LA-UR- 96-2953

Title: TRITIUM OXIDATION IN ATMOSPHERIC TRANSPORT

Author(s): P.Y. Pan, L. D. Rigdon

Submitted to: OSTI FOR ANNOUNCEMENT AND DISTRIBUTION

Los Alamos
NATIONAL LABORATORY

Form No. 836 R5
ST 2629 10/91

LA-UR- 96-2953

TRITIUM OXIDATION IN ATMOSPHERIC TRANSPORT

by

Paul Y. Pan and L. Douglas Rigdon*

* Nuclear Safety Division, Albuquerque Operations Office, DOE

DISCLAIMER

TABLE OF CONTENTS

List of Tables

List of Figures

ABSTRACT

This study has been performed in support of the DOE/AL Tritium Limits Working Group (TLWG). The goal of the TLWG is to provide a technical basis for nuclear facility tritium limits based on test data, accident experience, and sound analytical modeling.

Measured tritiated water/tritium (HTO/HT) have shown that an HTO/HT conversion ratio of 0.01 would bound all the measured data by a factor of 2 to 500 for a downwind distance of up to 15 km. The 99.7% HTO/HT quantile is 6.1×10^{-3}.

Use of the HTO/HT ratio of 0.01 with site-specific or conservative meteorological data for three Los Alamos tritium facilities would bound the doses calculated based on the dose factors derived from sophisticated tritium transport codes with conservative meteorological and physical conditions. As a result, the proposed methodology, i.e. use of an HTO/HT conversion ratio of 0.01 with site-specific meteorological data or conservative meteorological data would provide an upper bound for off-site doses, and is recommended.

1.0 INTRODUCTION

Tritium has almost the same physical and chemical characteristics as hydrogen. Tritium gas reacts with oxygen to form tritiated water, also replaces hydrogen atoms in organic compounds. Tritiated water is the most abundant chemical form of tritium. Incorporation of tritiated water into the human body can occur by three pathways: 1) lungs respire, 2) gastrointestinal tract ingests, and 3) skin absorbs. Tritium remains in the body in tritiated water and tritium-labeled organic molecules. Tritium is eliminated mostly from the body primarily with a biological half-life of 9.7 d and secondarily with half-lives of 30 d and 450 d.

Major tritium production in nature is from nuclear reactions of atmospheric atoms with cosmic rays in the upper atmosphere. A very minor fraction of natural tritium is produced in the earth's crust from the neutron capture reaction of Li^6 in rocks, where neutrons would be provided by spontaneous fission of uranium and by (α, n) reactions. Man-made activities from nuclear weapon tests and nuclear facilities (nuclear power

plants and tritium production reactors) have produced tritium about 50 times the natural occurring tritium.

Tritium has a specific activity of 9.6E3 Ci/g. The radiological impact of releasing tritium gas (HT) to the atmosphere depends on the rate of formation of tritiated water (HTO) vapor which is approximately 22,000 times more radiotoxic to humans than tritium gas. The dose conversion factor of HTO is about 95 rem/Ci, whereas the dose conversion factor of HT is about 3.5E-3 rem/Ci. As a result, the conversion mechanism of HT into HTO has become an important issue in off-site dose evaluations. Various processes can convert HT into HTO, oxidation in air, exchange reactions with H_2O, photochemical oxidation in air; oxidation in soil, and oxidation in plants.

Three major experiments have been conducted to explore the dominant conversion mechanisms of tritium gas in the atmosphere: 1) Canadian experiment on August 25, 1986: 18.5 Ci was released; 2) French experiment on October 15, 1986: 7,000 Ci was released; and 3) Canadian experiment on June 10, 1987: 95.7 Ci was released. It was found that little conversion of HT to HTO occurred in air and plants. Major conversion took place in the surface layer of soil. The converted HTO was re-emitted into the air from the soil. Also the soil HTO was taken up by plants and subsequently released into the air.

Subsequent to these experiments, many analyses have been performed to study in-field tritium transport and measurement. These studies also include dispersion code development and assessment of tritium transport and conversion in air, plant, and soil. Major tritium transport codes include ETMOD (Russell, 1988), UFOTRI (Raskob, 1990), and H3DISP (Edland, 1990).

2.0 MEASURED HTO/HT CONVERSION RATIOS

Figure 1 shows the measured data (Brown, 1987; Ogram, 1988; Burnham, 1988; Murphy, 1990; Morris, 1976) for HTO/HT from the three major experiments and tritium releases at SRS including an accidental release in 1974. Major findings are summarized as follows:

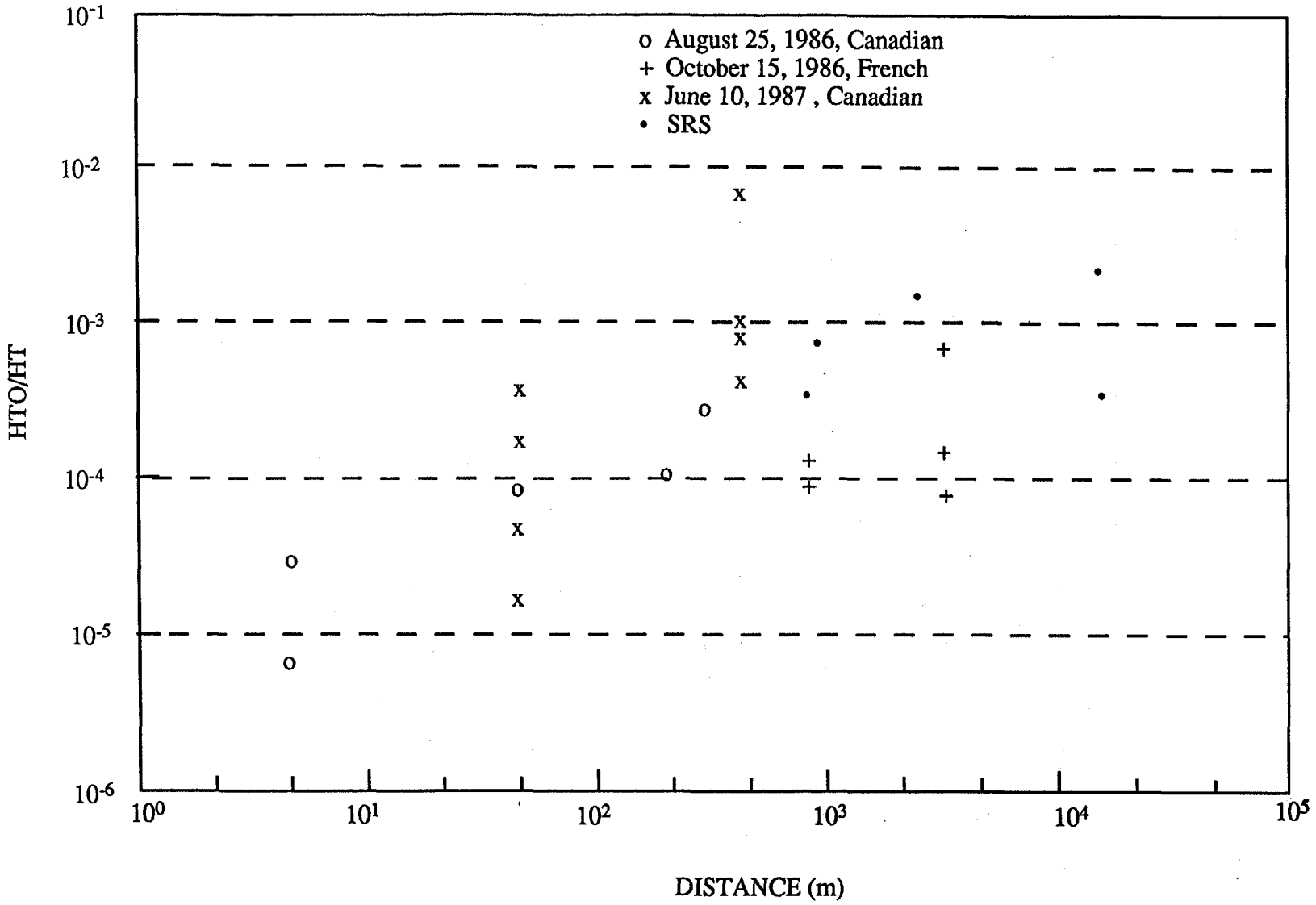

Figure 1. Measured HTO/HT Conversion Ratios

- The measured conversion ratio ranges between 10^{-5} and 10^{-3} for downwind distances of up to 15 km.
- The amount of HTO formed in the atmosphere is negligible. The formation of HTO through chemical kinetics is extremely small.
- Surface soils oxidized HT to HTO via microbial action.
- Part of the HTO formed in soils is re-emitted to the atmosphere. The soil is the dominant source of atmospheric HTO after the HT plume passage.
- Deposition velocities of HT to soil are in the order of 10^{-4} to 10^{-3} m/s, in good agreement among all measuring devices.
- Soil tritium re-emission rates are of the order of a few percent per hour for a period of a day after exposure.
- Wind speed, humidity, release height, and soil properties would dictate the conversion rate.

An HTO/HT ratio of 0.01 would bound all the measured data in Figure 1. The mean and median of the measured HTO/HT conversion ratios are 8.8×10^{-4} and 2.3×10^{-4} respectively. The 99.7% HTO/HT quantile is 6.1×10^{-3}.

3.0 TRITIUM ACCIDENT ANALYSIS

3.1 Tritium Dose Factors

Dose factors are defined as the committed doses in rems per gram of tritium release. Table 3.1 summarizes dose factors from tritium releases in both HTO and HT forms. Derivation of these dose factors is semi-empirical based on key measured parameters (e.g., tritium deposition velocity) in the French and Canadian tests. The complexity of these models ranges from sophisticated, mechanistic computer codes to simple, Gaussian dispersion models.

Table 3.1 Code and Model Predicted Dose Factors for HT and HTO

HT Form (rem/g)	HTO Form (rem/g)	Release Condition	Distance	Comments
1.0E-06, 2 min 4.0E-05, 8 h (1%) 1.0E-04, 8 h (3%) 1.5E-04, 8 h (5%) 3.2E-04, 1 d (5%) 4.5E-04, 3 d (5%) 4.5E-04, 5 d (5%)		2 min, night	1000 m	F stability, 2 m/s, w/wake, V_{HT}=2E-04 m/s, V_{HTO}=1.8E-02 m/s ( ) is for re-emission rate (Raeder, 1988)
4.0E-03		All French & Canadian data	1000 m	F stability, 0.4 m/s (Bartels, 1992)
1.0E-02		2 min, day	800 m	F stability, 0.5 m/s, w/wake (Taster, 1991)
	2.1E-02, 2 min 2.4E-02, 8 h (1%) 3.0E-02, 8 h (3%) 3.4E-02, 8 h (5%) 4.9E-02, 1 d (5%) 6.0E-02, 3 d (5%) 6.1E-02, 5 d (5%)	2 min, night	1000 m	F stability, 2 m/s, w/wake, V_{HT}=2E-04 m/s, V_{HTO}=1.8E-02 m/s ( ) is for re-emission rate (Raeder, 1988)
	1.5E-01	2 min, night	1000 m	F stability, 0.5 m/s, w/wake (Gulden, 1992)
	4.5E-02	1 h, night	1000 m	
	3.9E-03	1 h, rain	1000 m	D stability, 2 m/s, w/wake (Gulden, 1992)
	3.1E-02	1 h, morning	1000 m	E stability, 0.5 m/s, w/wake (Gulden, 1992)
	1.0E-01	2 min, morning	1000 m	
	5.0E-02	Recommended for NET and ITER,	1000 m	(Gulden, 1992)
	6.00E-02	All French & Canadian data	1000 m	F stability, 0.4 m/s, w/wake (Bartels, 1992)
	9.4E-02	2 min, day	800 m	F stability, 0.5 m/s, w/wake (Taster, 1991)

In the mechanistic models, dose factors predicted from the H3DISP and UFOTRI appear to be in good agreement. Both codes have used a relatively conservative meteorological condition as the initial condition for releases. Dose factors of 5% re-emission rate for 8-h exposure would represent the realistic exposure condition and should be used for the short-term exposure analysis.

Other simpler models have used straight forward extrapolation and scaling techniques to derive dose factors from the French and Canadian data. Overly conservative extrapolation schemes have been incorporated into these models to bound the modeling uncertainties. As a result, the final dose factors appear higher than those from the mechanistic dispersion computer codes by a factor of 2 to 25.

At the present time, a dose factor of 0.05 rem/g-HTO is being used to quantify the environmental impact of accidental releases from future fusion facilities like Next European Torus and International Thermonuclear Experimental Reactor. This factor falls in between the doses factors derived from the computer codes and simpler models.

3.2 Proposed Methodology

Doses, as total effective dose equivalent (TEDE), from a short-term exposure from an accidental release of tritium can be calculated based on either measured data or predicted analysis as depicted in Figure 2.

1. TEDE1: Doses are measured based on real-time HTO and HT concentrations.
2. TEDE2: Doses are calculated based on the bounding HTO/HT ratio (i.e., 0.01) and 95th percentile of meteorological data.
3. TEDE3: Doses are calculated based on the bounding HTO/HT ratio (i.e., 0.01) and F stability with 1.5 m/s wind speed.
4. TEDE4: Doses are calculated based on the dose factors using conservative extrapolation and scaling.
5. TEDE5: Doses are calculated based on the dose factors using simulated computer codes.

In theory, using one of the three computer codes, ETMOD, UFOTRI, or H3DISP, that has the capability to model the tritium deposition and re-emission processes would best predict the committed doses. However, using these codes would require excessive resources, e.g., user's fee and verification/validation process. In addition, site-specific data (e.g., weather, soil, vegetation, etc.) also need to be established for the codes.

In reality, no credible accidents in most DOE tritium facilities would involve enough tritium as the material at risk to result in 25 rem at the site boundary. Typical tritium releases at SRS in the past 20 years are summarized in Table 3.2 (Murphy, 1992). Based on the conversion factors in Table 3.1, approximately 700 g (~7 million Ci) of HTO or

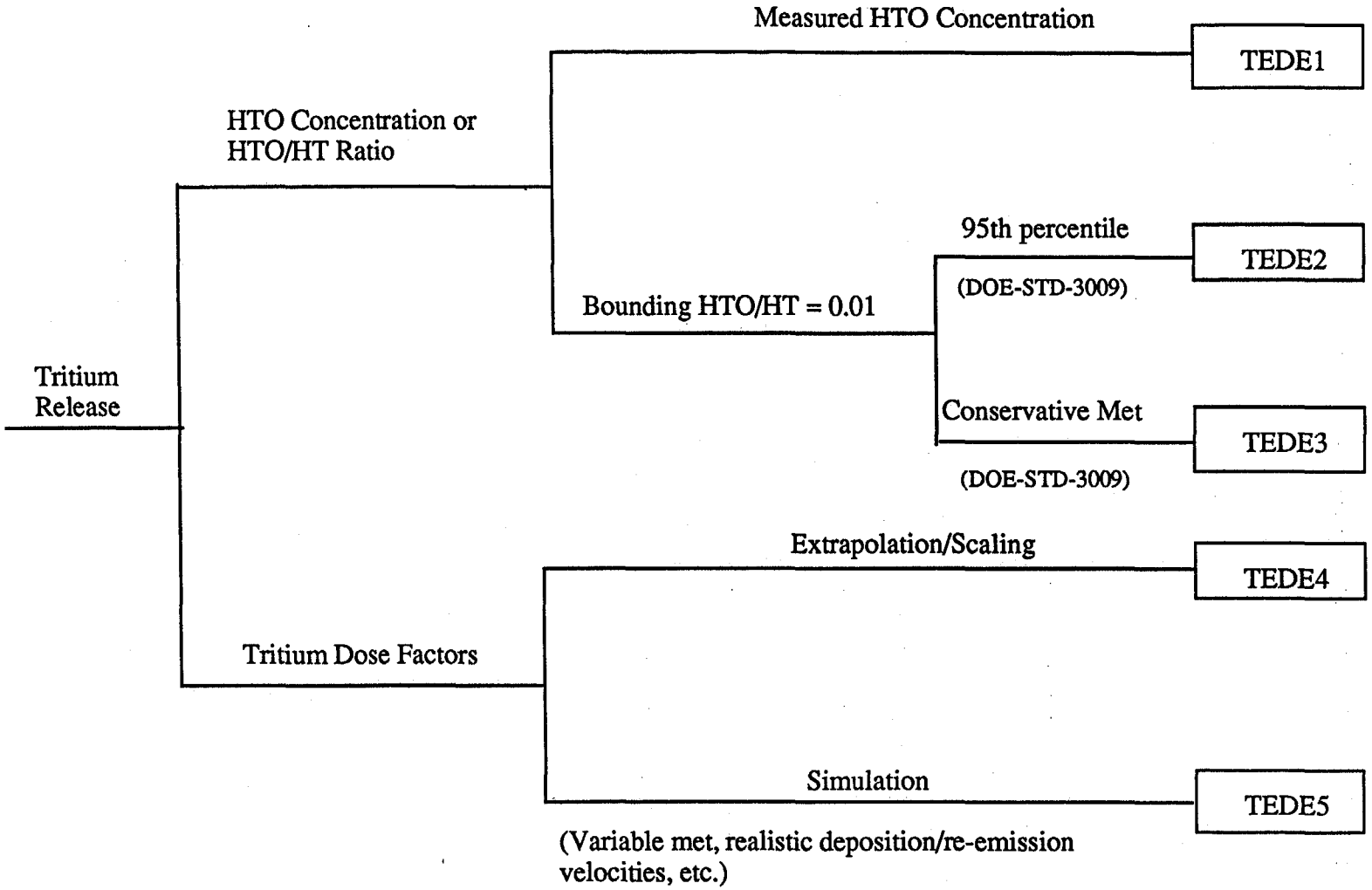

Figure 2. Tritium Dose Calculations

150,000 g of HT (~1.5 billion Ci) need to be released to result in a public dose at 1000 m that exceeds the evaluation guidelines. Because the material at risk in the present operation is typically much lower than the threshold quantities, using a sophisticated mechanistic code to evaluate the off-site doses may not be necessary.

Table 3.2 Tritium Releases at Savannah River Site

Date	Release (Ci)	HTO (%)	HTO (Ci)	Date	Release (Ci)	HTO (%)	HTO (Ci)
5/2/74	479,000	1.0	4,790	3/1/88	20,000	15.0	3,000
12/31/75	182,000	0.6	1,092	6/7/88	3,650	4.0	146
3/27/81	32,934	100.0	32,934	10/6/88	7,086	10.0	700
7/16/83	56,000	1.0	560	12/7/88	3,082	99.5	3,050
3/23/84	7,500	70.0	5,250	1/3/89	500	N/A	N/A
9/7/84	43,800	100.0	43,800	3/30/89	1,100	97.0	1,067
1/31/85	9,300	50.0	4,650	10/19/89	800	100.0	800
3/27/85	19,422	99.9	19,403	2/7/90	100	100.0	100
7/31/87	172,000	2.7	4,644				

A simple and direct application of the HTO/HT conversion ratio with site-specific weather data is proposed to provide a viable method to reasonably calculate the TEDE for a short-term exposure to tritium releases. Alternately, if the site-specific weather data are not available, a simple Gaussian straight-line dispersion should be used.

3.3 Sensitivity Study

Doses are calculated using the basic equation:

$$D = M * SP * DCF * X/Q * BR$$

where

D is the 50-yr TEDE, rem;

M is the material released, 1 g either in HT or HTO form;

SP is the specific activity, 9.6E3 Ci/g;

DCF is the dose conversion factor for HT, 3.5E-3 rem/Ci; or 95 rem/Ci for HTO;

X/Q is the atmospheric dispersion factor, s/m^3; and

BR is the breathing rate, 3.5E-4 m^3/s.

Doses are calculated and listed in Table 3.3 to compare with those calculated based on a method using HT and HTO dose factors (rem/g HT or HTO released) directly derived from the dispersion computer codes: 1.5E-4 for HT and 3.4E-2 for HTO. It is important to note that these dose factors are derived based on very conservative meteorological conditions and deposition velocities. Three tritium facilities, Tritium Science and Fabrication Facility (TSFF), Tritium System Test Assembly (TSTA), and Weapon Engineering Tritium Facility (WETF), at Los Alamos National Laboratory are used for the analysis. The 95th-percentile X/Q is generated from a dispersion code, GENII (GENII-S, 1992). Doses are calculated for various combinations of the HT and HTO ratios in the source term.

Table 3.3. Comparison of TEDE (in rems)

	HTO/HT= 0.01						DOSE FACTORS	
	W/ WAKE				W/O WAKE		W/WAKE	W/O WAKE
	RG1.145	TSTA*	TSFF*	WETF*	RG1.145	TSTA*, TSFF*, WETF*		
100% HTO	3.8E-02	1.4E-01	3.2E-02	4.8E-02	1.4E-01	2.3E-01	4.4E-02	1.4E-01
99%HTO, 1%HT	3.8E-02	1.4E-01	3.2E-02	4.7E-02	1.4E-01	2.2E-01	4.4E-02	1.4E-01
90%HTO, 10%HT	3.5E-02	1.2E-01	2.9E-02	4.3E-02	1.2E-01	2.0E-01	4.0E-02	1.3E-01
50%HTO, 50%HT	1.9E-02	6.9E-02	1.6E-02	2.4E-02	6.9E-02	1.1E-01	2.2E-02	7.1E-02
10%HTO, 90%HT	4.2E-03	1.5E-02	3.5E-03	5.2E-03	1.5E-02	2.5E-02	4.6E-03	1.5E-02
1%HTO, 99%HT	7.6E-04	2.7E-03	6.4E-04	9.5E-04	2.7E-03	4.5E-03	6.4E-04	2.0E-03
100%HT	3.8E-04	1.4E-03	3.2E-04	4.8E-04	1.4E-03	2.3E-03	2.0E-04	6.2E-04
* GENII-S results								

Examination of Table 3.3 indicates:

- Doses (at 1000 m) from using HTO/HT= 0.01 and the GENII 95th-percentile weather (with and without building wake) are approximately equal to or larger (by a factor of up to 10) than those using HT and HTO dose factors.
- Doses (at 1000 m) from using HTO/HT = 0.01 and a simple Gaussian model with building wake effects (Turner, 1994; USNRC, 1982) are appropriately equal to or larger (by a factor of up to 4) than those using HT and HTO dose factors.

For all combinations of HT and HTO ratios in the initial source term, the off-site doses from using the 0.01 conversion ratio with site-specific or conservative methodology are approximately equal to or larger, by a factor of 10, than those from using the dose factors based on sophisticated tritium codes.

4.0 CONCLUSIONS AND RECOMMENDATIONS

Experiments and tests have shown that the HTO/HT conversion ratio of 0.01 would bound all the measurements by a factor of 2 to 500 for a downwind distance of up to 15 km. The 99.7% HTO/HT quantile is 6.1×10^{-3}. Use of the HTO/HT ratio of 0.01 with site-specific or conservative meteorological data for three Los Alamos tritium facilities would bound the doses calculated based on the dose factors derived from sophisticated tritium transport codes with conservative meteorological and physical conditions. As a result, the proposed methodology, i.e. use of an HTO/HT conversion ratio of 0.01 with site-specific meteorological data or conservative meteorological data would provide an upper bound for off-site doses, and is recommended.

REFERENCES

H. W. Bartels, Max Plank Institut, "A Simple Model to Calculate Doses for Acute Tritium Releases, Based on HT Field Experiments in Canada and France," Fusion Technology, **21** (March 1992).

R. M. Brown, G. L. Ogram, and F. S. Spenser, " Field Studies of HT Oxidation and Dispersion in the Environment I, the 1986 August Experiment at Chalk River," AECL, Canada (January 1987).

C. D. Burnham, R. M. Brown, G. L. Gram, and F. S. Spenser, " An Overview of Experiments at Chalk River," Fusion Technology, **14** (September 1988).

O. Edlund, "H3DISP, A Computer Program for Calculation of Consequences Attributed to Atmospheric Release of Tritium," STUDSVIK/NS-90/89.

GENII-S, A Code for Statistical and Deterministic Simulations of Radiation Doses to Humans from Radionuclides in the Environment, SAND91-0561A, UC-721 (October 1992).

W. Gulden, Max Plank Institut, and W. Raskob, Klernforschungszentrum, "Accident Tritium Doses Based on Realistic Modeling," Fusion Technology, **21** (March 1992).

O. M. Morris and R. A. Scaggs, "Tritium Release from the SRP," DPWD-1375 (November 1976).

C. E. Murphy, Jr., "Estimating the Dose from Atmospheric Releases of HT," WSRC-TR-90-538, Westinghouse Savannah River Company (November 1990).

C. E. Murphy Jr., L. R. Bauer, and D. D. Hoel, "Tritium Atmospheric Transport and Deposition Following Acute Releases-Comparisons with A Simple Transport Model," Fusion Technology, **21** (March 1992).

G. L. Ogram, " The French Experiment on Environmental Tritium Behavior, October 15, 1986," CFFTP-88043 (December 1988).

J. Raeder and W. Gulden, Max Plank Institut, "NET Analysis and the European Safety and Environmental Programme," 15th SOFT Conference (December 1988).

W. Raskob, "UFOTRI, Program for Assessing the Offsite Consequences from Accidental Tritium Releases," KfK 4605 (July 1990).

S. B. Russell and G. L. Ogram, "Modeling Elemental Tritium Deposition, Conversion, and Re-Emission Using Ontario Hydro's Tritium Dispersion Code," Fusion Technology, **14** (1988).

Michael Taster and Claus Bunnengerg, University Hannover, and Werner Gulden, Max Plank Institut, "Maximum Permissible Amounts of Accidentally Released Tritium from An Environmental Experiment to Meet Dose Limits for Public Exposure," Fusion Technology, **20** (1991).

D. B. Turner, Workbook of Atmospheric Dispersion Estimates, An Introduction to Dispersion Modeling, 2nd Edition, Lewis Publishers (1994).

USNRC, Regulatory Guide 1.145, "Atmospheric Dispersion Models for Potential Accident Consequence Assessments at Nuclear Power Plants" (November 1982).