Analyzing Environmental Tritium Cycle for Fusion Energy and National Security

by

Liliana R. Arias

Submitted to the Department of Nuclear Science and Engineering
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ABSTRACT

Although tritium is a sought-after isotope of hydrogen for fusion fuel, it is important to consider the environmental impacts of its release into the environment. In order to prepare for the elevated tritium releases that may result from commercial fusion power, yearly tritium releases from different types of nuclear facilities are compiled, with an emphasis on fusion reactors. Atmospheric modeling using HYSPLIT and a Gaussian Plume model is then conducted in order to better understand current and future global tritium sources and concentrations and their release pathways in the environment. Despite elevated tritium levels near major sources, most emissions remained within regulatory bounds, although proximity to facilities still matters.

Thesis supervisor: Haruko Murakami Wainwright
Title: Assistant Professor of Nuclear Science and Engineering

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Chapter 1

Introduction

Tritium is a radioisotope of hydrogen with a half-life of about 12.32 years [1]. It is a pure beta emitter that releases low-energy beta radiation. Each year, cosmic rays colliding with nitrogen molecules create an estimated 210 grams (7.6×10^{16}Bq) of tritium [2]. Tritium is primarily created in the upper atmosphere through the reactions $^{14}\mathrm{N}(n,T)^{12}\mathrm{C}$ and $^{16}\mathrm{O}(n,T)^{14}\mathrm{N}$ [3]. The total natural abundance of tritium is estimated to be about 1-1.3 10^{18} Bq (about 2800-3600 g) [4]. Protium is the most abundant hydrogen isotope, followed by deuterium, with about 1 atom of deuterium for every 6,600 atoms of protium [5]. Once created, it can be readily incorporated into the global water cycle [6]. The relative natural abundance of tritium is about 1 atom of tritium for every 10^{18} atoms of protium, although anthropogenic releases of tritium in the environment have increased this abundance to about 1 atom of tritium for every 10^{17} protium atoms.

Global environmental tritium levels have been significantly affected by anthropogenic activities. In the past, the majority of tritium in the atmosphere was the result of frequent nuclear weapons testing, particularly from fusion bombs [1]. After the cessation of underground nuclear weapons testing in the 1990s, the nuclear power industry is now the main anthropogenic cause of tritium in the atmosphere [7]. Tritium is created and released by nuclear facilities such as fission power plants [3]. A significant amount is also released by spent fuel reprocessing plants, such as the La Hague reprocessing plant[8].

Because it can substitute for hydrogen atoms through isotopic exchange, tritium can take the place of hydrogen in water (HTO) and be absorbed by the human body [6]. Organisms can be exposed to tritium releases in the environment through inhalation, absorption, and ingestion of tritium, which poses a possible carcinogenic risk [6]. In the environment and in biological systems, tritium is mobile and at higher levels it can lead to a decrease in survival of species [9]. Cancer risk of tritium exposure has been studied in experiments involving mice, where some health effects were observed [6].

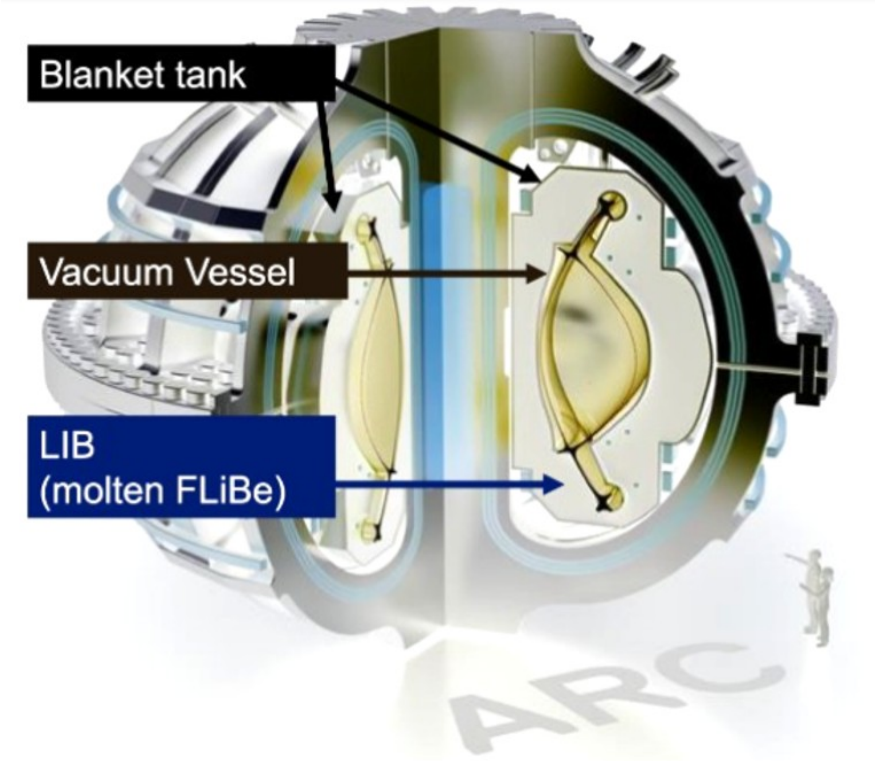


Figure 1.1: ARC, a future fusion power plant (Figure from Commonwealth Fusion Systems with colors inverted [10]).

1.1 Research Gaps

A forthcoming source of anthropogenic tritium is the development of fusion power plants (FPPs) [11]. Many FPP designs use the fusion reaction of deuterium and tritium to produce energy, and thus require significant amounts of tritium to fuel their plants [12]. As such, FPPs will release a significant amount of tritium in the environment [11]. Because commercial fusion plants do not yet exist, their projected tritium releases have not been studied as extensively as those of current facilities.

Additionally, although production of tritium from different sources has been discussed separately, there lacks a comprehensive synthesis of the source terms from different facilities. Also, there is a lack of studies that integrate atmospheric transport and detectability for different types of tritium releases.

1.2 Thesis Objective

This thesis aims to both synthesize tritium release information from different types of nuclear facilities and evaluate tritium concentrations in the environment in terms of detectability and safety.

While atmospheric modeling of tritium has been done before, my thesis applies methods of

atmospheric dispersion simulation in order to better understand the environmental footprint of different types of nuclear facilities. For this thesis, two models of atmospheric dispersion will be applied. The first model to be used is HYSPLIT, a model created by the National Oceanic and Atmospheric Administration (NOAA). The second is a Gaussian Plume model. For each of these models, release of a unit source will be simulated (1 g/hr), then the results will be scaled for different types of nuclear facilities. The results will be compared to background levels of tritium, minimum detectable concentrations of tritium, and regulatory standards for maximum concentration of tritium in order to better understand the tritium footprints of different types of nuclear facilities.

The simulation in tandem with the global tritium emissions estimate helps people to understand the significance of release from nuclear facilities such as NPPs, fusion plants, and reprocessing facilities in comparison to each other and to background levels. Additionally, the results illustrate how much the addition of each nuclear facility changes the background concentration of tritium, which makes it possible to understand if the nuclear facilities have significant environmental impacts, thereby informing policy on the addition of future nuclear facilities.

Ultimately, the goal of this thesis is to prepare society for potential higher levels of tritium releases from fusion reactors through developing a more complete understanding of current and future global tritium sources and concentrations and their release pathways in the environment.

Chapter 2

Background

Like protium, tritium is found in all compartments of the geosphere, including the atmosphere, hydrosphere and lithosphere, the biosphere and the anthroposhere [13]. Tritium produced in the atmosphere naturally forms tritiated water (HTO), its most common form in the natural environment, through oxidation or isotopic exchange, as described in equations 2.1 and 2.2, respectively [14] [13].

$$T_2 + \frac{1}{2}O_2 \rightarrow T_2O \tag{2.1}$$

$$T_2 + H_2O \rightarrow HTO + HT \tag{2.2}$$

Less commonly, tritium can also be found as tritiated hydrogen (HT) or sometimes tritiated methane in the atmosphere (CH_3T) [13]. HTO can be integrated into the global water cycle, and it can also be converted into OBT (organically bound tritium) when it gets incorporated into the molecules of living organisms [6] [13]. The key processes of tritium in the environment are illustrated in Figure 2.1.

Anthropogenically, tritium is created in nuclear facilities such as nuclear power plants (NPPs). The main sources of tritium in fission power plants are [3]:

1) triple fission of fuel nuclei (when three daughter products are created during nuclear fission rather than just two),

2) (n, γ) reactions on the deuterium in water (when water is used as the coolant)

3.) neutron capture by boron and lithium used in the coolant and control rods

4) the $^3\mathrm{He}(n,p)^3\mathrm{T}$ reaction in the He gas circuit of light-water graphite reactors (LWGR)

5) reactions of the type (n, T) and (n, p) between fast neutrons and nuclei such as ^{14}N, ^{6}Li, ^{10}B, and ^{40}Ca

In fusion reactors, tritium will be created through neutrons from the fusion reactions interacting with lithium, as described in Figure 2.2 [16]. Tritium releases for fusion will be in the form of both liquids and gases. Information about these releases will be used in the simulations described in the subsequent section.

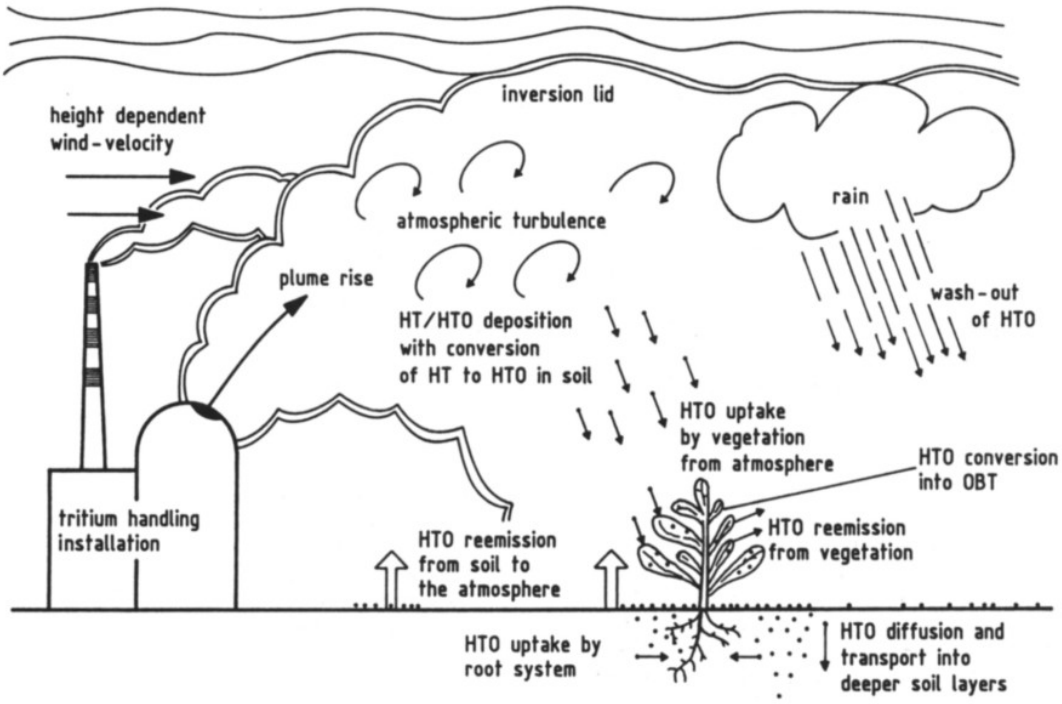


Figure 2.1: Key processes of tritium in the environment, taken from [15].

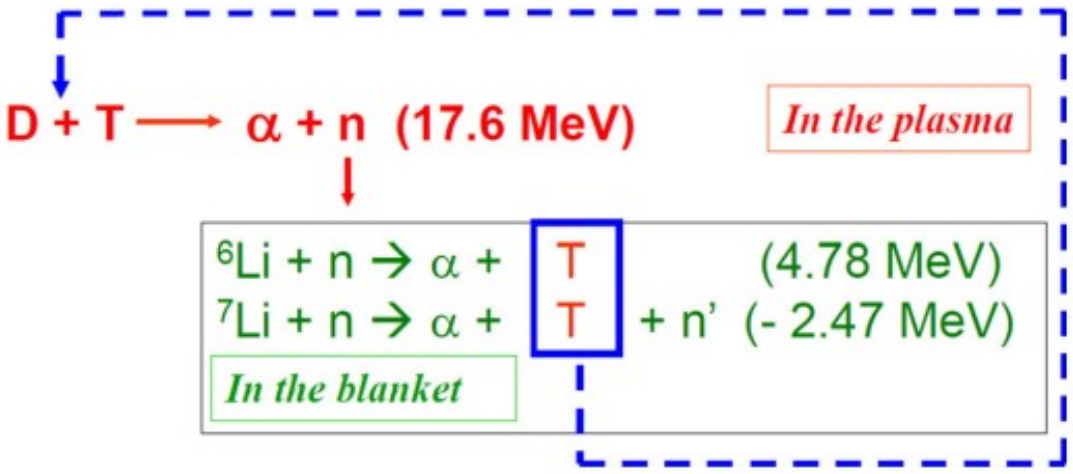


Figure 2.2: Tritium cycle in FPPs: neutrons from fusion reactions interact with lithium to form tritium. Taken from [16].

2.1 Atmospheric dispersion modeling

The atmospheric release of chemical, biological, and radiological (CBR) pollutants poses a threat to people today, as they are commonly released due to technological advances [17]. Because the release of CBR pollutants covers extensive amounts of space, it is important to be able to understand their dispersion for the safety of the public. Atmospheric dispersion modeling now plays an increased role in emergency response planning and accident response with regard to CBR pollutants.

Atmospheric dispersion modeling makes use of mathematical formulas in order to qualify the processes that distribute particles released from a source [18]. Through the use of meteorological and emissions inputs, it can predict the resulting concentration of particles at downwind locations. My research will focus on the NOAA's HYSPLIT and a Gaussian Plume model for Gaussian dispersion modeling.

Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) is the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory's model for atmospheric transport [19]. HYSPLIT is one of the most commonly used atmospheric transportation and dispersion models in the atmospheric sciences community. It has been used in various simulations that model the atmospheric transport of hazardous materials, including radioactive materials, such as in the deposition of fallout after nuclear weapons testing in the atmosphere, and behavior of the plume trajectory after the nuclear accident at the Fukushima reactor in Japan. HYSPLIT comes with post-processing programs that can both visualize and analyze simulation outputs. The simulations produce concentration maps by using a combination of the Lagrangian approach, in which it uses a moving frame of reference for advection and diffusion calculations, and Eulerian methodology, in which it uses a fixed three-dimensional grid as a frame of reference. The most relevant of HYSPLIT's abilities to this thesis are that it can be used to calculate the origin or destination of air masses, along with the transport and deposition of radioactive materials [19].

Gaussian Plume modeling is an analytical steady-state solution to the simplified form of the three-dimensional advection-diffusion equation, obtained by assuming constant wind velocity aligned with the positive x-axis, a wind velocity large enough that diffusion in the x–direction is much smaller than advection, isotropic diffusion, constant emission rate from a point source, negligible topography, and no contaminant penetrating the ground [20]. As of 2011, most NPP environmental impact assessments used a Gaussian plume model; however, the Gaussian Plume model has several limitations.

$$C(x,y,z) = \frac{Q}{2\pi u \sigma_y \sigma_z} \left\{ \exp\left[-\frac{(z-h)^2}{2\sigma_z^2} \right] + \exp\left[-\frac{(z+h)^2}{2\sigma_z^2} \right] \right\} \exp\left[-\frac{y^2}{2\sigma_y^2} \right] \tag{2.3}$$

where C is the resulting concentration at a location (x,y,z) (g/m^3), Q is the source release rate (g/s), u is the wind speed (m/s), h is the stack height (m), y is the crosswind distance (m), z is the vertical distance of the receptor from the ground (m) and σ_y (m) and σ_z (m) are the diffusion parameters in their respective directions, and are determined as a function of the upwind distance, x, and stability class [21] [22].

According to a specific study in which models using Gaussian Plume modeling and HYSPLIT were compared, HYSPLIT generally (though not always) predicts larger peak air

concentrations and depositions than Gaussian Plume modeling [23]. There did not exist a clear distance at which the Gaussian Plume model results began to diverge by more than an order of magnitude from the HYSPLIT results. Results from the HYSPLIT model were generally within a factor of two of the results from the Gaussian Plume model.

Gaussian Plume modeling has certain advantages over HYSPLIT, particularly in terms of computational time and required meteorological data [23]. In the study, the HYSPLIT model required about 1 TB of storage and 100-200 times the computational run time of the Gaussian model. However, these advantages mean less today than in previous decades due to the better computational power available.

2.2 Tritium regulatory limits, detection thresholds, and background

Tritium drinking water limits globally are summarized in Table 2.1.

Table 2.1: Tritium Limits in drinking water by country/region [24].

Country/Region	**Tritium Limit (Bq/L)**
Canada	7,000
EU	100
Finland	30,000
Australia	76,103
Russia	7,700
Switzerland	10,000
United States	740
WHO	10,000

The allowable tritium effluent air concentration in public settings in the United States is 1×10^{-7} μCi/ml [25], equivalent to 3.7 Bq/L. Over the course of a year, if inhaled/ingested continuously, this concentration would produce a total effective dose equivalent of 0.05 rem (equal to 50 millirem or 0.5 millisieverts).

The detection limit of tritium for short duration measurements is about 0.064 Bq/L [26]. This is for a method that consists of converting tritiated gases into tritiated water through burning the gases with a known quantity of hydrogen. The water vapor is then condensed and added to a liquid scintillator to be measured. The combustion period is 10 minutes, and the counting period is also 10 minutes.

When the sampling duration is about a week, the detection limit is between 2×10^{-4} Bq/L and 5×10^{-4} Bq/L for a method that measures tritium concentration by trapping tritium in air by bubbling through a water solution. [27].

Limited data is available on atmospheric tritium concentration in the United States, although it was measured in Kumamoto, Japan between 2003 and 2005 (Table 2.2). Assuming the levels are similar to those in Cambridge, MA, the total tritium background concentration from the different species sums to 2.82×10^{-5} Bq/L [28].

Table 2.2: Concentrations of atmospheric tritium [28].

Chemical species	**Concentration**
HT	12.5 ± 6.9 mBq m^{-3}
CH_3T	9.0 ± 8.2 mBq m^{-3}
HTO	6.7 ± 5.4 mBq m^{-3}
Total	2.82×10^{-5} Bq L^{-1}

Chapter 3

Approach and Methods

3.1 Source Terms

To quantify the yearly tritium releases, data is collected on both anthropogenic tritium releases from nuclear power and fuel reprocessing plants in addition to data on natural tritium creation and release. Much of this data is publicly available in reports online, such as in US Nuclear Regulatory Commission (NRC) and Canadian Nuclear Safety Commission effluent reports, and journal articles. We can thus compile it to create our estimate. Data on natural sources is taken from [2], and data from La Hague is taken from [8]. Additionally, data on tritiated water releases from Fukushima is taken from [29]. Gaseous and liquid release values from a report on different HWRs in Canada are averaged [30]. To find the gaseous and liquid T releases for a PWR and BWR, respective values for releases from PWRs and BWRs in the US available in an NRC report are averaged, and then divided by the electricity production of each plant [31] [32]. Finally, the values are scaled to be for a 1 GWe reactor. Data on yearly releases from different types of nuclear facilities will be used for simulations, as described in the following paragraphs.

Because commercial FPPs do not yet exist, their yearly tritium releases are based on estimates. One recent study estimated fusion releases to be between 0.3 and 37 g/yr (scaled for a 1 GWe fusion plant, assuming conversion efficiency of 0.33), based on a tritium permeation modeling code (TMAP4) that predicted tritium losses from the Fusion Nuclear Science Facility, an experimental device designed to generate fusion power [11]. The study chose to use a release rate of 6 g/yr-GWe in their model, and assumed all tritium releases to be gaseous. This thesis will also use those estimates for an FPP, with all releases assumed to be gaseous, as previous studies estimate liquid tritium releases to be orders of magnitude smaller than gaseous releases, or negligible [33] [34].

3.2 Atmospheric tritium transport

For both methods, we simulate a plume and concentration at a given time and space. We use a pre-calculated concentration and simulate a unit release rate in g/hr, with the release rate proportional to the concentration. HYSPLIT simulates 1g of tritium being released once per hour, whereas Gaussian Plume uses a constant release, so 1/3600 g of T is released each

second. Additionally, a stack height of 47 m, roughly the stack height of the MIT Research Reactor (MITR-III) in Cambridge, MA, is used for both methods [35]. The simulation conditions are summarized in Table 3.1.

We assume that our release rate is additive—that is to say, we assume that if the tritium source were 10 Bq instead of 1, we could just multiply our results by 10 to find the new answer. We also assume that we can find the results for multiple sources through using superposition. Additionally, we decouple atmospheric simulations and source terms, meaning that we can simply switch between fission and fusion by multiplying the results by the respective magnitude of the source term rather than rerunning the simulation. This ability to switch between results for fission and fusion is implemented in the thesis by providing results for a unit source, and then multiplying the source by the respective tritium releases in order to find results for typical NPPs (based on average tritium releases from PWR, HWR, BWR) and a theoretical fusion reactor. The unit source terms are described in Table 4.1.

HYSPLIT and the Gaussian Plume model each produce a scatterplot, to which curves are fitted (ignoring outliers), then scaled for different types of nuclear facilities using the values in Table 4.1.

Table 3.1: Comparison of HYSPLIT and Gaussian Plume modeling parameters.

Category	**Parameter**	**HYSPLIT**	**Gaussian Plume**
Source Location	Latitude / Longitude	42.3602, -71.0965	(0.0, 0.0) in local grid
	Stack Height	47 m	47 m
Emission Rate	Q	1 g/hr	1.0/3600 g/s (1 g/hr)
Duration	Simulation Time	1 year	1 year of hourly plumes
Meteorology	Wind Speed	From HRRR/NAMS meteorological input	Monthly average (converted to m/s)
	Wind Direction	From met input	Constant 270° (west)
	Stability Class	From met input	Cycles through different atmospheric classes
Grid Setup	Spatial Resolution	Polar: 64 angles, 0.5° width, 20 km radius	Cartesian: 200 m grid, $[-10\,\text{km}, 10\,\text{km}]$
	Vertical Domain	Receptor height at 50 m	Sampled at 50 m
Deposition / Aerosol	Particle Size	1.0 µm[a]	N/A

[a] Not the actual diameter of HTO, but HYSPLIT's default for particles when deposition is not computed [36].

Other values not specified were left as the model defaults.

3.3 HYSPLIT Simulation

We use HYSPLIT to simulate the transport and deposition of tritium for a given tritium release from a hypothetical facility in different environmental conditions, release conditions, and pathways of release [19]. The most relevant of HYSPLIT's abilities to this thesis are that it can be used to calculate the origin or destination of air masses, along with the transport and deposition of radioactive materials [19].

HYSPLIT is run using weather data from the National Oceanic and Atmospheric Administration (in this case, for Cambridge, MA).

For HYSPLIT, concentrations are generated for every day for a year in polar coordinates, with points places at the center of grid squares. The files contain concentration angles for different angles and distances from the source.

Rows are degrees from north clockwise with a total of 64 angles.

$$\text{Deg} = \frac{360}{\text{Angles}} \cdot \text{Row} + \frac{180}{\text{Angles}} \tag{3.1}$$

Row 0 is thus 2.8125 deg, and row 1 is 8.4375 degrees, etc.
Columns are distance from the source to 20 km in 0.1 km steps.

$$\text{Distance} = 0.1 \cdot \text{Column} + 0.05 \tag{3.2}$$

Column 1 is thus 0.05 km, and column 2 is 0.15, etc.

In order to create curves, a matrix of maximum values over all the files is created by looping through each file in order to find the element-wise maximum of all the data. Concentrations are then averaged over all angles in order to have data of maximum concentration vs. distance.

In order to differentiate between releases of HT and HTO, HYSPLIT was run for particles of diameter 0 and 1 micron, respectively. (A diameter of 0 is used for gases such as HT in HYSPLIT, while a diameter of 1 µm is used to define pollutants as particles [36].) Their plots of concentration vs. distance were extremely similar (see Figure 4.3), so only the HTO data were used from this point forward.

3.4 Gaussian Plume Averaging

In addition, a simulation using a Gaussian plume described by Equation 2.3 is run, using similar parameters as for HYSPLIT. The code for the model is taken from [37] and modified for the current setup. This allows us to compare the results for the two simulation methods, a valuable task since Gaussian Plume modeling is a much less computationally expensive way of modeling atmospheric transport. After running the Gaussian Plume model, the results are convert to be in units of Bq/L.

The Gaussian Plume was run for each combination of six atmospheric stability classes and 12 wind speeds based on each of the average monthly wind speeds in Cambridge, MA (Table 3.2), then averaged. A wind direction from the west (dominant wind direction in Cambridge) was used [38].

Table 3.2: Monthly Average Wind Speed (mph) [38].

Month	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Speed (mph)	8.5	8.7	8.1	7.0	5.8	5.1	4.8	4.8	5.5	6.4	7.2	7.9

For each combination of atmospheric stability and wind speed, the maximum concentration between 0 and 50 meters height was extracted, and the maximum concentration for each location was stored.

The maximums for each combination were then averaged, and for each radial distance bin, the maximum concentration across all grid points within that bin was taken to produce a final curve of maximum concentration vs. distance from the source.

In addition to curves, a line is plotted for the background concentration of tritium, the maximum public air concentration limit, and detectable levels of tritium, as described in section 2.2.

Chapter 4

Results

4.1 Tritium releases

The results on tritium releases from a variety of different nuclear facilities are summarized in Table 4.1 and Figures 4.3 and 4.2. Tritium release information for all nuclear facilities listed except for FPPs is based on measured data, whereas the value for an FPP is based on a middle-of-the-road estimate from the literature [11], converted to GWe by assuming 0.33 conversion efficiency.

As shown in Table 4.1 and Figures 4.3 and 4.2, more tritium is put into the atmosphere yearly by natural sources than by any of the individual nuclear facilities listed. Additionally, La Hague, a nuclear fuel reprocessing plant, releases more tritium in total than any of the nuclear power plants listed, by a large factor. After La Hague, the next largest tritium source is a theoretical future fusion reactor, while the smallest tritium source would be a BWR. In terms of gaseous anthropogenic release, a fusion power plant would release the most, while a PWR would have the smallest yearly gaseous releases. In terms of anthropogenic liquid releases, La Hague would have the highest, followed by an HWR. In terms of known values, a BWR would have the lowest yearly anthropogenic liquid tritium release.

Additionally, different nuclear facilities have different ratios of liquid to gas tritium releases, which reflect different effluent management. La Hague reprocessing facility releases the vast majority of its tritium in liquid form– specifically, 99.4%. PWRs are similar, as they also release the majority of their tritium (94.3%) in liquid form, although the percentage is slightly smaller than for La Hague. Conversely, both BWRs and HWRs have the majority of their releases in gaseous form- 88.2% and 53.1%, respectively. In terms of NPPs, an HWR has the most significant gaseous tritium releases, though the gaseous releases from an FPP are about 6 times greater than those from an HWR.

Nuclear Facility	**Gaseous T release (g/yr)**	**Liquid T release (g/yr)**
Natural sources [2]	210	0
La Hague [8]	0.22	38.5
Fukushima [29]	0	6.2×10^{-2}
Fusion power plant [11]	6 (estimated)	?
Pressurized Water Reactor (PWR) [31, 39]	3.29×10^{-3}	5.43×10^{-2}
Boiling Water Reactor (BWR) [31, 39]	3.68×10^{-3}	4.89×10^{-4}
Heavy Water Reactor (HWR) [40]	0.94	0.83

Table 4.1: Annual tritium (T) releases from various nuclear facilities, normalized per 1 GWe for power plants. For the purposes of this study, we assume all T releases from FPPs are gaseous releases.

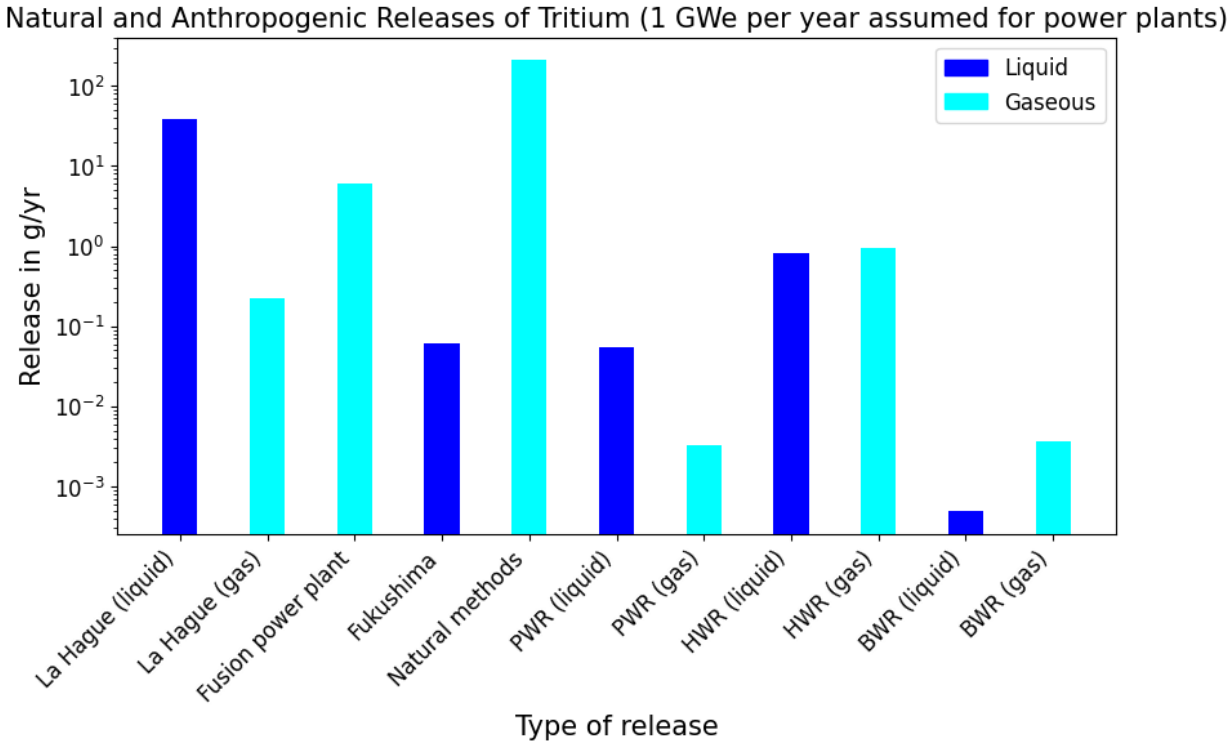


Figure 4.1: Summary of annual tritium releases, from natural and anthropogenic releases. Each power plant is assumed to be 1 GWe.

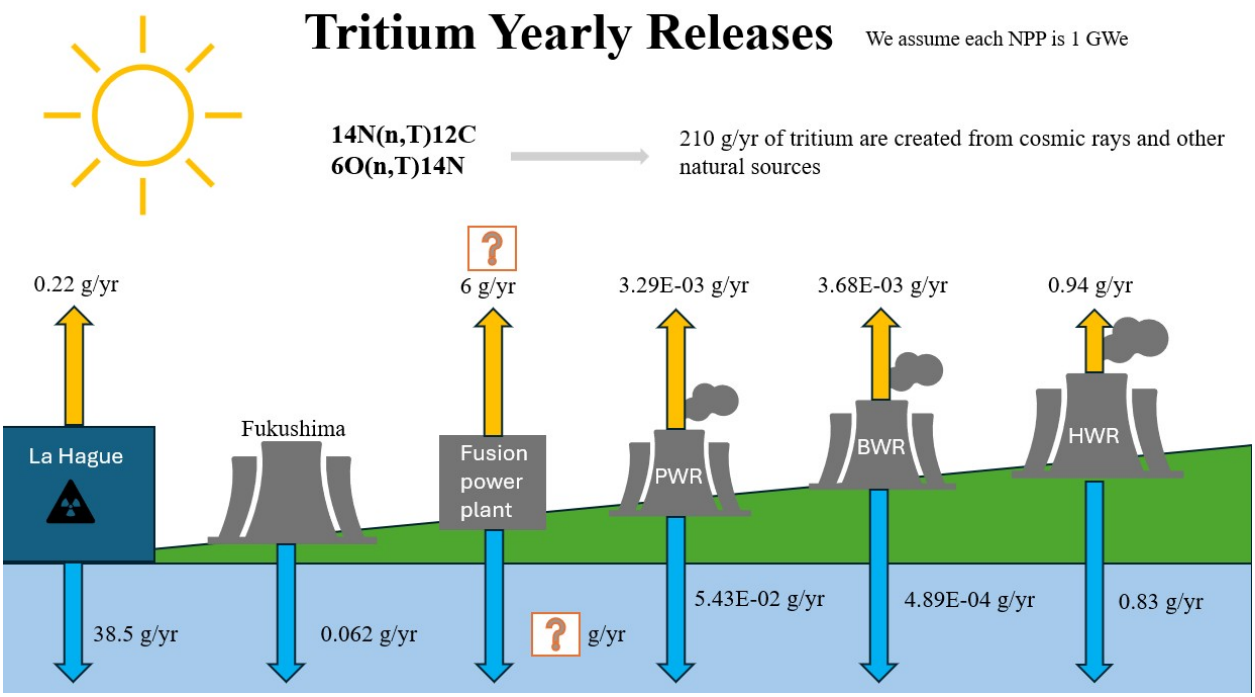


Figure 4.2: Visualization of annual tritium releases, from natural and anthropogenic releases. Each power plant is assumed to be 1 GWe.

4.2 Tritium modeling results

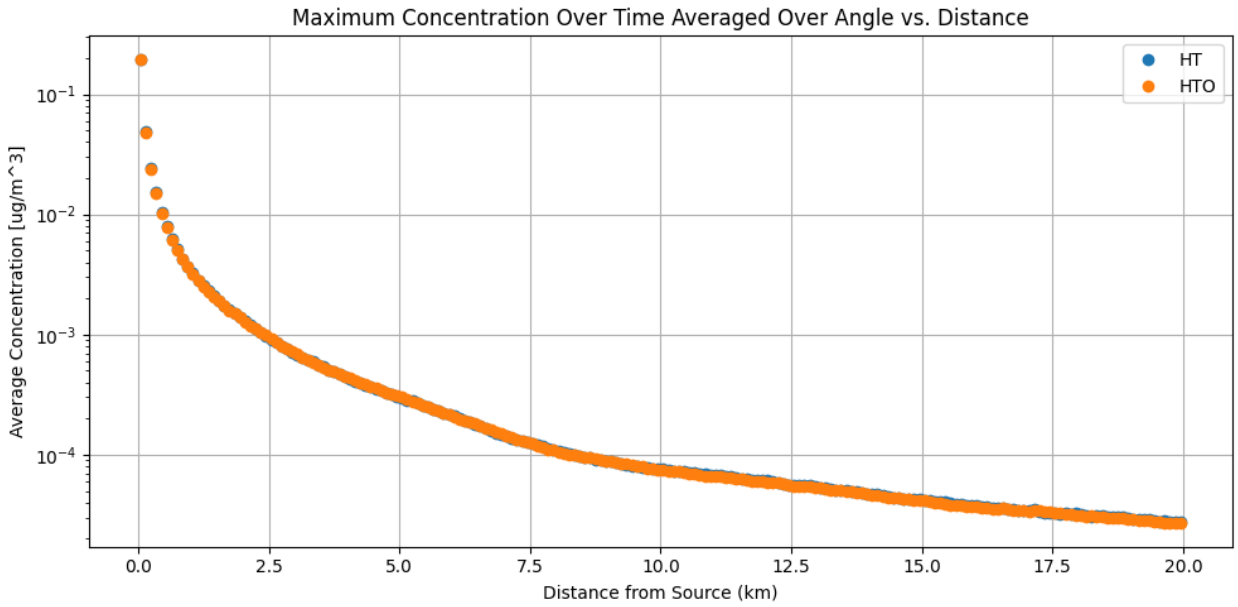


Figure 4.3: Plot comparing results of HT and HTO from HYSPLIT.

As previously mentioned, the plots of concentration vs. distance for HT and HTO

were extremely similar (see Figure 4.3), so only the data from HTO was used from this point forward. The results of maximum tritium concentration vs. distance were plotted as scatterplots (Figure 4.4), then a power law fit was applied to each, resulting in equations 4.1 and 4.2. As shown in Figure 4.4, HYSPLIT consistently estimates resulting concentrations that are higher than those of Gaussian Plume modeling (roughly 1.75 orders of magnitude higher on average). Additionally, although the fitted curves follow the data closely based on the high R^2 values, the results begin to diverge lower than the HYSPLIT fit past about 4.5 km, and slightly above the Gaussian fit past about 5 km. The curves were then scaled for the different types of nuclear facilities listed in Table 4.1, and plotted against the air concentration limit (described in section 2.2) in Figure 4.5.

$$\text{HYSPLIT Fit:} \quad C(r) = 1.19 \times 10^3 \cdot r^{-1.42}, \quad R^2 = 0.999 \tag{4.1}$$

$$\text{Gaussian Fit:} \quad C(r) = 2.53 \times 10^1 \cdot r^{-1.61}, \quad R^2 = 0.998 \tag{4.2}$$

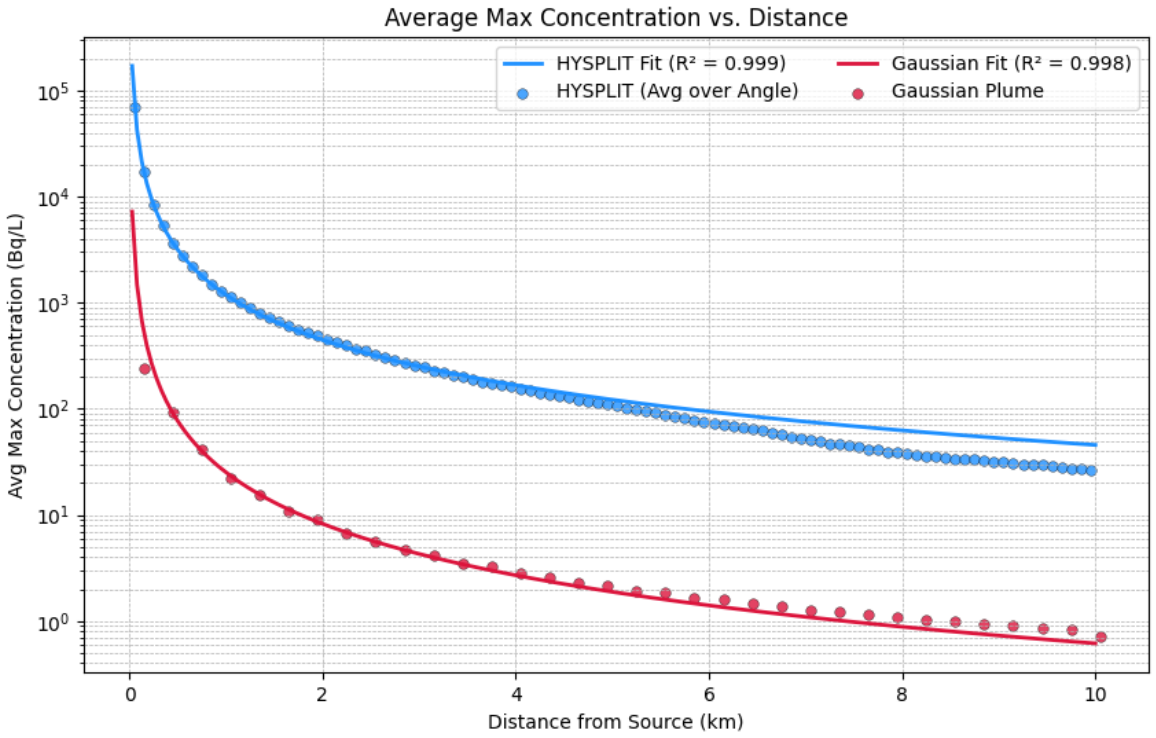
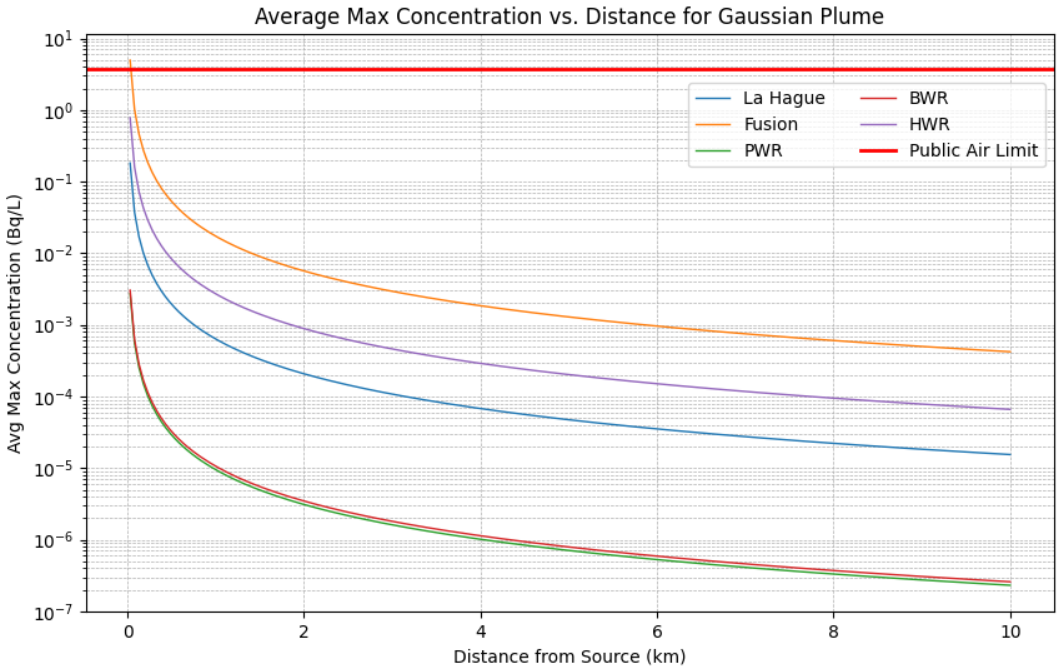
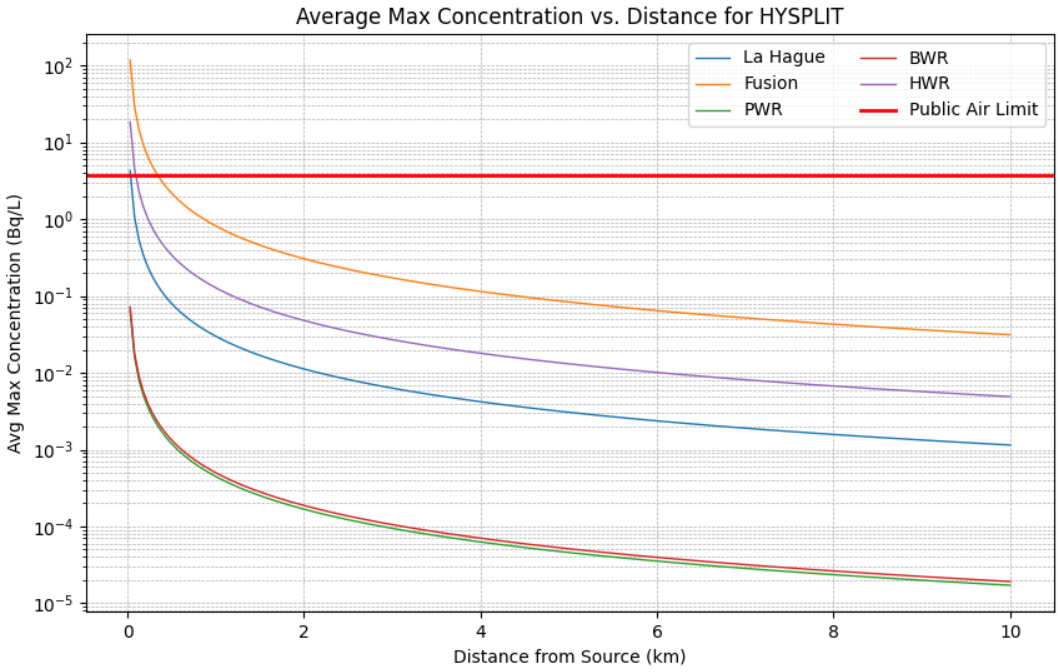


Figure 4.4: Raw modeling results, fitted to curves. Equation 4.1 is used for HYSPLIT and 4.2 is applied to the Gaussian Plume data.

Due to the different values used for the yearly gaseous tritium releases (Table 4.1), the resulting concentrations (Figure 4.5) are therefore different as well, with an FPP leading to the highest concentrations, followed by an HWR, La Hague, a BWR, and finally, a PWR. As shown in Figure 4.5, according to Gaussian plume modeling, the only offending facility is the



(a) Gaussian Plume: Public air concentration limit



(b) HYSPLIT: Public air concentration limit

Figure 4.5: The concentration exceeds the public air concentration limit until 0.35 km for a fusion reactor, 0.04 km for La Hague, and 0.10 km for an HWR according to HYSPLIT. According to the Gaussian Plume, only an FPP exceeds the limit for the first 0.05 km.

theoretical fusion plant, whose emissions only exceed the public air limit of 3.7 Bq/L until about 0.05 km (50 m) away from the plant. According to the HYSPLIT model, the resulting maximum tritium air concentration for a fusion reactor exceeds the public air limit for the first 0.35 km, for the first 0.10 km for an HWR, and for the first 0.04 km for La Hague, while no other facilities exceed the limit.

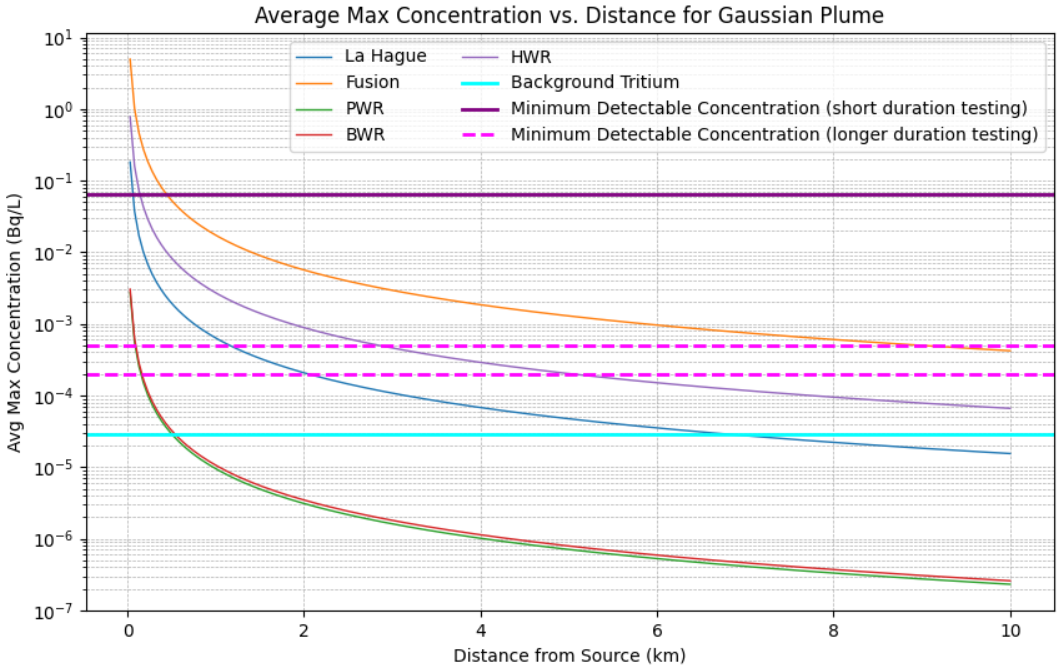
In Figure 4.6, the background tritium concentration and minimum detectable concentration limits, as described in section 2.2, are also plotted. According to HYSPLIT, tritium concentrations are always above the background tritium air concentration limit for the first 10 km for all but a BWR and PWR, which become less than background at 7.62 km and 7.04 km, respectively. According to the Gaussian Plume model, the resulting concentrations from both an HWR and FPP are above background tritium concentration for at least 10 km, while the curves dip below background concentration for La Hague, BWR, and PWR at 6.89 km, 0.55 km, and 0.51 km, respectively.

Additionally, according to HYSPLIT, the resulting tritium concentrations are always detectable for short duration testing until 6.03 km, 1.63 km, 0.59 km, 0.04 km, and 0.03 km for an FPP, HWR, La Hague, BWR, and PWR, respectively. According to the Gaussian plume model, they are detectable until 0.45 km, 0.14 km, and 0.07 km for a fusion power plant, HWR, and La Hague, respectively, and are never detectable for a PWR and BWR.

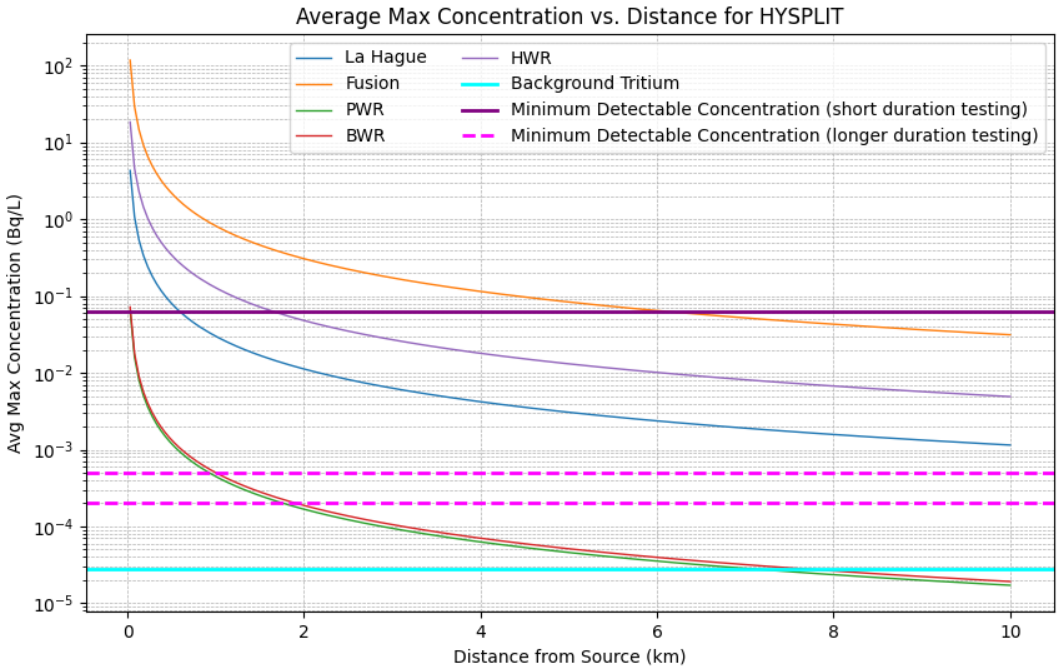
Also as shown in Figure 4.6, for longer duration testing, according to the HYSPLIT model, a fusion power plant, La Hague, and HWR are always above the minimum detectable concentration, whereas a PWR and BWR are above until between 0.93-1.77 km, and between 1.00-1.91 km. According to the Gaussian Plume model, a fusion power plant would be above the detection limit until 9 km or beyond, an HWR would be above until between 2.85 and 5.03 km, La Hague would be above until between 1.16 and 2.05 km, a BWR would be above until between 0.10 and 0.17 km, and a PWR would be above until between 0.09 and 0.16 km. Detectability is summarized in Table 4.2.

Table 4.2: Maximum tritium detectability distances for short and long duration testing.

Facility	**HYSPLIT Short Duration (km)**	**HYSPLIT Long Duration (km)**	**Gaussian Short (km)**	**Gaussian Long (km)**
FPP	6.03	10+	0.45	9+
La Hague	0.59	10+	0.07	1.16–2.05
HWR	1.63	10+	0.14	2.85–5.03
BWR	0.04	1.00-1.91	0	0.10–0.17
PWR	0.03	0.93–1.77	0	0.09–0.16



(a) Gaussian Plume: MDC (Minimum Detectable Concentration) for long and short duration testing and background



(b) HYSPLIT: MDC for long and short duration testing and background

Figure 4.6: Detectable limits and comparison to background for resulting concentrations.

Chapter 5

Discussion

To our knowledge, this is the first time that the estimates of tritium releases from a variety of types of different nuclear facilities are compiled and then modeled to find the resulting concentration vs. distance from the source. Although the result is limited to one particular location, the results suggest that some facilities may exceed the regulatory tritium air concentration limit in their proximity. In addition, the results suggest that tritium releases are detectable within tens of kilometers, though detectability distance varies with nuclear facility, detection methods and model types.

In this study, a key result is that HYSPLIT estimates larger tritium concentrations than the Gaussian Plume model does. This is consistent with previous work in which HYSPLIT generally predicted larger peak air concentrations and depositions than Gaussian Plume modeling [23]. In addition, HYSPLIT's higher results in this thesis could be due to the fact that it simulates releases occurring once an hour, which can lead to concentration peaks, whereas the Gaussian Plume model simulates a continuous release, thereby smoothing out such peaks. Additionally, HYSPLIT uses a much more detailed simulation, as it incorporates actual weather data for an entire year. The results show that model difference is significant in evaluating detectability and safety. It is important to consider multiple models when evaluating detectability and concentration limits.

Both HYSPLIT and Gaussian Plume modeling have known limitations that can affect the accuracy of the results. Previous work has shown that HYSPLIT may overestimate or underestimate concentrations depending on the stability of meteorological conditions [41]. In one study, HYSPLIT underestimated a measured peak activity concentration by a factor of 12 when simulating air concentrations of Kr-85, though it greatly overestimated the maximum concentration at other times [41]. Similarly, Gaussian Plume modeling can also overestimate or underestimate concentrations. A study conducted in Canada used a steady-state sector-averaged Gaussian plume model with 2006 annual emissions and meteorological data, and then compared the model results to annual average measured tritium concentrations for the same year at 31 locations between 0.25 and 2 km from the site [30]. On average, the model predicted concentrations to be 83% greater than in reality. About 60% of concentrations were within a factor of 2 of actual measured tritium concentrations. This result is consistent with previous studies that found Gaussian plume modeling to give a majority of results within a factor of 2 of measured annual average concentrations around nuclear power plants at low distances [42]. As such, there can exist a large margin of error due to the use of imperfect

atmospheric transport models. The complexity of each model should be carefully assessed when evaluating the resulting tritium concentrations.

Although tritium releases from fission reactors have been documented [43], our comparison includes fusion reactors, which could be a significant source of tritium in the future. As shown in the results, an FPP has the highest resulting concentrations, followed by an HWR, then La Hague, a BWR, and finally a PWR. The resulting concentrations from a fusion reactor were roughly similar to those from an HWR in terms of exceeding the concentration limit and detectability. Therefore, according to this study, resulting concentrations from fusion reactors are not strikingly high compared to other conventional nuclear reactors and facilities.

Differences in detectability are significant depending on the types of detectors, in addition to location of detectors. Based on the results, it would be necessary to use long duration testing to be able to detect any releases from a PWR or BWR (according to the Gaussian Plume model), although even with longer duration testing, they would still only be able to be detected until 1.77 km and 1.91 km at most. Longer duration testing would allow for more detectability and ability to distinguish releases from background overall, as according to HYSPLIT, FPP, HWR, and La Hague would be detectable over the whole simulation period of 10 km. Atmospheric modeling is a useful tool to estimate tritium concentrations [41], which can then be used to design the effective monitoring networks. It is important to have a monitoring network to detect accidental increased releases or any anomalies.

I acknowledge a number of limitations in this study. The atmospheric model is limited to Cambridge, MA, as it uses only meteorological data for this location, and due to the specificity of the data, the results are not generalizable to other locations because the atmospheric transport models are inherently dependent on location-dependent weather data and atmospheric conditions. However, this work provides a practical workflow for computing environmental effects of nuclear facilities given weather data. Additionally, we have not yet analyzed the uncertainty in source terms and other factors and thus need to conduct a sensitivity analysis in the future. Moreover, this study does not take into account tritium speciation (HT vs. HTO). HTO has a higher mobility and transferability, thus giving it a greater radiation impact than HT [44]. Another limitation is that HTO has both a greater wet and dry deposition velocity than HT, meaning that it is more important to take wet and dry deposition into account when modeling the atmospheric transport of HTO than for HT, although this study did not include deposition in the models used [45].

Chapter 6

Conclusion

This thesis has quantified different tritium sources from various types of nuclear facilities and modeled tritium transport in the atmosphere. As a demonstration, we use weather data specific to Cambridge, MA and two types of atmospheric dispersion models, HYSPLIT and a Gaussian Plume model. I have compared the two models as well as the tritium concentrations in the vicinity of each facility. The models show that tritium emissions from most plants are detectable within at least a few km, although this is highly dependent on type of detection. Despite elevated tritium levels near major sources, most emissions remained within regulatory bounds, although proximity to facilities still matters.

The investigation of source terms helps to put into perspective the environmental footprint of different types of nuclear facilities, and can thus play a role in regulatory preparedness and public perception of nuclear facilities. Additionally, this work has implications for future monitoring infrastructure, since tritium detection systems can be placed in locations where the concentration may be high enough to detect.

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