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Subject License Amendment Request for Tritium Dose Coefficients

## 1 Executive Summary

An experiment upgrade at the NBSR will result in the slow production of tritiated deuterium gas (DT) in a closed system. The equilibrium activity of the experiment after 60 years is 2.8 kCi ( $\beta^-$ , 18.6 keV). A review of the FSAR has indicated that the experiment upgrade meets criterion §50.59(viii), and thus requires a license amendment. The method of evaluation in question is the ICRP-66 dose coefficient for tritium gas, which has been adopted by the Department of Energy (DOE) and is used at other national laboratories per Federal Guidance Report (FGR)-13. The proposed LAR would solicit NRC approval to adjust the Derived Air Concentration (DAC) for HT in Part 20 Appendix B to the ICRP-66 values in accordance with §20.1302(c) for the new cold neutron source to be installed in 2023. This will be documented in Chapter 11 of the FSAR.

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## 2 Background

The NBSR is a 20 MW test reactor on the NIST campus in Gaithersburg, MD. The majority of the 2,000+ annual guest researchers who visit the laboratory use cold neutron beams (<5 meV) for nondestructive experiments in material science and condensed matter physics. The cold neutron moderators are

aluminum chambers filled with liquid hydrogen that reside in permanent thimbles adjacent to the reactor vessel.

In 2023, the laboratory will enter an extended shutdown to perform major upgrades, including the installation of a third-generation cold neutron source, Unit 3. The aluminum chamber volume will increase by a factor of 7 to accommodate the change to liquid deuterium, which has a larger neutron scattering length but lower absorption cross section than hydrogen. As a result, the neutron beam intensity at low energies is expected to increase by a factor of 2 or more. Similar to previous generations of cold sources, there is no credible accident scenario by which the reactor could be damaged by a cold source malfunction.

## 2.1 §50.59 Determination for the Unit 3 Upgrade

The buildup of DT gas was considered when performing the §50.59 evaluation for the Unit 3 upgrade. A review of the FSAR was conducted to determine whether an existing methodology could be applied to analyze a DT release from an experimental apparatus failure.

- In Chapter 11 of the FSAR [1], DTO evaporation from D<sub>2</sub>O primary coolant is evaluated using EPA Comply, a simple script that evaluates whether slow, continuous releases meet annual EPA requirements. This code is not suitable for short-term releases.
- Also in Chapter 11, the Part 20 Appendix B DAC for tritium is used as a benchmark for DTO buildup in the building due to primary coolant evaporation when the ventilation is turned off. However, the bulk of the new expansion volume will be located outdoors.
- In Chapter 13 [2], Hotspot is used to evaluate fission product release accidents. However, low-level tritium releases are not evaluated in this chapter and Hotspot's dose conversion factor for elemental tritium is based on ICRP-66, which conflicts with the Part 20 Appendix B DAC.

**Determination 1:** It was determined that the most appropriate tool to evaluate the DT release would be Hotspot, but this would meet criterion §50.59(c)(2)(viii), *“a departure from the method of evaluation used...in the safety analyses,”* for existing tritium releases in Chapter 11.

**Determination 2:** It was further determined that the ICRP-66 dose coefficient for HT is more appropriate than the Part 20 Appendix B DAC for this problem. §20.1302(c) allows the licensee to seek approval for this adjustment (discussed further below). However, the mechanism by which approval should be sought is unclear. A license amendment in conjunction with **Determination 1** has been identified as the most straightforward path.

## 2.2 System Technical Description

During normal operation, the bulk of the deuterium is a liquid at 23 K located in a thimble on the reactor vessel. When the refrigerator shuts down, the liquid evaporates into two expansion volumes, or ballast tanks, located outside confinement. The small ballast tank (2 m<sup>3</sup>) presently used with Unit 2 is stored in D-200, an independently ventilated room that evacuates to the stack. To accommodate the larger inventory of deuterium needed for Unit 3 a second volume, the large ballast tank (16 m<sup>3</sup>), will be installed in parallel with the small ballast tank. The large ballast tank will be located outdoors, outside the guide hall, 100 ft away from the stack.

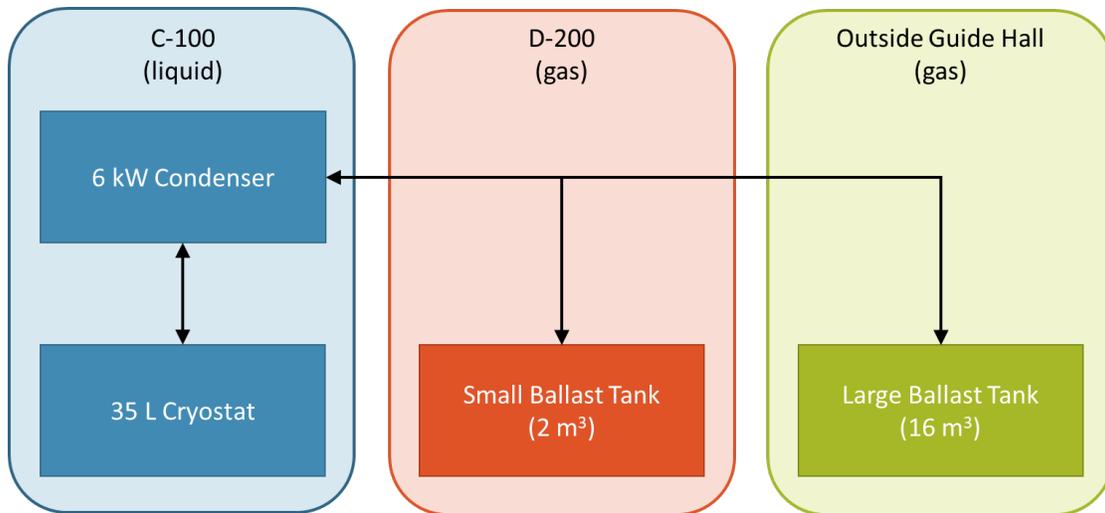


Figure 1. Block Diagram of the Unit 3 Deuterium Assembly.

The system is designed for passive safety: in the shutdown condition, liquid deuterium evaporates freely and the final, warm pressure of deuterium gas will be only 5 bar. This allows the system to be constructed with no relief valves as long as there are also no operable isolation valves between the cryostat and the ballast tanks. The code variances for NFPA 2 required to exclude pressure relief and isolation valves have been approved by the site AHJ at NIST (Roby Sagar, Fire Protection Group Leader). Because there are no relief valves, it is highly unlikely that the deuterium gas will ever escape the system. For reference, in 25 years of operation of the two existing hydrogen cold sources, the NCNR has never lost a gram of gas.

While the reactor is running, the liquid deuterium will activate by the  ${}^2\text{H}(n,\gamma){}^3\text{H}$  reaction with a half-life of 12.5 years. For a continuous operating schedule at 20 MW, the equilibrium activity would be 2.8 kCi after 60 years (this is impossible due to refueling and maintenance outages). Figure 2 depicts the continuous buildup estimate, as well as a more realistic 38.5/10 day operating cycle (2.3 kCi); but for conservatism, only the continuous scheme was used in the safety analyses. This conservatism is expected to provide a convenient margin for additional DT inventory in the future without the need for a separate safety analysis.

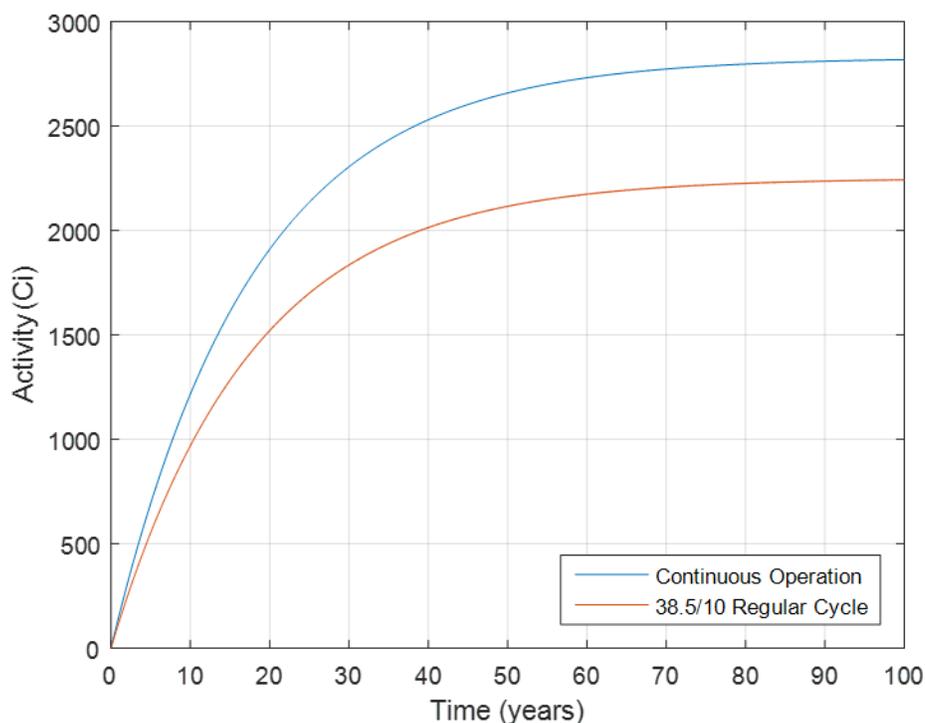


Figure 2. Tritium buildup as a function of time.

### 3 Safety Analysis of Outdoor DT Release

It was postulated that a major facility incident like a crane malfunction could cause an unanticipated release of DT gas. Numerous precautions are being taken to mitigate this, including a no-fly zone for the building crane, steel shielding around exposed components, concrete bollards outside, broad procedural requirements for maintenance activities, and signage at all access points. Thus, an “unspecified outdoor release” of warm DT gas was identified as the bounding safety concern.

#### 3.1 Dose Conversion Factors

DT dispersion is readily modeled using Hotspot [3], a gaussian plume simulator developed by LLNL. Per FGR-13 [4], which incorporates ICRP-66 guidance [5], Hotspot is able to discriminate between DT, DTO, and partial fractions of each. This contradicts NRC guidance for HT in Part 20 Appendix B, which is to use the DAC for 100% DTO regardless of chemical composition. The ICRP-66 guidance is based on research by Peterman, who investigated the uptake of DT and DTO in the lungs of mice and human volunteers, and found a difference in internal dose factors on the order of  $10^4$  [6]. No support is given for the Part 20 assumption.

It was found that the Part 20 assumption limited the scope of the proposed deuterium experiment to far less than the quantities used by DOE, as well as those used in comparable reactor cold source facilities in Australia and Germany, as these other institutions have already adopted ICRP-66 guidance. To continue pursuing our mission statement at NIST and stay relevant in a competitive research field, it is important to incorporate modern scientific knowledge and apply the latest health and safety standards. Thus, an adjustment per §20.1302(c) is requested. Specifically:

“Upon approval from the Commission, the licensee may adjust the effluent concentration values in appendix B to part 20, table 2, for members of the public, to take into account the actual physical and chemical characteristics of the effluents (e.g., aerosol size distribution, solubility, density, radioactive decay equilibrium, chemical form).”

Per §20.1302(c), the NBSR is submitting a LAR to adjust the DAC for our tritium gas to the ICRP-66 value to take into account the physical and chemical characteristics of elemental DT. The dose conversion factor for elemental DT is a factor of  $10^4$  lower than the factor for DTO.

### 3.2 New Tritium Release Paths

The deuterium distribution system spans indoor locations ( $2 \text{ m}^3$  both inside and outside confinement), and one designated exterior storage site (a  $16 \text{ m}^3$  ballast tank adjacent to the guide hall). A ground-level release from the outdoor ballast tank presents the greatest threat to public safety. The analyzed release point was 100 m from the stack, which corresponds to a system failure at the southwest corner of the guide hall. This point clearly bounds all conceivable releases paths to the 400 m boundary. The analyzed distance is 300 m.

Three release paths have been analyzed: a gaseous DT puff release (no heat); a slow DTO vapor release resulting from a deflagration (standing flame); and an instantaneous DTO release from a detonation (explosion). All cases were analyzed in HotSpot 3.0.3 using default settings unless otherwise noted. Class F (moderately stable) meteorological conditions and a wind speed of 1 m/s were found to be conservative in all cases except the deflagration.

#### 3.2.1 Deuterium Leak Resulting in Gaseous DT Puff

A tritium gas leak could result from inadvertent venting of the deuterium system, perhaps through gross mismanagement during a charging operation. In the analyzed scenario, the entire deuterium inventory is immediately discharged at ground level as DT gas (0% DTO).

As Figure 3 shows, gaseous diffusion occurs rapidly and the peak TEDE occurs well before the release path distance (300 m) is exceeded; this is due to the high buoyancy and natural diffusivity of DT in air. The maximum TEDE at 300 m is found with class F atmospheric conditions and a wind speed of 1 m/s. This TEDE is 0.4 mrem.

##### 3.2.1.1 Airborne DT-DTO Conversion

Two processes are possible to convert DT to DTO as it approaches the fence, and both will be shown to be negligible for this release path. The first process is an isotopic exchange (HX) with the outdoor air:  $DT + H_2 \rightleftharpoons HD + HT$  and  $DT + H_2O \rightleftharpoons HD + HTO$ . This process has been studied for heavy water production, during which a metal catalyst is used to achieve a useful efficiency – but no catalysts have been postulated for a tritium release to atmosphere. At STP with no catalyst, HX in air has been variously measured to occur at rates between 0.05 to 0.1 ml/mCi-hr, depending on the humidity [7]. The STP activity concentration of DT before mixing with air is:

$$C_{DT} = 2.8 \text{ kCi} / (5 * 18 \text{ m}^3) = 0.031 \text{ mCi/ml}$$

For the case-limiting 1 m/s winds, the DT plume would reach the fence line at 300 m approximately 5 minutes after release. Assuming perfect stoichiometry without any DT diffusion (impossible, conservative), the DT-DTO conversion factor as the gas crosses the boundary would be no more than:

$$C_{DT} * R_{HX} * t = \frac{0.31 \text{ mCi}}{\text{ml}} * \frac{0.1 \text{ ml}}{\text{mCi} \cdot \text{hr}} * \frac{5}{60} \text{ hr} = 2.6 \times 10^{-4}$$

The actual conversion rate will be much lower because of diffusion, which reduces the chemical potential driving the isometric exchange. Yet even ignoring this, the calculated exchange rate is trivially small.

The other process that can produce DTO is combustion:  $D_2 + DT + O_2 \rightleftharpoons D_2O + DTO$ . This conversion is exothermic, so any significant quantity of DTO that forms from DT gas would create a heat source, dramatically increasing airborne dispersion. It is physically impossible for a fraction of the DT cloud to combust without affecting the rest of the cloud, so this conversion process is not credible for a DT puff release type and must be analyzed separately.

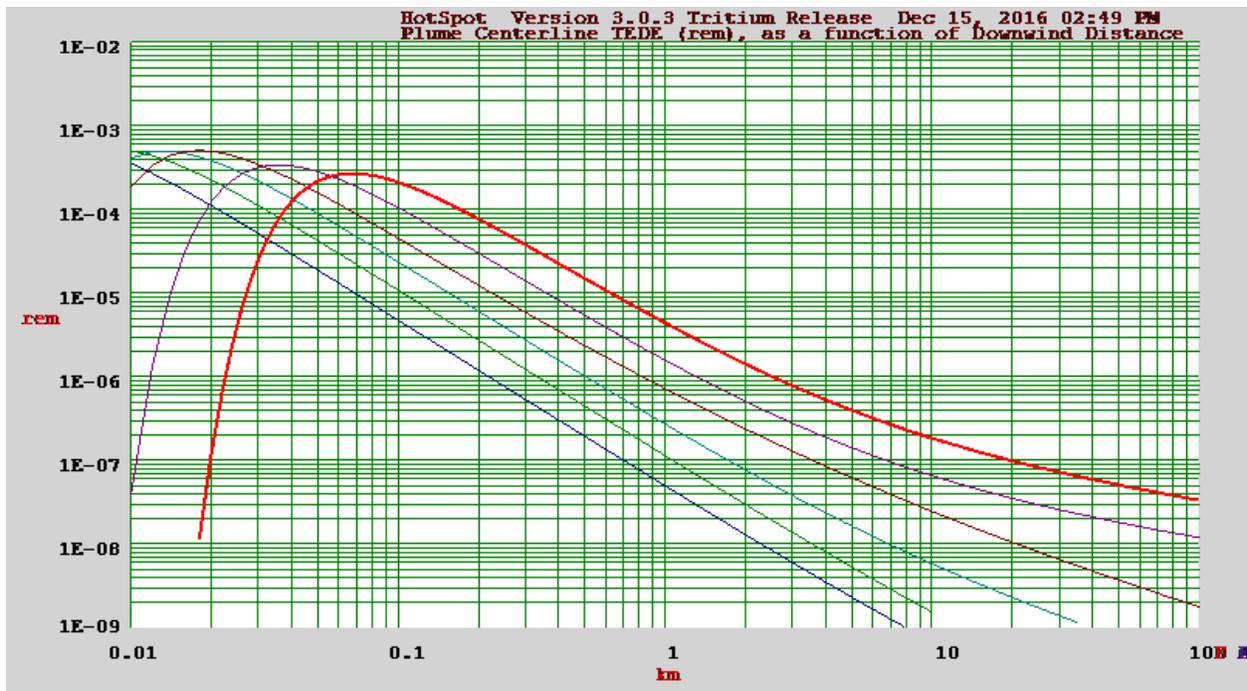


Figure 3. TEDE from Gaseous DT Release by Distance (Hotspot 3.0.3)

### 3.2.2 TEDE from DT Deflagration Resulting in DTO Vapor

If the DT should combust, either a slow continuous deflagration or a spontaneous combustion is possible. A deflagration is most likely to occur if a section of pipe is ruptured or the tank is punctured with a small hole. For this release path, the effluent DT gas is ignited by the same event that initiates the leak, forming a standing flame as it escapes. Bernoulli's equation is employed to calculate the release rate:

$$v = \sqrt{2 \Delta P / \rho}$$

Thus, for a 404 kPa differential pressure and STP helium density of 0.163 kg/m<sup>3</sup>, the ab initio escape velocity would be 2,226 m/s, or about mach 2.4. Above approximately mach 0.3, however, the gas mass flow rate will be choked due to the physical limit of the speed of sound, which is 933 m/s for deuterium

at 5 bar. The following equation for choked flow can be used to predict the tank depletion time in this pressure regime:

$$\dot{m} = C_d A \sqrt{k \rho_0 P_0 \left( \frac{2}{k+1} \right)^{\frac{k+1}{k-1}}}$$

where  $C_d$  is a discharge coefficient for the roughness of the hole (conservatively assumed here to be 1),  $A$  is the area of the hole, and  $k$  is the heat capacity ratio ( $c_p / c_v$ ). The choked flow condition associated with this equation is defined above the critical pressure,  $P^*$ , of the system, which is based on  $k$ . For deuterium,  $k$  is 1.3991, and  $P^*$  is approximately  $0.528 P_0$ . Below the  $P^*$  threshold, Bernoulli's equation may be applied instead. A finite element Matlab script was written to predict the gas mass flow rate and tank pressure as a function of time with the discontinuous function at  $P^*$  described above. The results of this calculation are shown in Figure 4.

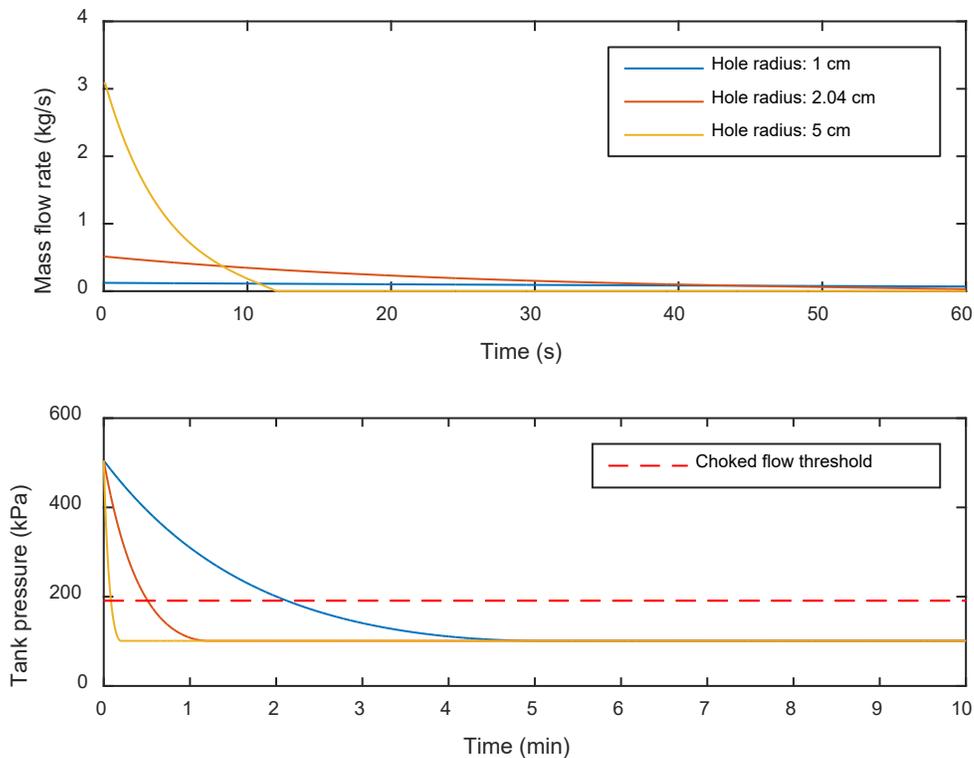


Figure 4.  $D_2$  ballast tank rupture profiles for select hole sizes. (Matlab script)

A hole radius of 2.04 cm corresponds to the 1-1/2" schedule 40 pipe that is proposed to connect the ballast tank to the remainder of the system, and smaller and larger hole radii are offered for illustrative purposes. For failure modes that produce a leak path larger than a pipe shear, it is assumed that the initiating event would cause an explosion, not a standing flame.

The Hotspot model, "General Fire," was used to model the 2.04 cm pipe break with the Fuel and Burn duration input option. The Burn Duration was set to 1.22 min as determined using Matlab above. The  $H_2 + O$  reaction energy of 241.8 kJ/mol was used to find the heat of combustion rate:

$$\frac{241.8 \text{ kJ/mol}}{4 \text{ g/mol}} = 1.44e4 \text{ cal/g}$$

and the liquid-equivalent fuel volume was calculated to be:

$$\frac{11.7 \text{ kg } D_2}{0.81 \text{ kg/l}} = 3.80 \text{ gal}$$

A “Physical Height of the Fire” parameter of 0 m was conservatively selected to model a downward facing penetration at the exit of the tank. Unique to this scenario, the most conservative meteorological condition was found to be Class E instead of Class F. These parameters were used to produce Figure 4, which shows that the maximum TEDE is 0.3 mrem at a public location 4 km from the release point. The dose to occupational workers is negligible.

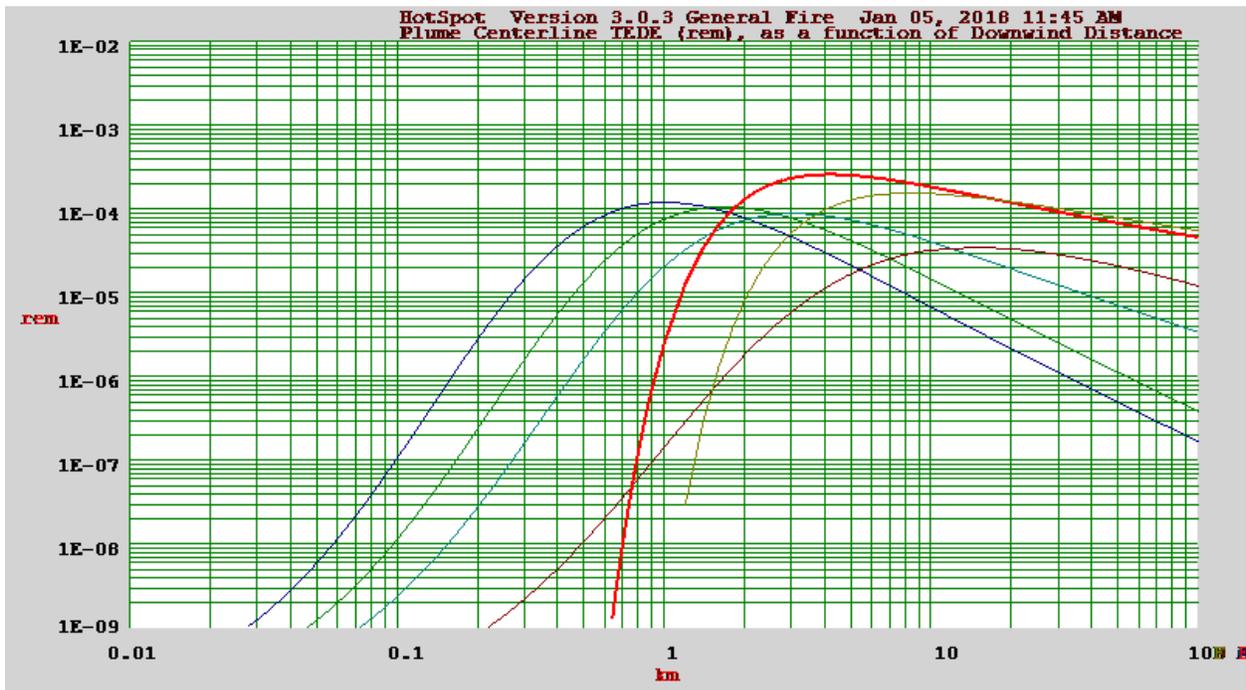


Figure 5. TEDE from a DT deflagration resulting in slow release of 100 % DTO. (Hotspot 3.0.3)

### 3.2.3 TEDE from Detonation Resulting in a DTO Puff

The third and final release path is a postulated detonation. Such an explosion could conceivably be caused by a shockwave, perhaps due to a major crane failure, a vehicular collision, or a high velocity impact like a bullet. The Hotspot model, “General Explosion” was selected with a High Explosive parameter of the deuterium inventory calculated by:

$$241.8 \text{ kJ/mol} * \frac{11.7 \text{ kg}}{4 \text{ g/mol}} * 4.184 \text{ MJ/kg TNT} = 373 \text{ lbs TNT}$$

This method was used to generate Figure 6, which shows the TEDE at the 400 m boundary (300 m from the release point) would be 1.5 mrem. The peak dose of 9 mrem would be inflicted on an occupational worker in the immediate vicinity of the tank. The physical damage to personnel at this distance is beyond the scope of this analysis.

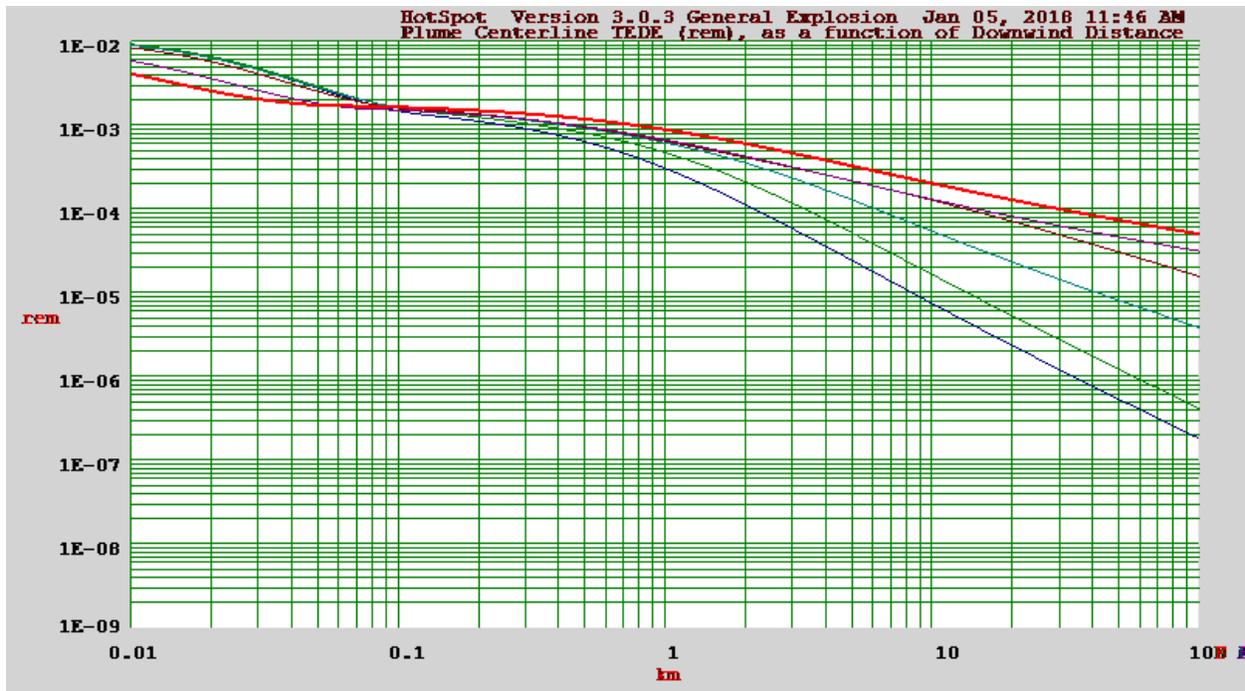


Figure 6. TEDE from a DT explosion resulting in 100 % DTO puff. (Hotspot 3.0.3)

## 4 Conclusion

The maximum public TEDE at any location for the DT puff, the DTO deflagration, and DTO explosion releases were 0.4 mrem, 0.3 mrem, and 1.5 mrem, respectively. For all cases the limiting meteorological conditions were considered, and the DTO releases are further conservative because 100 % chemical conversion was assumed, despite this being unlikely. This report concludes with confidence that in all cases the tritium release would not exceed the 100 mrem limit defined in 10 CFR 20 for members of the public. For this reason, the described events are not radiological “accidents” and do not require the special treatment of 10 CFR 50.

Furthermore, the public dose from a DT release would not exceed the extremely conservative 10 mrem prescribed by EPA for federal facilities other than nuclear regulatory commission licensees [8]. Although the NBSR is not required to meet this criterion, the fact that any DT release would be compliant with EPA standards supports our request to use ICRP-66 guidance for the DT dose conversion factor per FGR-13. A periodic feed-and-bleed program is not required to ensure compliance.

## 5 References

- [1] NIST Center for Neutron Research, "NBSR 14, Chapter 11 - Radiation Protection and Waste Management (rev 7)," NIST, Gaithersburg, 2015.
- [2] NIST Center for Neutron Research, "NBSR 14, Chapter 13 - Accident Analyses (rev 7)," NIST, Gaithersburg, 2015.

- [3] S. Homann and F. Aluzzi, "Hotspot, Health Physics Code Version 3.0 User's Guide," Lawrence Livermore National Laboratory, Livermore.
- [4] Environmental Protection Agency, "Federal Guidance Report No. 13: Cancer Risk Coefficients for Environmental Exposure to Radionuclides," Air and Radiation (6601J), Oak Ridge, 1999.
- [5] ICRP, "ICRP Publication 66: Human Respiratory Tract Model for Radiological Protection," SAGE Publications Ltd, 1995.
- [6] B. F. Peterman, J. R. Johnson and R. G. McElroy, "HT/HTO conversion in mammals," *Fusion Technol.*, 1985.
- [7] J. Phillips and C. Easterly, "Tritium Oxidation and Exchange: Preliminary Findings," Oak Ridge National Laboratory, 1978.
- [8] Environmental Protection Agency, "40 CFR 61, Subpart 1: National Emission Standards for Radionuclide Emissions From Federal Facilities Other Than Nuclear Regulatory Commission Licensees and Not Covered by Subpart H," Washington, DC, 1989.