Dose/ALARA
2000	24-hour (midnight-to-midnight) integrated release	25 Ci	Permit

# 2.2 Current Tritium Stack CAM Alarm Setpoints

In 2006, new tritium CAM software—called the PNNL OS3700 Tritium Monitoring System Software—was made operational at RPL (Barnett et al. 2005¹). In short, the OS3700 software converts the measured total (gross) tritium counts from the detector to real-time estimates of air concentration and integrated daily activity, which have accompanying alarm setpoints. A description of the process used by the OS3700 software to calculate the air concentration and integrated stack activity follows.

The tritium air concentration is calculated by subtracting the tritium background counts from the total gross counts measured during a 1-minute count interval and dividing by the detector efficiency and sampled volume (see Eq. 1) (Barnett et al. 2005; Rishel et al. 2019):

$$Trit_{conc} = \frac{c_{total} - c_{bkg}}{DE \times Volume \times (37,000)} \tag{1}$$

where:

 $Trit_{conc}$  = tritium air concentration ( $\mu$ Ci/ml) measured over a 1-minute count interval

 $C_{total} = \text{total gross tritium count rate (counts per second) measured over a 1-$ 

minute count interval

 $C_{bkq}$  = tritium background count rate (counts per second); treated as a fixed value

throughout the year based on annual CAM calibration measurements

DE = tritium detector efficiency (unitless); treated as a fixed value throughout the

year based on annual CAM calibration measurements

Volume = sampled volume (ml)

 $37,000 = \text{conversion factor to convert counts (Bq) to } \mu\text{Ci.}$ 

The daily integrated stack activity is calculated by multiplying the 1-minute tritium concentration (Eq. 1) by the 1-minute stack volume and summing the resultant activity over a day (i.e., a day extending from midnight-to-midnight; 1440 minutes) (Eq. 2):

$$Trit_{activity} = \sum_{i=1}^{1440} Trit_{conc_i} \times Stack Volume_i$$
 (2)

where:

 $Trit_{activity}$  = integrated tritium activity ( $\mu$ Ci)

 $Trit_{conc_i}$  = tritium concentration ( $\mu$ Ci/ml) in the i<sup>th</sup> 1-minute count interval

(1440 minutes in one day)

 $Stack\ Volume_i$  = stack volume (mL) over a 1-minute count interval.

Table 2 lists the current OS3700 alarm setpoints; these have been in use since the software became operational in 2006 and reflect the alarm setpoints in Table 1 from 2000, with the following implementation differences:

• The tritium concentration (Eq. 1) is conservatively averaged over a 5-minute rolling average instead of 2-hours; the 5-minute running average concentration is compared to the concentration setpoint every minute.

<sup>&</sup>lt;sup>1</sup> The OS3700 software was updated in 2019 (Rishel et al. 2019) to run on more recent operating systems, however the software functionally is identical to the 2006 version.

• The integrated activity (Eq. 2) is compared with the 24-hour (midnight-to-midnight) integrated release setpoint every 3 minutes; the integrated activity is automatically reset to 0.0 Ci at midnight.

Table 2. Current Tritium Alarm Setpoints Used in the OS37000 Software

Year	Alarm Description	Alarm Setpoint Verification Frequency	Documented Alarm Setpoint	Consideration(s)
2006	5-minute running average concentration	1 minute	2.0 × 10 <sup>-5</sup> μCi/ml	Dose/ALARA
2006	24-hour (midnight-to- midnight) integrated release	3 minute	25 Ci	Permit

Note: For ease of implementation, the concentration setpoint was conservatively reduced by 5 percent from  $2.1 \times 10^{-5} \,\mu\text{Ci/ml}$  (21  $\mu\text{Ci/m}^3$ ) to  $2.0 \times 10^{-5} \,\mu\text{Ci/ml}$  (20  $\mu\text{Ci/m}^3$ ). Technically, this results in a 9.5 Ci release (instead of 10 Ci) over a running 2-hour period at normal flow (140,000 scfm).

# 3.0 Evaluation of Tritium CAM Alarm Setpoints

This section evaluates the current (Table 2) alarm setpoints by evaluating alarm frequency using 5 years (2014–2018) of historical CAM data. Ideally, operational setpoints should identify larger releases that may require further attention while avoiding nuisance alarming.

# 3.1 Alarm Frequency

As discussed in Section 2.2 (Table 2), the OS3700 software uses two tritium alarm setpoints—air concentration and integrated daily activity. The tritium air concentration setpoint  $(2.0 \times 10^{-5} \,\mu\text{Ci/ml})$  is designed to alert the facility of a larger-than-normal release, that if allowed to persist up to 2 hours, could begin to approach conservatively defined ALARA- and dose-based objectives. The integrated daily activity alarm setpoint (25 Ci) is designed to inform the facility of larger daily emissions that if occurring routinely, could begin to consume the annual permit emission limit (currently 1,600 Ci over the calendar year period).

Alarm frequency was evaluated by comparing the tritium air concentration and daily integrated activity alarm setpoints against 5 years (2014–2018) of historical CAM data measured at RPL. Additionally, a maximum concentration ( $4.2 \times 10^{-5} \, \mu \text{Ci/ml}$ ) setpoint was evaluated; this setpoint reflects the 40-Ci release from the Sula (2000) report that was conservatively identified as an upper-limit tritium release that could begin to approach a 2-mrem acute dose to a member of the public. Table 3 lists the evaluated alarm setpoints along with the associated count frequency (i.e., "Count") and unique alarm days (i.e., "Days") for each year evaluated. Count refers to the number of 1-minute alarms (and therefore alarm minutes) that occurred in that year. Days is the number of unique days that had at least one alarm. In all cases, the alarms listed in Table 3 were associated with planned tritium work; there were no instances of unplanned alarms.

Table 3. Concentration and Integrated Activity Alarm Setpoints Count Frequency and Unique Alarm Days by Year (2014–2018)

Parameter	Setpoint	20	14	20	15	20	16	20	17	20	18
Concentration	(µCi/ml)	Count	Days								
Current	2.0 × 10 <sup>-5</sup>	1,162	9	54	4	48	5	21	2	80	8
Maximum	4.2 × 10 <sup>-5</sup>	202	8	3	1	9	2	0	0	22	4
Integrated Activity	(Ci)	Count	Days								
Current	25	805	2	0	0	0	0	0	0	0	0

As can be seen in Table 3, the current 1-minute concentration alarm setpoint results in alarms every year, with a count frequency ranging from 21 (2017) to 1,162 (2014). Unique alarm days ranged from 2 (2017) to 9 (2014). If the concentration alarm setpoint were set to the maximum value, the count frequency drops substantially, ranging from 0 (2017) to 202 (2014), and the unique alarm days also are reduced, ranging from 0 (2017) to 8 (2014). Although the current concentration setpoints result in more frequent 1-minute alarms (1,162 versus 202), thereby conservatively identifying work resulting in lower tritium emissions, the total number of unique alarm days (9 versus 8) is not appreciably different, meaning both concentration setpoints identify days where planned tritium work results in larger releases.

The 3-minute daily integrated activity alarm only activated in 2014, resulting in a total of 805 alarm-minutes, on two unique days. The near absence of this alarm indicates tritium work at

RPL is at sufficiently low levels so permitted annual emission limits are not challenged. As will be shown in Section 4.1, continuous tritium sampling, which is performed independently of the tritium CAM monitoring, is used to establish emissions for annual reporting; these data readily confirm compliance with the annual emission limit of 1,600 Ci and supports the low occurrence of this alarm setpoint.

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# 4.0 Tritium Dose Assessment

The acute dose calculations used to support the setpoints listed in Table 1 and Table 2 are not formally documented in the Sula (2000) report; therefore, this section includes a detailed dose assessment for completeness. As noted in Section 2.1, the original methodology considered a hypothetical 20-Ci release at RPL's normal stack flow of 140,000 scfm over a 2-hour period; the report stated this could result in a dose of 2 mrem, which met the desired constraint of 20 percent of the annual 10-mrem standard. For ALARA objectives, the assumed release was reduced to 10-Ci, with an implied dose no greater than 1 mrem. The concentration alarm setpoints in Table 1 and Table 2 reflect a 10-Ci release. The original dose assessment was based on "... historical meteorological data, environmental transport and radionuclide fate models." Furthermore, it was also noted that the predominant exposure pathway is ingestion through deposition uptake of HTO by foodstuffs; however, analysis details and references were not provided.

An RPL radiation dose safety analysis report (Cathey 2013) provides acute dose factors for a hypothetical 1-Ci tritium release over a range of release durations (3- to 120-minutes), release conditions (thermal buoyancy from fires), 2013 meteorology, updated dose coefficients from the ICRP (2001) for inhalation and from the Environmental Protection Agency Federal Guidance Report 13 (2002) for air/ground submersion, and the use of a different dispersion model (Chanin and Young 1998). Tritium was assumed to be released as HTO and the resulting dose coefficients were multiplied by 1.5 to account for skin absorption. Ingestion was not considered because this pathway can be effectively prevented by implementing protective measures (e.g., intervention to prevent consumption of affected crops and other foodstuffs). The report provides 95th percentile (i.e., worst-case meteorology) safety dose factors to the hypothetical MEI (570 m downwind).

The acute dose factors can be used to provide a conservative estimate of dose from the current tritium concentration alarm setpoint. Dose can be calculated using Eq. 3:

$$Dose (mrem) = Activity (Ci) \times Dose Factor \left(\frac{mrem}{Ci}\right)$$
 (3)

Table 4 lists the 95<sup>th</sup> percentile tritium dose factors from Cathey (2013) and the resulting 95<sup>th</sup> percentile doses (mrem) calculated using Eq. 3 and assuming a 10-Ci tritium release. The implied 95<sup>th</sup> percentile doses are at least a factor of 10 below the 2-mrem dose constraint for all release scenarios. Release scenarios 10 and 12 most closely match the hypothetical 2-hour release duration used in the derivation of the concentration setpoint; these scenarios result in an implied dose of 6.28 × 10<sup>-3</sup> mrem and 5.20 × 10<sup>-3</sup> mrem, respectively. These doses, which reflect conservative meteorology, are well below the original 1-mrem dose calculated by Sula (2000). It is hypothesized the Sula (2000) dose factor reflects an extremely conservative ground-level release, whereas the Cathey (2013) dose factors represent more probable releases, which in some cases include thermal plume buoyancy.

Tritium Dose Assessment

Table 4. RPL 95<sup>th</sup> Percentile Tritium Dose Factors Over a Range of Release Scenarios (Cathey 2013) and the Implied Acute Dose Using a 10-Ci Tritium Release

Release	Release		95 <sup>th</sup> Percentile	Implied 95 <sup>th</sup>
Scenario	Duration	Heat Release	Dose Factor	Percentile Dose
(#)	(min)	(MW)	(mrem/Ci)	(mrem)
1	3	0	2.19 × 10 <sup>-3</sup>	2.19 × 10 <sup>-2</sup>
2	15	0	1.72 × 10 <sup>-3</sup>	1.72 × 10 <sup>-2</sup>
3	15	0.1	1.60 × 10 <sup>-3</sup>	1.60 × 10 <sup>-2</sup>
4	15	1	1.13 × 10 <sup>-3</sup>	1.13 × 10 <sup>-2</sup>
5	30	1	1.08 × 10 <sup>-3</sup>	1.08 × 10 <sup>-2</sup>
6	60	1	1.03 × 10 <sup>-3</sup>	1.03 × 10 <sup>-2</sup>
7	15	5	$7.99 \times 10^{-4}$	$7.99 \times 10^{-3}$
8	30	5	7.50 × 10 <sup>-4</sup>	$7.50 \times 10^{-3}$
9	60	5	$6.87 \times 10^{-4}$	$6.87 \times 10^{-3}$
10	120	5	6.28 × 10 <sup>-4</sup>	6.28 × 10 <sup>-3</sup>
11	60	10	4.67 × 10 <sup>-4</sup>	4.67 × 10 <sup>-3</sup>
12	120	10	5.20 × 10 <sup>-4</sup>	5.20 × 10 <sup>-3</sup>

The acute dose factors considered thus far represent worst-case meteorological conditions (i.e., 95<sup>th</sup> percentile diffusion factors) that normally are used in safety analyses. These dose factors are intended to be very conservative because they are derived from meteorological conditions that are only expected 5 percent of the time. Acute dose factors also can be calculated using more realistic (e.g., average) meteorological conditions. For example, the U.S. Nuclear Regulatory Commission uses 50th percentile dose factors when evaluating dose impacts in environmental reports submitted as part of an application for a permit, license, or other authorization to site, construct, and/or operate a new nuclear power plant (NRC 2018). The RPL tritium CAM does not perform a safety function for the facility; its purpose is to provide an early indication of a larger-than-normal release to the environment for compliance purposes. Thus, the use of average acute dose factors for environmental releases is reasonable.

The Cathey (2013) report only provides  $95^{th}$  percentile dose factors for use in safety analyses. However, the report's calculation package provides mean dose factors as well. Table 5 lists the mean dose factors and implied mean dose (mrem) resulting from 10 Ci over a running 2-hour period at normal flow (i.e., the current concentration alarm setpoint of  $20 \, \mu \text{Ci/m}^3$ ). The implied mean doses are further reduced—100 to 1000 times lower than the 2-mrem dose constraint using this mean dose factor. Again, release scenarios 10 and 12 in Table 5 most closely match the implied 2-hour release duration used in the derivation of alarm setpoints; these scenarios result in an implied mean dose of  $1.02 \times 10^{-3}$  mrem and  $7.43 \times 10^{-4}$  mrem, respectively, to the nearest member of the public and are well below the 2-mrem dose constraint used in the development of the setpoint.

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Table 5.	RPL Mean Tritium Dose Factors Over a Range of Release Scenarios (Cathey 2013)
	and the Implied Acute Dose Using a 10-Ci Tritium Release

Release Scenario (#)	Release Duration (min)	Heat Release (MW)	Mean Dose Factor (mrem/Ci)	Implied Mean Dose (mrem)
1	3	0	4.51 × 10 <sup>-4</sup>	4.51 × 10 <sup>-3</sup>
2	15	0	4.06 × 10 <sup>-4</sup>	$4.06 \times 10^{-3}$
3	15	0.1	3.34 × 10 <sup>-4</sup>	3.34 × 10 <sup>-3</sup>
4	15	1	1.81 × 10 <sup>-4</sup>	1.81 × 10 <sup>-3</sup>
5	30	1	1.75 × 10 <sup>-4</sup>	1.75 × 10 <sup>-3</sup>
6	60	1	1.73 × 10 <sup>-4</sup>	1.73 × 10 <sup>-3</sup>
7	15	5	1.21 × 10 <sup>-4</sup>	1.21 × 10 <sup>-3</sup>
8	30	5	1.16 × 10 <sup>-4</sup>	1.16 × 10 <sup>-3</sup>
9	60	5	1.13 × 10 <sup>-4</sup>	1.13 × 10 <sup>-3</sup>
10	120	5	1.02 × 10 <sup>-4</sup>	1.02 × 10 <sup>-3</sup>
11	60	10	8.31 × 10 <sup>-5</sup>	8.31 × 10 <sup>-4</sup>
12	120	10	7.43 × 10 <sup>-5</sup>	7.43 × 10 <sup>-4</sup>

#### 4.1 Additional Dose Considerations

As discussed in Section 2.1, the original alarm setpoints were developed assuming a hypothetical short-duration (i.e., 2-hour) tritium gas release that would result in an implied dose less than 2-mrem to the hypothetical MEI; this value is 20 percent of the 10 mrem annual standard (40 CFR 61, Subpart H [2002]; Washington Administration Code Chapter 246-247 [2019]). As shown in the previous section, the current tritium concentration alarm setpoint would result in significantly lower doses than the 2-mrem dose constraint. The DOE public dose limit is 100 mrem/year (DOE 2020).

The 2-mrem acute dose constraint is consistent with the Nuclear Regulatory Commission dose standard, which limits the handling and use of radioactive materials such that no member of the public will receive a radiation dose of 2-mrem in any 1 hour period from external radiation sources in an unrestricted area (10 CFR 20, Subpart D [1991]). For context, the ICRP Publication 103 (2007) recommends exposures to the general public in planned exposure situations should be limited to no more than 100-mrem annually (ICRP 2007). Indeed, the average annual radiation dose per person in the United States is 620 mrem (NCRP 2009), of which 228 mrem is from natural background (i.e., radon and thoron).

As noted in Section 2.2, the current tritium concentration and integrated daily activity alarm setpoints have resulted in alarms at RPL during planned tritium work. However, as the dose assessment demonstrates, implied acute doses would remain well below the 2-mrem dose constraint. Continuous tritium sampling is performed separately at RPL to confirm that annual emissions are within permitted limits. Table 6 lists sampled tritium (HT and HTO) emissions and resulting doses for the last 7 years, including the 5-year period (2014–2018) used in Section 3.1 to evaluate alarm frequency.

Tritium Dose Assessment

From Table 6, it is clear 2014 has the highest total tritium emission (650 Ci) and associated annual dose (0.277 mrem). Table 3 showed 2014 also had the highest alarm frequencies. Thus, the CAM alarm setpoints are correctly identifying larger tritium releases that are separately reflected in the annual sampling results. Overall, Table 6 demonstrates that total annual emissions are well below 1,600-Ci annual permit limit and doses are well below 2-mrem dose constraint, thereby confirming the current alarm setpoints are meeting the desired objectives.

Table 6. Annual RPL Tritium (HT and HTO) Emissions and Associated Dose for 2014–2020

Year	HT Emissions (Ci)	HTO Emissions (Ci)	Total Emissions (Ci)	HT Dose (mrem)	HTO Dose (mrem)	Total Tritium Dose (mrem)
2014a	325	325	650	0.089	0.188	0.277
2015 <sup>b</sup>	133	282	415	0.020	0.047	0.067
2016 <sup>c</sup>	24	240	264	0.003	0.035	0.038
2017 <sup>d</sup>	16	160	176	0.002	0.026	0.028
2018 <sup>e</sup>	83	250	333	0.012	0.040	0.052
2019 <sup>f</sup>	87	170	257	0.013	0.029	0.042
2020 <sup>g</sup>	49	200	249	0.006	0.029	0.035

a DOE/RL-2015-12 (2015)

<sup>&</sup>lt;sup>b</sup> DOE/RL-2016-10 (2016)

<sup>°</sup> DOE/RL-2017-17 (2017)

d DOE/RL-2018-05 (2018)

e DOE/RL-2019-09 (2019)

f DOE/RL-2020-08 (2020)

<sup>&</sup>lt;sup>9</sup> DOE/RL-2021-12 (2021)

# 5.0 Summary and Recommendations

This report documents the historical and current tritium stack CAM alarm setpoints used at the RPL. Since 2000, two alarm setpoints have been used—tritium air concentration and daily integrated tritium activity out the stack. The tritium air concentration alarm setpoint is used to provide an early indication of a larger release that if allowed to persist, could begin to approach established dose and ALARA objectives. The integrated daily tritium activity alarm setpoint is used to serve as a timely warning of elevated emissions that if occurring routinely over many days, could begin to approach permitted annual emission limits.

Since 2006, alarming has been performed by PNNL-developed software called the "PNNL OS3700 Tritium Monitoring Software" (Barnett et al. 2005; Rishel et al. 2019). The OS3700 software takes pulse counts measured from the EG&G-Berthold LB110 Tritium CAM and calculates real-time air concentrations and integrated daily activities, and the values then are compared to defined setpoints for alarming purposes. The current tritium air concentration and integrated daily tritium activity alarm setpoints are  $2.0 \times 10^{-5} \,\mu\text{Ci/ml}$  and  $25 \,\text{Ci}$ , respectively.

The current alarm setpoints were evaluated against 5 years (2014–2018) of historical CAM data measured at RPL. Additionally, a maximum concentration (4.2 × 10<sup>-5</sup> μCi/ml) setpoint was evaluated that reflects an "upper-limit" tritium release hypothesized in the initial report that established the RPL tritium alarm setpoint technical basis (Sula 2000). It was shown that although the current concentration alarm setpoint results in more alarms as compared to the maximum hypothesized concentration setpoint, the total number of unique alarm days (9 days versus 8 days) is not appreciably different, meaning both the current (2.0 × 10<sup>-5</sup> μCi/ml) and maximum  $(4.2 \times 10^{-5} \, \mu \text{Ci/ml})$  concentration setpoints were effectively identifying the same days when planned tritium work resulted in larger releases; there were no unplanned tritium alarm events over the evaluation period. Importantly, the current concentration alarm setpoint is not set so low as to cause nuisance alarming on days when low-level tritium work is occurring. The evaluation also showed that the integrated daily activity alarm only activated on two unique days, and the near absence of this alarm indicates tritium work at RPL is at sufficiently low levels so permitted annual emission limits would not be challenged. This latter conclusion was confirmed by examining continuous tritium sampling data for RPL, which demonstrates annual emissions are well below the annual emission limit of 1,600 Ci.

An assessment of implied acute dose resulting from the current concentration alarm setpoint was performed using more recent dose factors from an RPL radiation dose and safety analysis report (Cathey 2013). It was shown that both extremely conservative safety (95<sup>th</sup> percentile) and more realistic environmental (mean) dose factors resulted in low acute doses, with implied doses 100 to 1000 times lower than the targeted 2-mrem dose constraint. Beyond using more up-to-date modeling methods and data, the significant reduction in acute dose using the dose factors determined by Cathey (2013) is attributed to thermal plume buoyancy as opposed to a ground-level release hypothesized in the Sula (2000) report. Annual tritium doses independently calculated from continuous sampling data for RPL further demonstrates tritium emissions result in very low doses.

In summary, the current tritium alarm setpoints are more than adequate at meeting permitted emission and established dose limits at RPL. Furthermore, the setpoints are effective at identifying larger releases without causing nuisance alarming. Therefore, the current tritium alarm setpoints (Table 2) are recommended for continued use at RPL, as they are meeting the stated objectives for current and expected future tritium work activity. Indeed, as the emission and dose evaluations demonstrate, there is sufficient margin to increase the setpoints should the need arise to accommodate additional tritium work.

# 6.0 References

10 CFR 20, Subpart D. 1991. *Standards for Protection Against Radiation*. Code of Federal Regulations, U.S. Nuclear Regulatory Commission, Washington, D.C.

40 CFR 61, Subpart H. 2002. *National Emission Standards for Emissions of Radionuclides other than Radon from Department of Energy Facilities*. Code of Federal Regulations, U.S. Environmental Protection Agency, Washington, D.C.

Barnett JM, LA True, and DD Douglas. 2004. *Review of Tritium Emissions Sampling and Monitoring from the Hanford Site Radiochemical Processing Laboratory*. In: Proceedings of the HPS 2004 Midyear Meeting – Air Monitoring and Internal Dosimetry, February 8-11, 2004, Augusta, GA. McLean, VA: Health Physics Society; 199-204.

Barnett JM, CJ Duchsherer, DR Sisk, DM Carrell, DD Douglas, and GL Carter. 2005. *PNNL OS3700 Tritium Monitoring System Software and Hardware Operations Manual, Revision 0*. PNNL-15491, Pacific Northwest National Laboratory, Richland, Washington.

Barnett JM and SF Snyder. 2018. Pacific *Northwest National Laboratory Facility Radionuclide Emission Points and Sampling Systems*. PNNL-15992, Revision 4, Pacific Northwest National Laboratory, Richland, Washington.

Berthold Technologies. 2009. *Tritium Monitor LB 110*. Id. No.: 80872 BA2. Rev. No.: 02. Bad Wildbad, Germany.

Cathey NG. 2013. *Radiation Dose Analysis for the RPL DSA and Implementing Procedures*. RPL-SA-R4, Revision 5, Pacific Northwest National Laboratory, Richland, Washington.

Chanin D, and ML Young. 1998. *Code Manual for MACCS2: User's Guide*. NUREG/CR-6613, Volume 1. U.S. Nuclear Regulatory Commission, Washington, District of Columbia.

DOE/RL-2015-12, Rev. 0. 2015. *Radionuclide Air Emissions Report for the Hanford Site, Calendar Year 2014*. U.S. Department of Energy, Richland Operations Office, Richland, Washington.

DOE/RL-2016-10, Rev. 0. 2016. *Radionuclide Air Emissions Report for the Hanford Site, Calendar Year 2015*. U.S. Department of Energy, Richland Operations Office, Richland, Washington.

DOE/RL-2017-17, Rev. 0. 2017. *Radionuclide Air Emissions Report for the Hanford Site, Calendar Year 2016*. U.S. Department of Energy, Richland Operations Office, Richland, Washington.

DOE/RL-2018-05, Rev. 0. 2018. *Radionuclide Air Emissions Report for the Hanford Site, Calendar Year 2017*. U.S. Department of Energy, Richland Operations Office, Richland, Washington.

DOE/RL-2019-09, Rev. 0. 2019. *Radionuclide Air Emissions Report for the Hanford Site, Calendar Year 2018*. U.S. Department of Energy, Richland Operations Office, Richland, Washington.

References 13

DOE/RL-2020-08, Rev. 0. 2020. *Radionuclide Air Emissions for the Hanford Site, Calendar Year 2019*. U.S. Department of Energy, Richland Operations Office, Richland, Washington.

DOE/RL-2021-12, Rev. 0. 2021. *Radionuclide Air Emissions Report for the Hanford Site, Calendar Year 2020*. U.S. Department of Energy, Richland Operations Office, Richland, Washington.

EG&G Berthold. 1993. *Alpha-Beta-Aerosol-Monitor LB 150 D* (Operating Manual). Laboratorium Prof. Dr. Berthold, Wildbad 1, Germany.

ICRP (International Commission on Radiological Protection). 2001. *The ICRP Database of Dose Coefficients: Workers and Members of the Public, Version 2.0.1.* Elsevier, New York, New York.

ICRP (International Commission on Radiological Protection). 2007. *The 2007 Recommendations of the International Commission on Radiological Protection*. Elsevier, Oxford, United Kingdom.

National Council on Radiation Protection and Measurements (NCRP). 2009. *Ionizing Radiation Exposure of the Population of the United States*. NCRP Report No. 160, Bethesda, Maryland.

Pacific Northwest National Laboratory (PNNL). 2006. 300 Area Pacific Northwest National Laboratory Facility Radionuclide Emission Points and Sampling Systems. PNNL-15992, Pacific Northwest National Laboratory, Richland, Washington.

Rishel JP, JM Barnett, GL Pace, EB Dutcher, and GL Carter. 2019. *PNNL OS3700 Tritium Monitoring System Software and Hardware Operations Manual*. PNNL-15491-1. Pacific Northwest National Laboratory, Richland, Washington.

Shields KD. 1999. Facility Effluent Monitoring Plan for the 325 Radiochemical Processing Laboratory. PNNL-12157, Pacific Northwest National Laboratory, Richland, Washington.

Sula MJ. 2000. *Tritium Monitor Alarm Setpoint Revision Radiochemical Processing Facility Exhaust Stack*. Unpublished report, Pacific Northwest National Laboratory, Richland, Washington.

- U.S. Department of Energy (DOE). 1990. *Radiation Protection of the Public and the Environment*. DOE Order 5400.5, Washington, D.C.
- U.S. Department of Energy (DOE). 2020. *Radiation Protection of the Public and the Environment*. DOE Order 458.1, Washington, D.C.
- U.S. Nuclear Regulatory Commission (NRC). 2018. *Preparation of Environmental Reports*. Regulatory Guide 4.2, Revision 3, Washington, D.C.

Washington Administrative Code. 2019. *Radiation Protection – Air Emissions*. Chapter 246-247. Olympia, Washington.

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# **Appendix A – Tritium Monitor Alarm Setpoint Revision Radiochemical Processing Facility Exhaust Stack (2000)**

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# TRITIUM MONITOR ALARM SETPOINT REVISION RADIOCHEMICAL PROCESSING FACILITY EXHAUST STACK

#### Summary

Since 1994, a monitor has continuously measured RPL exhaust stack tritium concentrations and activated a facility alarm whenever tritium concentrations exceeded 40  $\mu$ Ci/m³ over a three minute averaging period (i.e., a 0.5 Ci release at normal stack flow). This alarm point, which is well within the '20 Ci per two hour' criteria in the RPL Facility Effluent Monitoring Plan, was based on ALARA considerations at a time when relatively little tritium was being used in the facility.

Tritium Target Qualification Program (TTQP) work scheduled for summer 2000 is expected to bring 88,000 Ci of tritium into the RPL and generate short-term tritium releases of up to several curies during routine work<sup>1</sup>. Consequently, the current ALARA based alarm setpoint is no longer appropriate.

In response to expected tritium releases during forthcoming TTQP work, Effluent Management is reconfiguring the tritium monitor alarm function to avoid alarm activation during normal TTQP operations while continuing to use the monitor-alarm as an environmental ALARA tool. Under the new configuration, an alarm will be generated within one minute following a 10 Ci release within the previous two-hour period or if 25 curies are released within a calendar day. The new alarm conditions thus consider both acute and protracted release situations.

#### **Emission Monitoring Requirements**

Battelle environmental requirements specify continuous emission monitoring for any system with a potential of greater than once per year to exceed 20% of the 10 mrem/y offsite emission dose standard. Two acute tritium releases from TTQP work at the RPL over the past two years were theoretically capable of exceeding 2 mrem, although actual doses were much less than 2 mrem. Thus, it is concluded that tritium monitoring under the FEMP plan is required for TTQP work in the RPL<sup>3</sup>.

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<sup>&</sup>lt;sup>1</sup> An agreement between Battelle and the State of Washington (TTQP Notice of Construction, AIR 00-208, 2/23/2000) establishes maximum emission rates for TTQP work at 1,800 curies/year.

<sup>&</sup>lt;sup>2</sup> Per the RPL Facility Effluent Monitoring Plan (FEMP; PNNL-12157). This requirement is based on guidance in the Department of Energy's Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance (DOE/EH-0173T).
<sup>3</sup> Note that neither the State (TTQP Notice of Construction) nor the RPL Safety Analysis Report require continuous stack tritium monitoring during TTQP operations. The Tritium Target Qualification Program Notice of Construction (TTQP-NOC) does not specifically require continuous tritium emission monitoring, nor does it acknowledge that continuous monitoring will be performed during the project.

#### Tritium Monitor Function

The RPL stack tritium monitor measures the concentration of tritium over successive intervals of time (update interval) as programmed by the user, and displays the average concentration for each update interval as well as the moving average of the concentration over a user defined long-term averaging period. The monitor can also provide the accumulated release over successive 24-hour periods (e.g., midnight to midnight) but cannot effectively accumulate releases over long periods of time (i.e., annual release quantities), or accumulate releases associated with specific events.

The tritium monitor does not distinguish between the various forms of tritium in the stack exhaust.

#### Basis for Revised Alarm Setpoints:

The revised tritium monitor alarm setup considers both acute and protracted releases.

Acute Release - The RPL Facility Effluent Monitoring Plan (FEMP) prescribes detection of sudden releases that could result in an offsite emission dose (hypothetical maximum exposed individual) of 2 mrem (i.e., 20% of annual dose standard). A "sudden" release has historically been defined as one lasting less than 2 hours. Based on historical meteorological data, environmental transport and radionuclide fate models predict that a short duration release of 20 curies could cause a dose of 2 mrem. Thus, 20 Ci is the upper bound release for which an alarm is desired. ALARA considerations provide the basis for seeking a lower alarm level (e.g., 10 Ci) if this can be reasonably achieved.

Protracted Release - The 25 curie/day release alarm provides facility personnel with a timely warning of emissions that, if allowed to continue, could significantly consume the 1800 curie annually allotted tritium release specified in the TTQP Notice of Construction. Visual checks of the tritium monitor by operations personnel each workday normally provide an adequate means to identify low-level but continuous type releases. However, facility management is concerned that releases occurring during extended weekends when operations checks were not performed could significantly erode the allotment. The 25 curie/day accumulated release alarm point would, under worst case conditions, annunciate a release condition prior to an accumulated release of 150 curies (<10% of the annual allotment). This scenario assumes the release begins shortly after the daily inspection on the day before a four-day weekend and the alarm activates shortly before inspection on the day following the weekend. In terms of potential offsite dose, a protracted release of 150 curies would contribute approximately 0.013 mrem to a maximum exposed individual's dose from Hanford operations.

The RPL SAR states that a continuous tritium monitor will be operated when required by State and Federal regulations.

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<sup>&</sup>lt;sup>4</sup>. The two-hour period was chosen for consistency with acute release scenario assumptions used in the RPL Safety Analysis Report. The two-hour release period is used because dose impacts from releases lasting longer than two hours are reduced due to natural variability in wind direction over longer periods of time.

<sup>&</sup>lt;sup>5</sup> However, for such a dose to be realized, the release must be of tritiated water vapor (HTO) and take place during local crop harvest, since the primary exposure pathway is via deposition/uptake by foodstuffs.

#### Summary of Revised Tritium Monitor Alarm Settings

The tritium monitor will alarm in the event of:

- > acute release of 10 Ci or more of tritium within any running two-hour period, or
- > accumulated release of 25 Ci or more of tritium during any calendar day.

The alarms are established by the following monitor settings:

Acute Release of 10 Ci in 2 Hours:	Tritium Monitor Alarm Settings Alarm concentration = 20 μCi/m3 Averaging period = 120 minutes Update period = 1 minutes.
Accumulated Release of 25 Ci in 24 Hours:	Tritium Monitor Alarm Settings Stack flow rate = 140,000 cfm Accumulation Interval = 24 hours Interval sync. = 24 hour Quantity = 25 Ci

The acute alarm settings will activate an alarm within one minute of accumulating a release of 10 curies within a two-hour period (120-minute moving average exceeds 20  $\mu$ Ci/m³). Release scenarios that would activate the tritium monitor acute release alarm using these settings are illustrated in Figure 1.

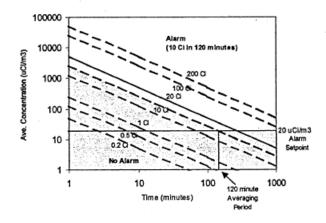


Figure 1. Release Scenarios at Recommended Acute Alarm Setpoint

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#### Appendix A.

#### Explanation of Tritium Monitor Alarm Setpoints

Two user definable parameters affect the way in which tritium concentration is measured by the monitor. These are:

- · concentration averaging period, and
- measurement update interval.

The basic formula for relating stack concentration and release over period of time T = toct1, is:

$$R = CxExTx1E-6$$

where:

 $R = Curies released from t_0 \zeta t_1$ ,

C = average concentration (μCi/m³) during t<sub>0</sub>ςt<sub>1</sub>, and

E = stack exhaust rate (m³/min), and

T = period (min).

For the RPL stack, which has a relatively constant flow rate of 4,000 m3/min (140,000 cfm), the formula is:

$$R(Ci) = 0.004 \times C \times T$$

and for a release of 20 Ci (minimum acute alarm criteria):

$$5,000 \mu \text{Ci-min/m}^3 = \text{C} \times \text{T}$$

Since our performance criteria constrain T to from 0 - 2 hours, the minimum average concentration that could result in a 20 Ci release over the range of possible time periods is

$$C_{min} = 5000 = 40 \,\mu\text{Ci/m}^3$$
.

The relationship between R, C, and T is shown graphically in Figure A.1.

If the alarm concentration setpoint is tied to a moving average, the alarm condition will be tested each time the average is updated; that is, the update interval establishes the time between reaching a 20 Ci release and activation of the alarm.

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