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# **Tritium Radioluminescent Devices Health and Safety Manual**

**R. J. Traub  
G. A. Jensen**

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**June 1995**

**Prepared for the U.S. Department of Energy  
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest Laboratory  
Operated for the U.S. Department of Energy  
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Pacific Northwest Laboratory  
Richland, Washington 99352

**RADIOLUMINESCENT (RL) - The property of a material to emit light at ambient temperature when bombarded by energy resulting from decay of radioactive material**

# Summary

For over 30 years, the U.S. Department of Energy (DOE) and its predecessor agencies, have investigated the science and technology of materials that are produced during nuclear processes in an effort to find beneficial uses for society. This work has made substantial contributions to science, medicine, industrial processing and other products. Some of these processes are complex, such as the use of monoclonal antibodies for treatment of cancer, and others are relatively simple, such as the sterilization of medical and food products by exposing them to gamma radiation to avoid the spread of disease. Other applications are functionally complex but simple in use, such as the household smoke detector that uses radioactive americium to reliably detect the products of combustion, saving lives and property. The use of RL technology is one of these applications. The physical process that produces light is similar to that of the smoke detector but its use is simple and requires no external power supply.

Tritium-powered radioluminescent (RL) lighting has been commercially available for over 25 years. In existing commercial RL technology, glass tubes coated internally with a phosphor are filled with tritium and sealed. The tubes are then mounted into plastic or other housing materials. The initial quantities of tritium contained in these devices have ranged from a few millicuries in watch dials and other instrument illuminators to over 30 Ci in commercially available exit signs.

This document consolidates available information on the properties of tritium, including its environmental chemistry, its health physics, and safe practices in using tritium-activated RL lighting. It also summarizes relevant government regulations on RL lighting. Chapters are divided into a single-column part, which provides an overview of the topic for readers simply requiring guidance on the safety of tritium RL lighting, and a dual-column part for readers requiring more technical and detailed information.

Interest has been growing in using RL technology for lighting remote and military aviation runways and helipads in remote locations where electric power is difficult to obtain or perhaps difficult to maintain once acquired. It is also useful as temporary and portable airfield lighting until permanent lighting can be installed at these locations. Demonstration airfield runway and helicopter landing pad lighting systems have been installed at Eskimo villages and other remote locations in Alaska and Canada and have served these locations on a semipermanent basis. Radioluminescent lighting is also useful where highly corrosive soil corrodes buried electrical conduits and wiring. Its independence of electrical sources also has made RL lighting useful in emergency exit lighting for military and civilian vehicles and ships.

When it decays, tritium emits a low energy (maximum 18.4 keV) beta particle and an antineutrino. This beta particle (electron) has an energy less than that emitted from an electron gun in a computer monitor or television set. The relative safety of the use of tritium-activated RL devices is demonstrated by the fact that there have been very few serious incidents involving tritium that have been documented since its discovery in 1939 in the laboratory and in nature in 1949. To be sure, there have been tritium releases from damaged RL devices but the exposures have been very limited and have caused essentially no injury.

There are concerns by users of RL technology and these are justified. The use of RL technology must be truly beneficial when compared to health, safety, and environmental risk that may be created. It must be used knowledgeably and with care. There has been a need for a compilation of information on the health and safety requirements tritium use in RL devices for a long time. It was to meet this need that this document was prepared.

This document is intended to be a consolidation of the essential available information on tritium's properties and environmental chemistry, its health physics, and safe practices in using tritium-activated RL lighting.

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- Robin L. Hill, for her contributions on tritium dosimetry.



# Acronyms

AGR	advanced gas cooled reactors
ANSI	American National Standards Institute
APHS	
ASTM	American Society for Testing Materials
BWR	boiling water reactors
CANDU	Canadian Deuterium Uranium
CFR	Code of Federal Regulations
DEB	1,4-di-(2)-phenylethynyl benzene
DOD	U. S. Department of Defense
DOE	U. S. Department of Energy
FBR	fast breeder reactor
FR	Federal Register
GCR	gas-cooled reactor
GI	gastrointestinal tract
ICRP	International Commission on Radiation Protection
IAEA	International Atomic Energy Agency
LLNL	Lawrence Livermore National Laboratory
LSC	liquid scintillation counting
NCRP	National Council on Radiation Protection and Measurements
NRC	U.S. Nuclear Regulatory Commission
PET	positron emission tomography
PHWR	pressurized heavy water reactor
PNL	Pacific Northwest Laboratory
PVA	polyvinyl
PVC	polyvinylchloride
PWR	pressurized water reactor
RL	radioluminescent
RPO	radiation protection officer
RPT	radiation protection technician
RSO	radiation safety officer
SEE	specific effective energy

TU tritium unit

WHO World Health Organization

# Symbols Used

## Chemical Symbols

D	deuterium
H	hydrogen
<sup>3</sup> He	helium atomic weight 3
<sup>85</sup> Kr	krypton-85
Hg	mercury
O	oxygen
<sup>147</sup> Pm	promethium-147
<sup>226</sup> Ra	radium-226
T or <sup>3</sup> H	tritium

## Symbols Used in Equations

A	individual's age, y
A	area, m <sup>2</sup> , cm <sup>2</sup>
a	atmosphere pressure, atm
BR	breathing rate, m <sup>3</sup> /sec
C	air concentration of tritiated water, μCi/mL
Ci	curie, 3.7 x 10 <sup>10</sup> disintegrations/second
C <sub>i</sub>	coefficient of the ith compartment
°C	degrees centigrade
cm	centimeter
cmHg	centimeters of mercury pressure
c/Q	relative integral air concentration, sec/m <sup>3</sup>
dpm	disintegrations per minute
e	radiation energy absorbed in the organ, MeV
eV	electron volt
g	gram
g-Atom	one gram atomic weight of an element
g-Mole	one gram molecular weight of a substance
H	dose equivalent rate, rem/day
H <sub>e</sub>	committed effective dose equivalent, rem
H <sub>T,50</sub>	committed dose equivalent, rem
h	hour
K	permeation constant
°K	degrees kelvin
k	kilo
keV	kilo electron volts
kg	kilograms
I <sub>e</sub>	retention coefficient for tritium, effective half time of tritiated water in body, days

$\lambda_e$	effective removal constant, $\text{time}^{-1}$
$\lambda_l$	exfiltrate removal constant, $\text{time}^{-1}$
$\lambda_p$	physical decay constant, $\text{time}^{-1}$
M	general symbol for a metal cation
M	mega
M	mass of the organ, g
M	mass of soft tissue, g
MeV	million electron volts
m	meter
mL	milliliter
mm	millimeter
mol	mole pressure
mrem	millirem
N	north
n	nano
Pa	pressure in Pasquills
p	pico
R(t)	retention of tritiated water at time t, in $\mu\text{Ci}$
Q	quality factor, dimensionless
q(t)	activity of radioactive material in the tissue, $\mu\text{Ci}$
q(0)	initial amount of activity in the body, or at time of intake, $\mu\text{Ci}$
s or sec	second
T	exposure time, min
$T_e$	effective half-time of tritium in the body, days
t	time, sec, min, h, day, y
U	wind speed(source to receptor), m/sec
V	volume, L
W	watts
W	individuals weight, kg
y	year

### Subscripts

h	high pressure side (pressure)
l	low pressure side (pressure)
$t_1$	at time 1
$t_2$	at time 2

### Greek Letters

$\tau$	antinutrino, flow in diffusion
$\beta$	beta particle
$\sigma_y$	horizontal dispersion coefficient, $\text{m}^{-1}$
$\mu$	micro
$\lambda_i$	retention coefficient for the ith component, $\text{days}^{-1}$
$\sigma_z$	vertical dispersion coefficient, $\text{m}^{-1}$

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## **1.0 Introduction**

# 1.0 Introduction

For over 30 years the U.S. Department of Energy and its predecessor agencies have investigated the science and technology of materials that are by-products of nuclear processes in an effort to find beneficial uses for them. This work has made a substantial contribution to science, medicine, industrial processing, and products. Many of these applications are extremely complex, such as Positron Emission Tomography (PET) Scan, a diagnostic technique that uses gamma emissions to study human metabolism. Some are simple in concept, such as the sterilization of products by exposing bacteria to gamma radiation to avoid the spread of disease. Still other applications are complex in function but simple in use; the common household smoke detector uses radioactive americium to reliably detect the ionized products of combustion, saving life and property.

Radioluminescent (RL) devices produce light by a fairly complex physical process. The processes that are used in RL devices are similar to smoke detectors. Both serve simple yet important purposes, such as emergency exit signs in airplanes and smoke (fire) detection in homes. However, as with any application of technology, the use of nuclear by-product material must be truly beneficial when compared to health, safety, and environmental risk that the use may create. The frivolous use of radioactive material should not be tolerated in a responsible society. Neither should the responsible use of radioactive material be rejected in society when the benefit clearly outweighs the risk.

## 1.1 Uses of RL Lighting

Interest has been growing in using RL technology for lighting civilian and military runways in remote locations where electric power is difficult to obtain and perhaps difficult to maintain once acquired. Demonstration airfield runway lighting systems have been installed at Eskimo and other remote communities in Alaska and are now serving several Alaskan locations on a semipermanent basis. Radioluminescent lighting is also very useful where highly corrosive soil corrodes buried electrical conduits and wiring. Being independent of electrical sources has also made RL lighting useful in emergency exit lighting for military vehicles and ships. Radioluminescent lighting has been tested and approved by the U.S. Air Force for use in base and tactical runways.

Radioluminescent lights have been used in safety and other applications since the early 1960s. Their largest use today is for emergency exit signs in aircraft, buildings, and small hand-held military lighting applications. While there have been releases of tritium from damaged exit signs or other lighting uses, there have been no fatalities related to the use of tritium for RL lights. Considering that the beta particle released during tritium decay has a maximum energy of 18.6 keV, less than that of the electron released in the electron gun in a television or computer monitor, this is not surprising.

Published information on releases is sparse, but in general those receiving exposures have not been harmed. Seelentag (1973) attributed two deaths to tritium exposure. Both individuals had, however, worked with other radioactive isotopes for many years prior to their exposure to tritium. Their exposure to rather high levels of tritium was in an uncontrolled laboratory over a long period of time. Because of their long-term exposure to other radioisotopes, the conclusion was that while the tritium exposure was a contributor in the individuals' deaths, it was not the only cause. The only release in which individuals received significant exposures to tritium and were carefully monitored occurred in Switzerland in a RL light manufacturing facility (Lloyd et al. 1986). During the final stage of filling RL lights with tritium, a worker was required to heat the filled tube with a gas torch to seal the glass container. However, the system was pressurized and when the glass was heated and became soft, tritium escaped. The worker received an estimated dose of 80 R that was reduced to about 40 R by dialysis treatment in a

local hospital. She was also monitored for a period of 6 months following the incident and no adverse health problems were observed. The conclusion from the paper was that the treatment was somewhat extreme and that by forcing liquids the same reduction in dose could have been achieved.

Additional releases have occurred at DOE facilities over the years since tritium was discovered in the laboratory in 1939 and in nature in 1949. These releases are reviewed in Section 3.0. No serious injury to the environment or individuals was identified in these documents.

## 1.2 Structure of This Document

This document is intended to consolidate available information on tritium's properties and environmental chemistry, its health physics, and the safe practices in using tritium-activated RL lighting.

The sections that follow are divided into three major parts: 1) the properties of tritium and its behavior in the environment, 2) radiation protection and health physics concerns in handling RL lighting, and 3) the Federal regulations applicable to RL lighting. Most chapters comprise two parts: an introductory section (printed in single column format) that provides an overview of the topic of that chapter, and a two-column section that provides more technical information. Those readers wanting an overview of the topics discussed can find sufficient information in the introductory sections of the chapters. Health physicists, radiation protection technologists, and others requiring more detailed information will probably want to read each chapter in its entirety.

## 1.3 References

Lloyd, L. L., A. A. Edwards, J. S. Prosser, A. Aufder Maur, A. Etzweiler, U. Weikhardt, U. Gossi, L. Geiger, U. Noelppt, and H. Rosler. 1986. "Accidental Intake of Tritiated Water: A Report of Two Cases," *Radiation Protection Dosimetry*, 15(3):191-196.

Seelentag, W. 1973. "Two Cases of Tritium Fatality." In *Tritium*, eds. A. A. Moghissi and M. W. Carter, pp. 267-280. Messenger Graphics Publishers, Phoenix, Arizona.

## **2.0 Properties of Tritium**

## 2.0 Properties of Tritium

Tritium chemistry is the same as that of hydrogen, except that tritium is radioactive and has a larger mass that affects its reactions. As the radioactive isotope of hydrogen, tritium decays by the following reaction:



where  $\beta$  is a beta particle (electron) and  $\nu$  is an antineutrino. The progeny of tritium decay is helium-3, which is not radioactive.

### 2.1 Common Forms of Tritium

There are several forms of tritium, the most common being  $T_2$  and TH (gases) and tritiated water (THO or  $T_2O$ ). Tritiated water is the most prevalent form of tritium in the environment (THO). Table 2.1 identifies important radiological properties of tritium. It is important to note that when tritium decays, no gamma ray is emitted but some x-rays are.

Radioluminescent lights are normally filled with very pure tritium gas. However, the tritium compounds of primary interest for RL safety and usage are elemental tritium and THO.

**Table 2.1.** Radiological Properties of Tritium

Radiation Emitted	Beta (No Gamma)
Beta energy	
Maximum energy	0.0186 MeV <sup>(a)</sup>
Mean decay energy	0.00568 MeV <sup>(b)</sup>
Half-life	12.3 y <sup>(a,b)</sup>
Disintegration probability	$1.78 \cdot 10^{-9}$
Specific activity ( $T_2$ gas)	$9.62 \cdot 10^3$ Ci/g
Specific activity (THO)	$1.45 \cdot 10^3$ Ci/g
Power density ( $T_2$ gas)	$3.24 \cdot 10^{-1}$ W/g
Activity density ( $T_2$ gas, 1 atm)	2.59 Ci/cm <sup>3</sup> (0°C) 2.37 Ci/cm <sup>3</sup> (25°C) 2.28 Ci/cm <sup>3</sup> (37°C)
(as $T_2$ molecules)	$5.80 \cdot 10^4$ Ci/(g-mole)
(as T atoms)	$2.90 \cdot 10^4$ Ci/(g-atom)

(a) From Lederer and Shirley (1978).

(b) From Souers, pp. 207-8 (1986).

## 2.2 Hazards of Tritium

Elemental tritium gas ( $T_2$ ) is the major portion of the total activity in RL lights. Although a very small quantity of tritiated water (either in the THO or  $T_2O$  form) is found, it is only a very small portion of the total activity of an RL light. Tritium gas that is released to the environment will eventually form THO. Tritiated water is the greater hazard to health and safety because it is absorbed instantaneously in the lungs and through the skin and is retained by the body with a half-life of 10 days. The gas states ( $T_2$  or TH), which are only minimally absorbed and quickly eliminated from the body, are four orders of magnitude less dangerous than the aqueous forms. Although tritium may enter the body by several paths (ingestion, inhalation, injection, or percutaneous absorption), it is important to note that intakes can occur only if an RL light is cracked or broken. The other reactions may occur within a light unit during its service life, but only the THO that is produced or contained in a device becomes important when the unit is returned for recycle or disposal, or is destroyed.

If the tritium is released indoors, the conversion rate from the gas phase to THO is accelerated because of temperature, humidity, catalytic surfaces, electric discharges from electrical equipment, and other factors related to ventilation. However, tritium gas released directly to the atmosphere reacts very slowly to form tritium oxide (THO). The THO concentration produced depends on how rapidly tritium disperses into the atmosphere when released. Assuming that dispersion is rapid in an atmospheric release, the exchange of tritium to produce THO will take many hours (perhaps years) before significant quantities build up. In recent studies, tritium was purposely released to the atmosphere, as would occur in the destruction of an RL device, and was monitored downwind for several kilometers. Researchers reported that the only THO or tritiated water vapor above background tritium identified was at ground level, probably produced from natural conversion by bacteria in the soil (Brown et al. [1988] and Paillard et al. [1988]).

## 2.3 Properties of Tritium in Radioluminescent Devices

The chemistry inside RL devices is important, as well. Because tritium is radioactive and has a high specific activity, radiolysis occurs. The tritium interacts with the binders (used to attach the phosphor to the glass tubing in RL lights) and with surface impurities to produce THO and other tritiated compounds (see Table 2.2). Tritium may also react with the binder or the phosphor and be retained in the solid. The chemistry of these reactions inside RL devices is not well understood now and may be a safety concern for handling damaged light units or for disposing of them. Some data have been obtained on the products of reactions inside RL devices by destroying small RL devices (two aiming posts and four range indicators) and 50-Ci RL sources produced at the Oak Ridge National Laboratory 2 to 4 years after their manufacture and subjecting the resulting gas to mass spectrographic analyses. Data from these analyses shown in Table 2.3 provide evidence that tritium may have reacted with something attached to or inside the tube wall. Also, some commercial RL units have been broken and more contamination was found than would be expected from a release involving only  $T_2$  gas (Bowerman and Czajkowaski 1990). The NRC commissioned Brookhaven National Laboratory to determine the chemical forms and quantities of the tritium in each of four building exit signs. Tritiated water content in these signs ranged from 2% to 12.2%. The newest signs (2 years old) had the least tritiated water and the oldest sign (14 years old) had the greatest amount of tritiated water (Bowerman and Czajkowaski 1990).

The development of solid-state RL devices by incorporating tritium into inorganic or organic materials should improve the safety of these devices because the tritium will be contained in a chemical structure rather than as a gas in a glass container.

**Table 2.2.** Analysis of Pure Tritium Gas for RL Applications<sup>(a)</sup>

Element/ Compound	Mole %	Element/ Compound	Mole %
N <sub>2</sub>	0.01	HDO	0.01
O <sub>2</sub>	0.00	H <sub>2</sub> O	0.00
Ar	0.00	TH	0.64
H <sub>2</sub>	0.02	DT	2.08
D <sub>2</sub>	0.01	THO	0.03
HD	0.00	T <sub>2</sub> O	0.01
T <sub>2</sub>	97.19		
Total T ( <sup>3</sup> H) 98.60			

(a) Data supplied by J. A. Tompkins, ORNL, personal communication, 1988.

**Table 2.3.** Comparison of Total Tritium in RL Devices by Calorimetry and PVT/MS Analysis of Gas Released on Fracture (no bakeoff)<sup>(a)</sup>

Item	Calorimetry (Ci)	PVT/MS (Ci)	Percent on Walls (%)	A/V
Aiming Post (Green)	8.39 ± 0.04	8.13 ± 0.12	3	7
Aiming Post (Orange)	7.89 ± 0.06	7.53 ± 0.11	5	7
Range Indicator 1	0.55 ± 0.05	0.49 ± 0.01	11	20
Range Indicator 2	0.59 ± 0.05	0.49 ± 0.01	17	20
Range Indicator 3	0.71 ± 0.03	0.65 ± 0.01	8	20
Range Indicator 4	0.73 ± 0.05	0.60 ± 0.01	18	20

(a) The difference between calorimetry and PVT/MS is a measure of tritium adsorbed on the walls. Residual tritium on the walls is proportional to the ratio of surface area to volume (A/V).

The following sections cover in more detail the radiological, physical, and chemical properties of tritium and the effects of tritium in the body.

## 2.4 Radiological Properties of Tritium

The following sections discuss the significant radiological properties of tritium, including its recoil energy, its bremsstrahlung and fluorescence, and the range and interactions of its beta radiation.

### 2.4.1 Recoil Energy

When the tritium electron is released during the decay process, momentum (kinetic energy) is imparted to the daughter  $^3\text{He}$  nucleus by emission of the electron and there is a disturbance in the electron cloud surrounding the nuclei due to the sudden increase in the nuclear charge ("shaking effect"). The energy absorbed by the nucleus in this process is called recoil energy and has a maximum 3.6-eV energy (Cacace 1975). Theoretical calculations show that 75% to 80% of the decay ions are formed in their ground state and receive negligible or recoil excitation, most often no more than 0.08 eV (Cacace 1975). The effects of these processes are not fully known for complex molecules except that  $^3\text{He}^+$  and an electron will be produced. The  $^3\text{He}^+$  may react with hydrogen if present and can cause biological damage. Although the maximum recoil energy of 3.6 eV is normally insufficient for bond rupture (normally carbon-carbon bonds require at least 4 eV) and the primary effects of decay in chemical structures appear to be minimal (Evans 1974), further studies must nonetheless be done for advanced solid-state RL lighting and its applications.

### 2.4.2 Bremsstrahlung and Fluorescence

When the beta particle released by tritium decay decelerates as it penetrates matter, it produces additional radiation, either bremsstrahlung (x-rays) or fluorescence (Souers 1986). The x-rays produced are more penetrating than the original beta particle. It has been shown, for instance, that for solids, a beta particle having an energy of 20 keV will produce x-rays that penetrate 1500  $\mu\text{m}$  through  $\text{SiO}_2$  but only 6.8  $\mu\text{m}$  through gold; the ability to absorb x-rays goes

up rapidly with  $Z$  (atomic number). The maximum energy of tritium betas is 18.6 keV.

Because the average tritium beta has an energy of 5.6 keV and produces x-rays that penetrate approximately 28  $\mu\text{m}$  through  $\text{SiO}_2$ , the glass containment should shield most of the bremsstrahlung produced in existing RL lights. However, the characteristic x-rays from the ZnS produced by the decay of the small amount of tritium contained in tritium-activated RL watches has been known to set off radiation detection devices at nuclear power plants.

### 2.4.3 Range and Interactions of Radiation

Because of the very limited energy of its beta particle, tritium cannot be detected at great distances from its source. Table 2.4 shows the range of the tritium beta particle in water and air. The very short range of the tritium beta particle is the reason that it is difficult, if not impossible, to detect tritium on surfaces by means of survey instruments. It is common to hold the instrument 2 to 3 cm from the surface being measured, but this distance would be greater than the range of the tritium beta particle in air.

**Table 2.4.** Range of Tritium Beta Particle (cm)

Medium	$1.8 X_{90}$ <sup>(a)</sup>	Range <sup>(b)</sup>
Air	no data	$5.6 \times 10^{-1}$
Water	$3.9 \times 10$	$5.8 \times 10^{-4}$

(a)  $1.8 X_{90}$  is an arbitrary value chosen by Berger (1971) as a cutoff point for dose calculation purposes; according to his methods, insignificant radiation doses accrue beyond this point.

(b) From Tables 3 and 4 in Cross et al. (1982), ~99.5% of all beta energy is deposited within a sphere having this radius.

## 2.5 Physical Properties of Tritium

Being an isotope of hydrogen explains some of tritium's physical properties. Isotopic effects help to explain the distribution of natural tritium in the environment, for instance, and the triple-point temperatures of THO and T<sub>2</sub>O, which are both higher than that of water.

The natural abundance of the three hydrogen isotopes is found in Table 2.5. Selected physical properties of the oxides of the isotopic forms of hydrogen containing tritium are listed in Table 2.6. Physical properties of the isotopic forms of elemental hydrogen containing tritium are also shown in Table 2.7.

### 2.5.1 Diffusion of Tritium Through Materials

A very important characteristic of the hydrogen isotopes is their ability to permeate materials that contain them. Currently, for RL lights, we have only the information developed by Tompkins (1988) from which to extrapolate. Tompkins completed some permeation experiments for the U.S. Air Force at room temperature and found that the actual permeation rate was at least two orders of magnitude below that predicted for tritium contained in Pyrex tubes. The diffusion rate of tritium gas through the glass containers used for light sources can be described by Equation (2.2):

**Table 2.5.** Natural Abundance of Hydrogen Isotopes<sup>(a)</sup>

Isotope	Atom (%)
Hydrogen ( <sup>1</sup> H, protium)	99.985
Deuterium ( <sup>2</sup> H)	0.015
Tritium ( <sup>3</sup> H)	5.10 · 10 <sup>-18</sup> (approx.) <sup>(b)</sup>

(a) Dean (1978), Evans (1974).

(b) Before weapons testing.

**Table 2.6.** Selected Properties of the Oxides of Hydrogen Isotopes

Property	THO	H <sub>2</sub> O	D <sub>2</sub> O	T <sub>2</sub> O
Boiling Point (°C)	100.76	100.00	101.42	101.51
Triple-Point Temperature (°C)	2.25	0.01	3.82	4.49
Triple-Point Pressure (mm Hg)	4.73	4.58	5.02	4.87
Heat of Vaporization at the Boiling Point (kcal/mole)	9.91	9.72	9.90	10.10

**Table 2.7.** Properties of the Selected Isotopic Forms of Hydrogen<sup>(a)</sup>

Property	H <sub>2</sub>	D <sub>2</sub>	T <sub>2</sub>	TH	DT
Triple Point (°K)	13.96	18.73	20.62	17.70	19.79
Boiling Point (°K)	20.41	23.67	25.04	22.91	24.37
Critical Temperature (°K)	33.24	38.35	40.44	37.13	39.42
Critical Pressure (MPa)	1.298	1.665	1.850	1.571	1.773

(a) For H<sub>2</sub>, D<sub>2</sub>, and T<sub>2</sub>, ortho and para forms exist.

The data provided are for the normal equilibrium mixture.

$$\frac{d\tau}{dt} = \frac{KA(P_h - P_l)}{L} \quad (2.2)$$

where  $d\tau$  = the tritium flow out of the low pressure side

$K$  (mol/m·s·Pa) = the permeability constant

$L$  = the thickness of the permeable ceramic or glass material (m)

A = the area through which the tritium is permeating (m<sup>2</sup>)  
P<sub>h</sub> and P<sub>l</sub> = the pressures on the high-pressure and low-pressure sides (psi), respectively.

Permeability constants for selected glasses are presented in Table 2.8 for permeability constant, K. If the specific activity of tritium is taken into account, K can be calculated as shown in the second column in Table 2.8.

Because hydrogen diffuses through glass as a molecule rather than an atom, the tritium diffusing from the glass container will be in the form of the diatomic elemental gas (Souers 1986).

### 2.5.2 Solubility in Metals and Adherence to Paint and Metals

An experiment that investigated the evolution of tritium from stainless steel planchets that had been exposed to elemental tritium found that at 350°C, tritium begins to evolve after about 15 minutes and is essentially completed after 1 hour. Tritium loss rates were observed to be 0.17%/day for planchets that were degreased after exposure and 0.91%/day for planchets that were not cleaned after exposure to the tritium gas (Surett and McElroy 1988).

Later studies by Dickson and Miller (1991) and Antoniazzi et al. (1991) evaluated the adsorption/absorption of HT and HTO on aluminum, brass, copper, mild steel, tin-plated steel, galvanized steel, and stainless steel. Dickson and Miller (1991) also looked at nonmetallic building materials such as concrete, vinyl floor tile, porcelain tile, painted Gyproc, and unpainted concrete. Aluminum samples painted with an oil-base semi-gloss paint with an undercoat of primer were also evaluated. Most Gyproc samples were coated with an interior latex satin finish paint. In addition, Ono et al. (1991) evaluated absorption and desorption of tritiated water on paints.

The following characteristics of tritium that adheres to paint and metal have been observed experimentally:

- Tritiated water sorbed to the surface of enamel paint can be desorbed by heating the paint at 250°C. Adsorption and desorption of tritium for acrylic paints was smaller than for silicone, epoxy, or fluorine paints. Residual tritium was largest for epoxy paints although the permeability was smallest for those tested (Ono et al. 1991).
- About 10% (range 1% to 21%) of surface tritium activity on 304 and 316 stainless steel can be removed by means of filter paper swipes (Surette and McElroy 1988).

**Table 2.8.** Permeability Constants for Selected Glasses, Extrapolated to 300 K

Glass Type Permeated	cm <sup>3</sup> /mm	Ci/mm	Data Source
	s cm <sup>3</sup> cm Hg	s cm <sup>3</sup> cm Hg	
Silica glass	1.8·10 <sup>-13</sup>	1.6·10 <sup>-13</sup>	Souers (1986)
Pyrex glass	6.0·10 <sup>-15</sup>	5.2·10 <sup>-15</sup>	Souers (1986)
M-15 glass	8.9·10 <sup>-17</sup>	7.8·10 <sup>-17</sup>	Souers (1986)
M-30 glass	8.9·10 <sup>-20</sup>	7.8·10 <sup>-20</sup>	Souers (1986)
Soda lime	2.8·10 <sup>-18</sup>	7.2·10 <sup>-18</sup>	Neimeyer (1969)

- Approximately all surface activity (90%) on stainless steel and other metal planchets was in the form of tritiated water even though the planchets were exposed to (almost) pure T<sub>2</sub> gas and not to tritiated water (Surette and McElroy 1988; Antoniazzi et al. 1991; Dickson and Miller 1991).
- Tritium is sorbed to a greater degree by nonmetallic surfaces (except glass) than metallic surfaces but conversion of HT to HTO is less for nonmetallic surfaces than metallic surfaces. Most samples sorbed more HTO than HT although the HT concentration was larger. HTO is more readily absorbed than HT and is more difficult to desorb than HT (Dickson and Miller 1991).

## 2.6 Chemical Properties of Tritium

A range of the chemical properties of tritium are covered in this section, including decay, oxidation, and radiolysis, as well as environmental properties and reactions with organic materials.

### 2.6.1 Decay Chemistry

Pure tritium is a diatomic gas, whereas helium is a monatomic gas; the decay of 1 mol of tritium gas ultimately yields 2 mol of helium:



Pressure builds up as the tritium decays in sealed containers, such as the sealed glass containers used for current RL devices that contain diatomic tritium gas.

### 2.6.2 Oxidation and Radiolysis

Tritium reacts with oxidants as ordinary hydrogen does. The flammable/explosive limit for hydrogen

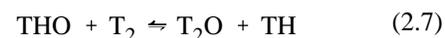
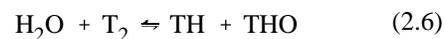
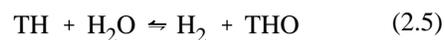
in air ranges from 4% to 75% by volume (Olishifski and McElroy 1971). These air concentrations equate to 0.0948 to 1.778 Ci/cm<sup>3</sup> air for T<sub>2</sub> and 0.0474 to 0.889 Ci/cm<sup>3</sup> for TH (at 25°C). The flammability or potential explosiveness of tritium may be an important environmental consideration, depending on the quantities of tritium used or stored. When released to the atmosphere, T<sub>2</sub> reacts spontaneously but slowly with other isotopes of hydrogen and water to form TH, DT, THO, and DTO.

Tritiated water (T<sub>2</sub>O) in concentrations above 2.0% and other tritiated compounds self-radiolize, that is, they decompose (Whittemore and Lehman 1957, Evans 1974). For example, T<sub>2</sub>O in its own radiolytic field forms T<sub>2</sub> and O<sub>2</sub> and can produce pressures from 20- to 600-cm Hg (Evans 1974).

Because tritium oxide and other tritiated compounds auto-decompose, any direct use of T<sub>2</sub>O or tritiated compounds used for RL light technology development will need to be carefully evaluated. For example, absorption of T<sub>2</sub>O into zeolites, metal tritides, or polymers used as RL light sources need to be carefully evaluated. For T<sub>2</sub>O, total decomposition depends upon the equilibrium pressure in the containment vessel and upon the available catalytic materials (Evans 1974).

### 2.6.3 Environmental Chemistry

THO can be formed in the atmosphere by isotopic exchange via the following reactions:





Similar reactions are possible in the presence of deuterium and tritium but both of these hydrogen isotopes are in small concentrations in the environment. Gas-liquid and gas-phase equilibrium constants have been calculated for the above reactions and favor the formation of TH and THO. Reactions (2.4) and (2.6) are of the greatest concern when a "gas tube" RL unit is destroyed and the contained tritium (pure T<sub>2</sub>) is released. When tritium is released indoors, catalytic surfaces and other factors affect the reaction processes.

Based on a survey of laboratories and reactors by Robins, Bartoszek, and Woodall (1984), later experimental studies by Blet et al. (1991) and Dickson and Miller (1991), and the results of reported decontaminations of stainless steel and tritium facilities by Anatoniazzi et al. (1991), Christ and Wehner (1991), and Maienschein et al. (1991), a few generalizations about the conversion to TXO may be made:

- In laboratory facilities or reactor halls, about 50% of the TX released (where X is either H or T, but normally H) can be expected to convert to TXO before leaving the exhaust stack. Even changing the air in glove boxes once every few minutes does not prevent substantial conversion.
- At least a few percent of the TX released would be converted to TXO each day, even under carefully controlled conditions.
- Rapid conversion from TX to TXO takes place on surfaces within tritium facilities, a fact which is consistent with slower results obtained experimentally using glass bulbs. Factors probably increasing the observed conversion rates in tritium facilities are temperature, humidity, gas turbulence in ductwork, and contact with catalytic surfaces. For example, temperatures as high as 400°C have been reported where contact

with fan blades, motors, and hot reactor piping is common. Humidity also accelerates tritium conversion by one or more orders of magnitude.

## 2.7 Reactions in Gas-Filled RL Light Tubes

A number of chemical reactions may occur between the phosphor coating and its binder on the inside surface of an RL light tube and the contained tritium. Residual water and residual chemicals also appear to react within the tubes. For the most part, these reactions are not well defined but data are available from studies conducted at Mound Laboratories that demonstrate that such reactions may occur (private communication, R. E. Ellefson, Mound, to G. A. Jensen, Pacific Northwest Laboratory).

Reactions in four types of gas-filled light tubes will be discussed below. The first type, which contains the least tritium, consists of a small device with less than 1 Ci. These devices are used for backlighting instrument dials (e.g., reticules in gun sights) and have been used for illumination of liquid crystal displays on watches. The second type is the 1- to 2-Ci devices used for backlighting scales on inclinometers for mortars and howitzers and to make personnel markers for the military. Some manufacturers spell "EXIT" with 1- to 2-Ci segments. The third type is the 5- to 10-Ci tubes used as lighting segments of aircraft and commercial exit signs and for aiming post devices and map reading lights for the military. The fourth type is the 30- to 166-Ci tubes used for airfield runway lighting, taxiway signage panels, and signal wands. Recovery and disposal options for RL lights have been assessed (Jensen et al. 1991).

### 2.7.1 Expected Composition of Gas from RL Tubes

Two studies of the gas composition in small (2- and 8-Ci) commercial and ORNL (50-Ci) tritium devices have been performed at Mound since 1984 (private communication, R. E. Ellefson, Mound, to G. A.

Jensen, Pacific Northwest Laboratory). The findings of each study give similar results: The tritium from direct fracture has nearly the expected helium-3 content (implying little loss by diffusion), some protium (H) ingrowth, and a small indication of water vapor present in the gas ( $T_2O$ , see Table 2.9). The sealed glass envelope is a good storage vessel for tritium gas. Residual gas adsorbed on the phosphor accounts for 7% to 20% of the total tritium and is proportional to the surface-to-volume ratio of the tube (see Tables 2.3 and 2.10). In Table 2.3 the area-to-volume ratio is 7 and in Table 2.10 the ratio is 20; in both device types, the fill pressure is 1.3 atm. These data imply that 80% to 90% of the tritium can be released by fracture alone without high-temperature processes to remove absorbed tritium.

An additional study of commercial RL exit signs was commissioned by the NRC as a result of several incidents involving damaged building exit signs (Bowerman and Czajkowski 1990). Personnel at Brookhaven National Laboratory examined RL light tubes from four of six signs forwarded from several manufacturers. The signs were dismantled, the RL light sources were smeared, and then the tubes were broken in such a fashion that the tritium gas in the tubes could be analyzed for their water content. The smears showed no contamination above background on the outside of the panels; however, the smears of the tubes showed contamination that ranged from less than 100 disintegrations/minute to over 2000 disintegrations/minute. The two new signs had tritiated water contents of 2% and 4.5% water. The sign that was 6-years-old had a water content of 14.5% and the oldest sign, 13-years-old, had a tritiated water content of 12.2%. Since the work was scoping in character, a discussion on why tritiated water was present was not included.

## 2.7.2 Reactions with Organic Materials

Organic tritium compounds have been made into luminous paint for watch dials and other RL applications. Tritium labelling in this fashion occurs on the

backbone chain in the polymeric molecule, as shown in Figure 2.1. However, these polymer chains are subject to breakage and leakage of the tritium because the polymer is destroyed as the tritium decays. With breakage of the polymer chain, the tritium becomes inert helium and outgasses as elemental helium and probably HT. This tritium release is measurable: urine samples from individuals wearing watches having luminous dials was found to contain 1.1 to 12 nCi/L of tritium. Individuals not wearing luminous dial watches had average tritium concentrations in their urine of about 0.7 nCi/L (Evans 1974).

Researchers at the Pacific Northwest Laboratory have developed a method for producing tritiated polymers in which up to five tritium atoms are exchanged for hydrogen on the aromatic pendent ring in polystyrene (Jensen et al. 1989, 1992). The backbone ethylene chain is not tritiated; thus, the polymer is expected to be stable and the tritium released is expected to be minimal.

In addition, researchers at Sandia National Laboratories have been investigating 1,4-Di-(2)-phenyl ethynyl benzene (DEB) and similar compounds as hydrogen (and tritium) getters (Shippodd and Smith 1990). To use DEB as a getter requires a palladium catalyst to cause the tritium to add to the ethylene linkage between the aromatic portions of the molecule.

However, the tritium will leak from the DEB if hydrogen or water is present. By adding LiH, the getter material (DEB/Pd on  $Al_2O_3 + LiH$ ) can be used as a getter for the released tritium or tritium in tritiated water. The resulting materials are stable and can be used to improve the safety of RL light units.

## 2.7.3 Metal Tritides

Relatively stable metal tritides such as titanium tritide ( $TiT_2$ ) have been proposed for use as RL light sources. Tritium reacts with metals to form hydrides by the reaction:

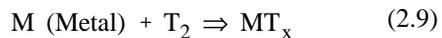
**Table 2.9.** Tritium Gas Composition for Various RL Lights (%)

	Fill Gas <sup>(a)</sup> (Glass)	Range Indicator	Aiming Post	ORNL Tube Gas <sup>(b)</sup>	ORNL Solid Bakeoff <sup>(b)</sup>
N(Ci)	6.66	0.68	4.74	36.57	3.41
H(Mol%) <sup>(c)</sup>	0.66	6.96	5.14	2.20	27.60
D <sup>(c)</sup>	0.87	0.90	1.02	0.84	0.77
T <sup>(c)</sup>	92.76	83.25	92.52	69.97	60.41
He-3	5.56	7.08	2.24	26.72	3.56
H <sub>2</sub>	0.02	0.46	0.36	0.00	14.50
HD	>0.05	0.15	<0.05	<0.05	0.29
HT	1.28	12.84	9.56	4.37	25.92
D <sub>2</sub>	<0.02	<0.02	<0.02	<0.02	<0.02
DT	1.73	1.64	2.04	1.65	1.23
T <sub>2</sub>	91.26	76.02	85.57	66.05	46.84
22-T <sub>2</sub> O	0.01	0.31	0.03	0.20	0.55
24-CT <sub>4</sub>	0.15	1.47	0.20	0.38	0.01
28-N <sub>2</sub>	<0.01	0.02	<0.01	0.10	5.65
32-O <sub>2</sub>	<0.01	<0.01	<0.01	<0.01	0.03
40-Ar	<0.01	0.01	<0.01	0.08	0.23
44-CO <sub>2</sub>	<1.0	<0.01	<0.01	0.05	1.57

(a) Fill gas sealed and stored in a spherical glass envelope by a vendor.

(b) Two analyses of same ORNL tube: one as free gas and one as gas absorbed on solid.

(c) Total of H, D, and T represent elemental hydrogen percentage.



where x depends upon the valence state of the metal and how much hydrogen the metal will absorb. The chemistry of tritides is essentially the same as that for hydrides, and they can be handled similarly. For example, titanium tritide powder (or thin films of titanium), when reacted with tritium, produces titanium tritide (TiT<sub>2</sub>). The films (or powder) could be used as source materials for further fabrication into RL light units. Unfortunately, titanium hydride (tritide) and many other hydrides (for example, Zr,

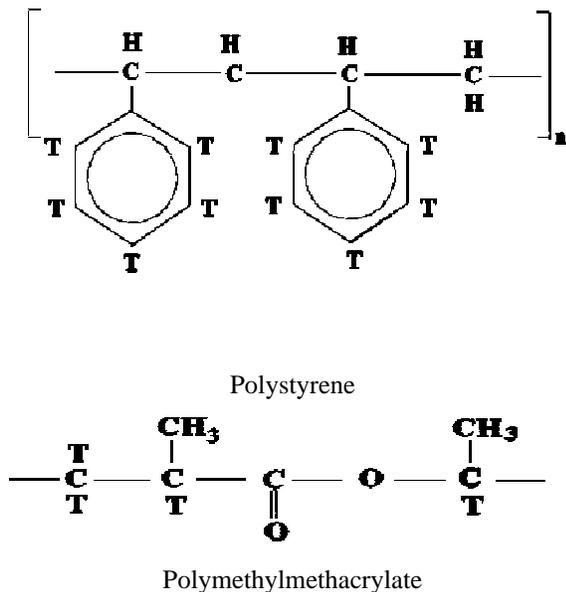
Hf, and Y tritides) are pyrophoric, that is, they ignite in air. For titanium tritide, the compound is pyrophoric at temperatures above 350°C (Muller et al. 1968) and, when finely divided, near room temperature. Particle size, purity, porosity, hydrogen content, and water vapor all affect pyrophoricity and ignition temperature. Rare earth tritides (hydrides) are reported to be more stable (Muller et al. 1968) but may be less useful because of the lower specific activity of the tritium contained in them. Care will

**Table 2.10.** Tritium Content of ORNL 10 in. x 1/2 in. Tubes (Gas released during room temperature fracture and additional gas released during a 500°C bakeoff of the tube.)

Tube No.	Gas, $n_g$ ( $\text{cm}^3$ (STP))	Tritium, $T_g$ (Mol%)	Solid, $n_s$ ( $\text{cm}^3$ (STP))	Tritium, $T_s$ (Mol%)	Tritium, $T_{\text{sol}}^{(a)}$ (Mol%)
1	15.93	68.55			
2	15.18	67.81	1.74	52.76	8.2
3	15.76	69.44	1.90	55.35	8.8
4	13.04	65.98	1.65	56.60	9.8
5	17.42	70.70	1.98	56.32	8.3
6	19.50	69.67	2.11	60.41	8.6
7	18.93	71.40	1.89	58.95	7.6
8	18.22	74.73	2.02	62.17	8.4
9	19.43	70.60	1.81	61.87	7.5
10	19.39	71.29	1.91	63.49	8.1
11	16.43	68.16	2.36	55.17	10.4
12	15.04	68.90	2.23	49.90	9.7
13	12.56	69.56	1.79	53.08	9.6
14	13.27	68.72	2.09	47.38	9.8
15	18.70	69.18	2.32	49.25	8.1
16	17.22	66.98	1.69	56.69	7.7
17	18.34	72.41	1.89	40.71	5.5
18	18.01	72.98	1.52	50.22	5.5
19	15.44	66.36	1.90	57.82	7.7
20	14.21	67.20	1.82	48.43	8.4
21	13.01	67.63	2.06	47.84	10.1
22	14.29	67.47	1.59	59.78	9.0
23	18.10	65.29	2.65	51.88	10.4
24	13.77	69.21	2.15	45.17	9.3
25	18.84	71.46	1.30	44.46	4.1
26	20.07	71.16	1.98	43.28	5.7
27	18.01	71.49	1.23	41.03	3.8
28	18.98	68.38	1.71	65.15	7.9
Average	16.68	69.39	1.90	53.15	8.1
Std. Dev.	2.3	2.19	0.30	6.84	1.8

(a) Calculated as follows:

$$T_{\text{sol}}(\%) = \frac{100T_s(\text{mol}\%)\cdot n_s(\text{cm}^3)(\text{@STP})}{T_s \cdot n_s + T_g \cdot n_g}$$



**Figure 2.1.** Tritium in Polymeric Molecules

have to be taken to ensure that the tritides are properly encapsulated to avoid spontaneous ignition and that they will survive reasonable temperatures in a fire.

## 2.8 References

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### **3.0 Tritium in the Environment**

## 3.0 Tritium in the Environment

This section summarizes information on levels and sources of tritium in the environment, including natural tritium, major man-made sources, and the location of tritium in the biosphere.

### 3.1 Concentration of Tritium in the Environment

Different tritium concentrations are found in the atmosphere, groundwater, and seawater at different latitudes, and over land areas or the oceans (NCRP 1979). Higher concentrations of tritium are found in the mid-latitudes than in the Arctic latitudes. This distribution parallels the tritium release from all sources since the advent of nuclear weapons testing and nuclear power. Tritium concentrations in the biosphere are largest in the northern hemisphere because most of the weapons testing and nuclear power development have occurred there.

The tritium concentration in rainfall is normally higher over continental areas than over oceans. Oceans and other large water bodies act as sinks for tritium because of the rapid mixing that occurs down to about 75 m. Thus, water evaporated from oceans or large bodies of fresh water will have a lower tritium concentration than that found in the original rainfall (NCRP 1979).

### 3.2 Natural Tritium

About 4 MCi/y of tritium is formed naturally by cosmic-ray bombardment of nitrogen and oxygen in the upper atmosphere (Moghissi and Carter 1973). Additional tritium may be introduced into the atmosphere by solar flares. The world inventory of natural tritium in the biosphere is estimated at 69 MCi. Natural (and man-made) tritium reaches the earth's surface chiefly by precipitation and distributes roughly 90% into the hydrosphere and 10% into the atmosphere. Natural tritium concentrations in the hydrosphere ranged from  $1.61 \cdot 10^{-9}$   $\mu\text{Ci/mL}$  to  $16.1 \cdot 10^{-9}$   $\mu\text{Ci/mL}$  before the beginning of thermonuclear weapon testing (NCRP 1979).

#### 3.2.1 Tritium from Weapons Testing

Weapons testing has increased the amount of tritium in the environment. Estimates for the amount of tritium released from weapons testing have ranged from 25 to 150 times the tritium inventory produced naturally until the weapons test ban treaty went into effect in 1963 (NCRP 1979; Moghissi and Carter 1973; Brown and Workman 1986). Most of this tritium was injected into the stratosphere, where it remained for some years because of its slow rate of mixing with the troposphere. Once in the troposphere, tritium is washed rapidly into surface waters. The half-life of tritium in the troposphere is on the order of  $35 \pm 5$  days, and concentrations of tritium in rainwater following bomb tests were several hundred times the concentrations produced by natural processes. Decay of the tritium produced by bomb testing is expected to return the content of tritium in the environment to its steady-state natural inventory of 69 MCi by 2030 (NCRP 1979). Tritium concentrations in streams reached concentrations approaching  $1.4 \cdot 10^{-6}$   $\mu\text{Ci/mL}$  in 1963 and are now near  $1 \cdot 10^{-7}$   $\mu\text{Ci/mL}$  (NCRP 1979).

### 3.3 Tritium from U.S. Department of Energy and Other Weapons Production Facilities

In 1991, the Assistant Secretary for Environment, Safety, and Health (EH) commissioned an internal DOE Task Group to "review tritium facility management practices and identify measures to improve operations." Tritium releases for the years 1986-1990, inclusive from DOE tritium facilities, were identified.

The cumulative tritium release was reported to be 186.6 grams or 1,795,000 Ci. Approximately 90% of this tritium was released at the Savannah River Site (SRS) (DOE 1991). The tritium releases from the SRS K-Reactor were approximately 238 grams or about  $2.3 \cdot 10^6$  Ci from the start of operations through 1985. Emissions from K-reactor represent approximately 35% of the emissions from SRS during the study period. Using these percentages as an approximation, about 960 grams of tritium have been released at DOE sites during the last 35-40 years.

Data for production of nuclear weapons in the former U.S.S.R. and other countries is not available. However, from available information, the U.S.S.R. produced close to the same number of weapons that were produced in the U.S. Therefore, assuming the tritium release from weapons production from the U.S.S.R. and others would at least equal that released by the U.S. world-wide releases would have approached 2000 grams (19.4 MCi) or more over the past 35-40 years. This equates to approximately 30% of the natural steady-state tritium inventory.

### 3.4 Tritium from Nuclear Power Plant Emissions

Tritium is formed also in nuclear reactors (boiling water reactors [BWRs] and pressurized water reactors [PWRs] by ternary fission, and in PWRs, BWRs, and CANDU reactors by light-element activation). Most of the tritium produced in PWRs and BWRs is captured as zirconium tritide in the zirconium alloy cladding used for encapsulating the fuel. Projections of release and accumulation of tritium from nuclear power production and other nonnatural sources are included in Table 3.1. These estimates include BWRs and PWRs for the years 1990-2000.

Production of tritium in pressurized heavy water reactors (PHWRs) in Canada is estimated to approach 26 MCi/y, and nearly all this will be recovered for disposal or sale (Drolet et al. 1984). Releases of tritium from PHWRs usually occur when tritium is released from heavy water ( $D_2O$ ) escaping to the atmosphere. Drolet (1984) reported that about 0.5 kg/h  $D_2O$  escapes and contains about 65 Ci/kg of tritium. Concentrations of tritium in the heavy water will be reduced to ~5 Ci/L once the planned tritium recovery for disposal or sale is underway.

Fast breeder reactors (FBR), advanced gas-cooled reactors (AGRs), and gas-cooled reactors (GCRs) all produce tritium by nuclear fission. Most of the fuel used in them is either plutonium or mixed uranium-plutonium fuel, and more tritium is produced per megawatt electrical than in PWRs or BWRs. Nearly all of it is captured in the liquid metal coolant or the materials used to encapsulate the fuel. At present, the total tritium released to the environment is small (less than 1 kCi/y). Data for light-water graphite reactors (LWGRs) and high-temperature graphite reactors (HTGRs) are not available, but assuming that tritium release from these sources is on the order of magnitude of PWRs and BWRs, about 2 kCi/y from these sources is anticipated.

**Table 3.1.** Projected Release and Accumulation of Tritium in the Environment from Nuclear Power Production and Other Nonnatural Sources (MCi)

Year	Nuclear Power	Weapons Tests	Other	Total
1990	4.43	665	1.7	671
1991	4.83	630	1.7	637
1992	5.03	595	1.7	602
1993	5.23	560	1.7	567
1994	4.50	530	1.7	536
1995	5.73	500	1.7	507
1996	5.93	475	1.7	483
1997	6.13	450	1.7	458
1998	6.43	425	1.7	433
1999	6.53	400	1.7	508
2000	6.53	380	1.7	388

Source: NCRP 1979.

### 3.5 Tritium Distribution Cycles

Considering an inventory of natural worldwide tritium of 69 MCi, man-made tritium constitutes about 88% of the tritium currently in the environment, and man is adding about 12% to 14% annually to the amount found naturally. Tritium concentrations vary according to latitude and type of area, i.e., whether tritium is in the atmosphere, groundwater, or seawater (NCRP 1979). Table 3.2 shows average concentrations for a 1-Ci/y release over land masses and oceans, and divides releases by latitude.

Data available on environmental concentrations in the southern hemisphere are minimal, but they are expected to be nearer those occurring naturally.

Both the high rainfall over ocean areas and the vapor exchange over oceans result in greater atmospheric removal of tritium than over land. Tritiated water

(THO) has a lower vapor pressure and hence would be expected to move more readily into the water phase than into the vapor phase if the tritium content of the vapor phase is higher. Related to this effect, the diurnal effects for tritium transfer have also been observed (see Moghissi 1975). Overnight, the tritium in stagnant air over a lake was reduced from 157 tritium units to 68 tritium units (a tritium unit, T.U., is 1 tritium atom in  $10^{18}$  hydrogen atoms [Evans 1974]), while the moisture content in the air increased from  $7.1 \text{ g/m}^3$  to  $10.4 \text{ g/m}^3$ .

Tritium reaching the ground can be recycled to the atmosphere by re-evaporation or by transpiration through plants, or can be carried away by surface runoff. Slightly over one-third (37%) of all rainfall over continental areas is absorbed into the soil or is removed in runoff (Rankama and Sahama 1949). Tritium absorbed into the ground is essentially lost from circulation except where it reappears in springwater. Because rainfall over continental areas is highly variable, tritium concentrations will vary

**Table 3.2.** Average Ground Level, Atmospheric, and Surface Water Concentrations, Assuming 1-Ci/y Atmospheric Release in Latitude 30° to 50°N<sup>(a)</sup>

Latitude (°N)	Land Masses <sup>(b)</sup>		Oceans <sup>(c)</sup>	
	Air (pCi/m <sup>3</sup> · 10 <sup>8</sup> )	Water (pCi/L · 10 <sup>7</sup> )	Air (pCi/m <sup>3</sup> · 10 <sup>8</sup> )	Water (pCi/L · 10 <sup>7</sup> )
0 - 10	4.0	0	0.13	0
10 - 20	4.2	171.0	0.3	0.15
20 - 30	14.0	224.0	2.8	2.0
30 - 50	35.0	287.0	15.3	25.8
50 - 60	15.0	54.0	3.8	0.5
60 - 70	5.9	13.0	0.6	4.9 · 10 <sup>-3</sup>
70 - 80	1.9	3.3	0.1	3.4 · 10 <sup>-5</sup>
80 - 90	0.8	0	0.03	0

(a) Table from NCRP (1979).

(b) Assumed depth = 0.5 m.

(c) Assumed depth = 75 m.

depending on latitude, or climatological factors, or whether man-made or natural sources are present. Man-made tritium emitted as a plume from a stack or other means will eventually reach ground surface, and on the order of 1% of the tritium released as T<sub>2</sub> or TH will be converted to THO in the soil (Djerassi and Lesigne 1988). The remainder will eventually be washed from the atmosphere in rainfall or will decay in the atmosphere (NCRP 1979).

### 3.6 Organically Bound Tritium

Man-made tritium released to the environment (and natural tritium) is usually absorbed through the lungs or the skin. There is another pathway that must be considered where there is either a continuous or episodic release of tritium. In these cases, tritium released is incorporated into living plant or animal tissue as organically bound tritium (OBT). Thus, OBT may represent a longer-term exposure to organisms living near the release site than the tritium exposure from the tritium released to the environment

in its water or elemental forms (Murphy 1993; Diabaté and Strack 1993).

Tritium and the other isotopes of hydrogen cycle in the environment similarly. Although physical and chemical isotopic effects exist and are found in some transport mechanisms and chemical processes, they are not significant relative to the creation of OBT. Thus, there is no concentration of tritium by organisms in the foodchain other than that created by the usual transport process for hydrogen (Murphy 1993).

The absorption of tritium by plants and algae during daylight hours when photosynthesis occurs is the most important mechanism for collecting tritium as OBT (Diabaté and Strack 1993). Tritium absorbed in this manner reacts into the chemical structure of the leaf by photosynthesis and enzyme processes. The chemical components formed are transported to other parts of the plant where they are deposited in other edible parts of the plant. Most of the tritium

absorbed is liable and does not exchange with the C-H bonds of the organic structure. This tritium ultimately returns to the water pool in the organism and may be excreted. Where the tritium is exchanged with the hydrogen in the C-H bond, this represents a potential long-term exposure (Diabaté and Strack 1993). The tritium available for incorporation into plants is largely dependent upon the tritium concentration of the tritium-containing water absorbed and other water entering the organism. Thus, the organic concentration of tritium will be lower than the concentration in the organism's water pool.

A chronic or acute short-term environmental release of tritium to the atmosphere will affect biota including humans only if the tritium reaches the surface as rain or is directly absorbed. In humans, tritium is absorbed through the lungs or skin, where it can be converted to OBT by enzymatic processes in the body or is present in the food consumed. The rainwater containing released tritium must reach the water available to the plants or animals consumed. Although OBT that results is small, it represents a higher concentration of tritium in a pathway compartment to be addressed when such releases occur (Murphy 1993).

### 3.7 Environmental Effects of RL Devices

Tritium released from RL lights should have the same fate as manmade tritium from any other source released to the environment. Over land areas, the tritium concentrations vary in the atmosphere and in surface waters. Thus, care must be taken to ensure that the local tritium concentration in surface waters, groundwaters, and plants is known so that the results of a release from an accident involving RL light units can be properly evaluated.

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## **4.0 Emergency Preparedness for Tritium RL Devices**

## 4.0 Emergency Preparedness for Tritium RL Devices

During use, RL devices may be broken or damaged. When this occurs, tritium will be released, appropriate safety concerns will need to be addressed, any necessary cleanup of the tritium will need to be initiated, and the damaged device will need to be recovered for disposal. The following sections discuss what steps will need to be taken in handling accidents.

### 4.1 Procedures in Dealing with a Broken RL Light

The tritium released immediately after a light breaks will most likely be elemental hydrogen; in that case, the greatest dose will be to the lung. However, the very small uptake will make it difficult to quantify the dose to the lung. As it may be some time before the tritiated water associated with the light volatilizes (Niemeyer 1969), total exposure to tritium can be minimized by placing the light in a shipping container as soon as the damage is noticed.

Protective clothing is generally not required for individuals who deal with RL devices. If a device is broken, however, precautions should be taken to reduce the possible radiation dose to any individual responsible for cleanup. The protective clothing required for this will probably be limited to gloves. Persons who handle a broken device should wear gloves and wash their hands immediately after handling it.

### 4.2 Health Physics Kit

Although the equipment may never be needed, an emergency kit is recommended for use by a designated radiation protection technologist (RPT) for use in case of breakage of RL devices:

- filter papers for taking smears
- ethylene glycol or glycerol (to moisten filters)
- liquid scintillation counting (LSC) vials
- LSC cocktail
- urine collection bottles (to contain at least 100 mL) with labels
- a tight container for broken/damaged units (aluminum containers are relatively impermeable to hydrogen gas [Gause et al. 1984])
- gloves for handling broken/damaged units
- silica gel (place in container with broken units to collect tritiated water)
- incident report forms.

### 4.3 Monitoring Persons Exposed to Tritium from Damaged RL Devices

The individual(s) in a room where an RL device is damaged should be monitored for exposure to tritium. In case of an accident outdoors, individuals near the damaged device, especially those downwind, should be monitored for tritium exposure. Whoever handles the damaged light should also be monitored.

### 4.4 Decontamination of Broken RL Lights

Because tritium in RL devices is predominantly in the form of elemental gas, smears of RL devices will yield little information unless the device has been broken. When tritium-containing RL lights are broken, tritiated water and possibly phosphors may be found on the surface of the devices.

Decontamination of surfaces can be difficult because tritium permeates into materials. Areas thought to have been decontaminated can appear to be recontaminated because tritium that had diffused into the surface is now diffusing out from it. When the tritium leaves the surface, the process is said to be "bleeding out" or "outgassing." For any tritium removal process to be truly effective, all subsurface tritium activity as well as surface activity must be removed. Residual phosphor on the surfaces of damaged RL devices can also be misleading concerning the effectiveness of the cleanup operation. This is because phosphor absorbs light energy and can cause errors in the analysis of the surface contamination.

At times, it may be necessary to obtain smear samples to determine whether a light source is leaking or, if the light's containment has been broken, to determine the extent of the contamination. Details for the procedure are discussed in Section 4.8.

### 4.5 Training

The training requirements for users of RL devices is minimal. However, a trained custodian for the lights should be identified who is responsible for proper disposal of the devices if they are broken. This individual should be trained in emergency procedures, including packaging a broken device, collecting bioassay samples, and surveying the potentially contaminated area.

### 4.6 Area Monitoring

Area monitoring can indicate the exposure of workers in normal working conditions and warn when locally derived working limits are exceeded (IAEA 1980, para. 4.4.1).

The methods usually employed in area monitoring are (IAEA 1980, para. 4.4.2.1):

- measurement of external radiations
- sampling of airborne contaminations

- survey of surface contamination.

In the case of tritium, there is no easily measurable external radiation field produced.

Air sampling is needed to help quantify the activity inhaled by the individual. The basic assumption is that the air concentrations that are inhaled by the radiation worker are represented by the measured air concentration (IAEA 1980, para. 4.4.2.4). However, because concentration gradients can, and often do, exist, it is desirable to sample the air as close as possible to the worker's breathing zone.

Surface contamination can be determined directly by means of survey instruments or indirectly by means of smear techniques (IAEA 1980, para. 4.4.2.7). The range of the tritium beta particle in air, only about 0.6 cm, makes detection of tritium surface contamination by instruments very difficult; smear techniques are generally used. Because tritium in RL devices is predominantly in the form of elemental gas, smears of RL devices will yield little information unless the device has been broken. When RL devices are broken, tritiated water may be found on the surface of the devices so that smearing the damaged devices will yield information on the surface contamination.

## 4.7 Interactions with Surfaces

The mechanisms of tritium interactions with surfaces are detailed in Section 7.0. Thus, in this chapter it is necessary only to be aware that tritium can, and does, adhere to surfaces as tritiated water or elemental tritium (Easterly 1982). Tritium physically permeates into a surface, diffuses through the material, and may eventually leave from the other side. To minimize surface contamination, hydrogen-containing materials should be avoided, especially where high concentrations of tritium may be possible.

Braun and Williamson (1985) found that coatings with a low sheen, particularly those that are epoxy-based, can be decontaminated more readily than those with a high gloss. This occurs because tritiated water is desorbed faster from the more open film structure associated with coatings that have a high pigment-to-binder ratio.

## 4.8 Survey Procedures

At times, it may be necessary to obtain samples to determine whether a light source is leaking or, if the light's integrity has been compromised, to determine the extent of the contamination. Surveys can be made either by sampling the air or by smearing the surface of some object. (Air-sampling methods are discussed in Section 7.0.)

Swipes or smears are often used to determine how much radioactivity is on a surface. The technique is to wipe the surface of the object with an absorbent material (cotton swab, filter paper, or commercially available smears). The surface area to be swiped should be about 100 cm<sup>2</sup>. The activity on the smear is then quantified to determine the extent of "removable" activity on the surface. Gas-flow proportional counters may be used to count the smears. Liquid scintillation counting (LSC) is more convenient and may be more accurate. When counting by LSC, the sample to be counted should be left in the counting chamber in the dark for at least 24 hours and recounted to be sure that the absorption of light by any phosphor contaminating the smear has been accounted for. Although the activity on the surface of the object may be quantified and corrections made to account for changes in counting efficiency and efficiency in extracting the tritium from the absorbent material, often this is not done.

To increase the fraction of the surface activity that is removed by the swipe and to prevent evaporation of water, the swipes should be wetted by glycerol or ethylene glycol before taking the swipe. Some authors consider ethylene glycol to be better than glycerol. Table 4.1 shows NRC criteria for surface surveys.

## 4.9 An Emergency Kit

Although the equipment may never be needed, the kit described in Table 4.2 is recommended for use by the designated radiation protection technologist (RPT).

## 4.10 Protective Clothing

Protective clothing is generally not required for individuals who deal with RL devices. If a device is broken, however, precautions should be taken to reduce the possible radiation dose to any individual responsible for cleanup. The protective clothing required for this will be limited to gloves. Several kinds of gloves are available through chemical and medical supply houses. The type of glove necessary

**Table 4.1.** U.S. Nuclear Regulatory Commission Criteria for Tritium

Acceptable Frequencies for Surveys <sup>(a)</sup>	Air Sampling			Surface Contamination		
	Weekly	Monthly	Quarterly	Weekly	Monthly	Quarterly
Amount (Ci) in process at any one time or placed into process in any 3-mo period within any room requiring surveys	≥10	1 ≤ x <10	1	≥100	10 ≤ x <100	<10

Recommended Action Levels <sup>(b)</sup>	Type of Surface	Level (μCi/m <sup>2</sup> )
Removable Surface Contamination in Manufacturing Plants	Unrestricted areas	1 · 10 <sup>-6</sup>
	Restricted areas	1 · 10 <sup>-2</sup>
	Personal clothing	1 · 10 <sup>-2</sup>
	Protective clothing	1 · 10 <sup>-4</sup>

Acceptable Surface Contamination <sup>(b)</sup>	Average	Maximum	Removable
Levels (for release of materials)	5,000 dpm/100 cm <sup>2</sup>	15,000 dpm/100 cm <sup>2</sup>	1,000 dpm/100 cm <sup>2</sup>

(a) From Regulatory Guide 8.21 (NRC 1979).  
 (b) From Regulatory Guide 1.85 (NRC 1989).

depends on the work to be performed or whether perforation resistance is necessary or not. (See Table 4.3 for compatibility of gloves with various chemicals). Hafner has prepared a table of commercially available gloves (see Table 4.4). For the column "THO Permeation Characteristics," he did not quantify the differences between poor, fair, good, and excellent. The literature seems to indicate that whatever kind of glove is used, water permeates through it within 10 minutes to 1 hour, depending on the glove thickness.

The effect of different types of clothing on the absorption of tritium is shown in Table 4.5. Protection values for different types of clothing as determined by Osborne (1966) are shown in Table 4.6.

## 4.11 Emergency Actions

If a device is broken:

- Identify the affected individuals and write down their names.
- Get the health physics kit.
- Put on the rubber gloves.
- Place the broken device in a "broken device box" and sprinkle the device with silica gel.
- Seal the "broken device box."

**Table 4.2.** Emergency Kit Contents

Item	Number
Filter papers for taking smears	50
Ethylene glycol or glycerol to moisten filters	100 mL
LSC vials	20 vials
LSC cocktail	500 mL
Urine collection bottles (to contain at least 100 mL) with labels	10
A sufficiently large, tight container for broken/damaged units and residue from the immediate area <sup>(a)</sup>	2
Gloves for handling broken/damaged units	20 pair
Silica gel (place in container with broken units to collect tritiated water)	500 gm
Incident report forms or notebook	1

(a) Aluminum containers are relatively impermeable to hydrogen gas (Gause et al. 1984).

**Table 4.3.** Chemical Compatibility of Available Liquid-Proof Glove Materials<sup>(a)</sup>

Material	Recommended for	Not Recommended for
Butyl	Dilute acids and alkalis, ketonic solvents, gas and vapor permeation protection	Petroleum oils, distillates, and solvents
Natural Rubber	Ketonic solvents, alcohols, photographic solutions	Petroleum oils, distillates, and solvents
Neoprene	Concentrated nonoxidizing acids and concentrated alkalis	Halogenated or ketonic solvents
Neoprene/Natural Rubber Blends	Dilute acids and alkalis, detergents, and photographic solutions	Halogenated or ketonic solvents
Nitrile	Petroleum-based solvents, distillates, and oils	Halogenated or ketonic solvents
PVC	General purpose, low-risk hand protection	Halogenated or ketonic solvents
PVA	Halogenated solvents, paint shop applications	Water or water-based solutions
Viton	Halogenated solvents, concentrated oxidizing acids	Aldehydes, ketonic solvents

(a) Table prepared by R. S. Hafner of Lawrence Livermore National Laboratory (LLNL). It is based on glove manufacturers' data sheets. Used with permission of R. S. Hafner (Teleconference May 4, 1988).

**Table 4.4.** Physical Properties of Commercially Available Gloves<sup>(a)</sup>

Glove Material	Length (in.)	Thickness (in.)	Shelf Life	Relative Toughness	THO Permeation Characteristics
PVC	11	0.005	Fair	Fair	Poor
PVC	11	0.010	Good	Good	Good
PVC	11	0.020	Excellent	Excellent	Good
Neoprene/natural rubber blend	14	0.020	Good	Good	Good
Neoprene	11	0.015	Excellent	Good	Good
Neoprene	18	0.022	Excellent	Good	Good
Natural rubber	11	0.015	Poor	Fair	Good
Nitrile	13	0.015	Excellent	Excellent	Good
Nitrile	18	0.012	Excellent	Excellent	Good
Butyl	11	0.012	Excellent	Poor	Excellent
PVA <sup>(b)</sup>	12	0.022	Good	Excellent	Poor
Viton	11	0.012	Excellent	Excellent	Excellent

(a) Table prepared by R. S. Hafner of LLNL. It is based on the glove manufacturers' data sheets. Used with permission of R. S. Hafner (teleconference, May 4, 1988).

(b) As a coated, flock-lined fabric, the thickness of PVA gloves can vary by as much as  $\pm 20\%$ . Because the PVA coating is water-soluble, other properties of PVA gloves can also be expected to vary, depending on their long-term exposure to moisture.

**Table 4.5.** Transpercutaneous Intake Rates<sup>(a)</sup>

Protection Level	Intake Rate (range in $\mu\text{Ci}/\text{min}$ per $\mu\text{Ci}/\text{L}$ )
No protection	6.6 - 13.4
Oronasal mask plus cotton overalls	5.4 - 13.0
4-mil PVC	0.6 - 1.0
4 mil PVC and cotton	0.09 - 0.19
6-mil 2-piece PVC and cotton	0.2 - 0.5
Overalls between two 4-mil PVC	0.01 - 0.04

(a) From data in Osborne (1966).

**Table 4.6.** Clothing Protection Values<sup>(a)</sup>

Protection	Protection Factor	
Air-supplied mask plus 1 piece 4-mil PVC suit	20/36	16/15
Air-supplied mask plus overalls plus 2-piece, 6-mil PVC suit	40/120	40/30
Air-supplied mask plus overalls plus 1-piece, 4-mil PVC suit	120/140/160	15/15/16
Air-supplied mask plus multilayer system: dry (two 4-mil PVC suits)	>480/>640	--
Air-supplied mask plus multilayer system: wet ~50 mg/cm <sup>2</sup> (wet cotton overalls between two, 4-mil PVC suits)	>1900	12,000

(a) Table 8 of Osborne (1966).

- Have one or more individuals submit a bio-assay sample within 24 hours of handling a broken light. The sample should be obtained at least 6 hours after the accident.
- Remove the gloves (toss in contaminated waste).
- Wash hands.
- Smear the surrounding area with (several) moistened smears. Place the smears in individual LSC counting vials. Uniquely mark the cap of each LSC vial.

## 4.12 Contamination Levels

The draft standard "Surface Radioactivity Guides for Materials, Equipment, and Facilities to be Released for Uncontrolled Use" describes the criteria for surface contamination levels. The criteria are based on direct ingestion of material, ingestion of material in water, and inhalation. The published draft does not consider radionuclides that emit beta particles having an energy less than 150 keV, so tritium is not included in the standard. However, the rationale of the standard may be employed for tritium. Based on discussions in the appendix to the draft standard, it appears that total surface contamination of tritium of up to 3  $\mu\text{Ci}/100\text{ cm}^2$  would be acceptable from a dosimetric standpoint.

The standard also considers removable contamination, which levels are lower than the total contamination level by a factor of 5 to 25, depending on the radionuclide. Hafner indicates that for tritium a smear may remove only about 10% of the surface activity. Thus, we can think of acceptable levels of removable tritium contamination on the order of  $24 \cdot 10^3\text{ dpm}/100\text{ cm}^2$ . If we use a multiplier of 5 as the difference between the average contamination level and maximum removable contamination level (in accordance with Regulatory Guide 1.86 [NRC 1974]), we have an average contamination level of  $120 \cdot 10^3\text{ dpm}/100\text{ cm}^2$  and a maximum of about  $360 \cdot 10^3\text{ dpm}/100\text{ cm}^2$ . These contamination values meet the criteria of the draft standard and will result in very low doses to the public. However, these levels are also greater than the contamination levels that can be achieved, especially for items of new manufacture. The committee that drafted the standard was of the opinion that items released to the general public should not have contamination levels in excess of  $5000\text{ dpm}/100\text{ cm}^2$ .

If contamination levels are maintained below the level of  $5000\text{ dpm}/100\text{ cm}^2$ , it becomes difficult to envision how an individual could receive a dose equivalent in excess of 1 mrem. One millirem is the dose equivalent to members of the public below which the NRC (in their proposed revisions to 10 CFR 20) would not require inclusion in collective dose evaluations.

Regulatory Guide 8.21 (NRC 1979) has a recommended action level for low-risk beta-emitters in unrestricted areas of manufacturing plants at  $1 \cdot 10^{-6} \mu\text{Ci}/\text{cm}^2$  (i.e., 220 dpm/100  $\text{cm}^2$ ). Note however, that release of contaminated items to the general public may become a public relations problem.

## 4.13 References

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## **5.0 Tritium in the Body**

## 5.0 Tritium in the Body

This section describes the mechanisms by which tritium compounds may enter the body, the incorporation of tritium in the body, the distribution patterns of tritium compounds in the body, and the excretion of tritium.

### 5.1 Intake Mechanisms

An understanding of how tritium enters the body can be used in finding ways to reduce the total magnitude of intakes. Moreover, the information can be used to estimate the intake that will result from an accident.

Tritium can be taken into the body by four different mechanisms: inhalation, percutaneous absorption, ingestion, and injection. Inhalation and percutaneous absorption are the most important intake mechanisms for the user of RL devices.

#### 5.1.1 Inhalation

Tritium may be inhaled if someone is exposed to airborne tritium. The total quantity inhaled (the intake) is the product of the air concentration of the material, the breathing rate, and the total time the individual was exposed to the airborne material.

Following inhalation of water vapor, absorption is rapid, being transferred from the lung to the blood with an apparent half-time of 1.9 to 2.7 minutes (Balonov, Dolgirev, and Likhtarev 1974), and complete, with virtually all (~99%) THO vapor inhaled by humans being absorbed (Pinson and Langham 1957; Balonov, Dolgirev, and Likhtarev 1974; ICRP 1979).

A small amount of inhaled elemental tritium is also retained by the body. This retention is due to the slight solubility of hydrogen gas in body fluids and oxidation of elemental tritium to tritiated water (Pinson and Langham 1957).

The fraction of the inhaled elemental tritium converted to tritiated water in the body is approximately 0.0044% (ICRP 1975).

#### 5.1.2 Percutaneous Absorption

The skin is very permeable to tritiated water but relatively impermeable to elemental hydrogen. When an individual is exposed to tritiated water vapor, the intake due to absorption through the skin may range from one-half that due to inhalation (ICRP 1979) to an amount equal to that due to inhalation (IAEA 1987).

#### 5.1.3 Ingestion

Ingestion may occur when tritium has been incorporated into either food or drink. For the user of RL devices, this will not be common. However, ingestion may occur if an individual has handled a device that has external contamination and then eats without first having washed his hands.

All tritiated water ingested will be taken into the body (Pinson and Langham 1957; ICRP 1979). Absorption of tritiated water through the gastrointestinal (GI) tract begins 2 to 9 minutes after ingestion (Pinson and Langham 1957). The intake is essentially complete and equal tritium concentrations have been found in urine and blood levels from 2 to 2.5 hours after an intake (Balonov, Dolgirev, and Likhtarev 1974).

### 5.1.4 Injection

Injection may occur if someone handles an RL device that breaks and parts of the device (e.g., glass shards) penetrate the skin and force tritium into the body. Injection can be avoided by taking care when handling the devices and by wearing leather gloves when handling broken devices.

## 5.2 Biodistribution

Tritium will distribute itself in the body following the characteristics of its chemical form. Because tritium in RL devices is either tritiated water or elemental tritium, the tritium taken into the body following an intake of tritium from these devices will be distributed in the body water.

### 5.2.1 Retention and Excretion

The retention of tritiated water in the body may be described as a sum of exponentials (ICRP 1979) as shown below:

$$R(t) = q \sum_{i=1}^n C_i \exp(-\lambda_i t) \quad (5.1)$$

where  $R(t)$  = the retention of tritiated water at time,  $t$ , in  $\mu\text{Ci}$   
 $q$  = the intake of tritiated water,  $\mu\text{Ci}$   
 $C_i$  = the coefficient for the  $i$ th compartment, dimensionless  
 $\lambda_i$  = the retention coefficient for the  $i$ th compartment,  $\text{days}^{-1}$   
 $t$  = the time after the intake, days.

Each exponential term can be interpreted to represent a compartment within the body. The literature indicates that the equation may have up to four exponential terms. The first, and major, term is associated with the body water that is rapidly

exchanged. The other terms are associated with tritium that has been exchanged with organic hydrogen, etc.

Most researchers who follow the excretion of a person who has been exposed to tritiated water may observe only a one-compartment excretion curve. The second compartment, and particularly the third and fourth compartments, will all be observed only under rigidly controlled, research-type conditions. Because of the difficulty in observing the longer-lived compartments, reports on the excretion of tritiated water may commonly refer only to the shorter half-life component. Interpretation of bioassay data is based on the use of only the single major component. Because the minor components indicate a longer retention than would be estimated from a single-compartment excretion curve, the final dose estimate must be corrected to account for the increased retention.

### 5.2.2 Factors Affecting Biological Half-Time

The half-time of tritium in the body is affected by those factors influencing perspiration, consumption of water, or the body's retention of water. Such factors include the season, the amount of water consumed, the taking of diuretics, and age.

**Seasonal Variation.** The season of the year will influence the biological half-time of tritiated water. Butler and Leroy (1965) observed that half-times were shorter when assimilation occurred in warmer weather (8.3 days for spring and summer, but 10.4

days for fall and winter). The seasonal variation may be because of both increased intake of water and increased perspiration during the warmer months.

**Water Consumption.** A major factor affecting the biological half-time of tritiated water in the body is the body's rate of water turnover. Because water is itself a diuretic, its increased consumption will speed its turnover. The faster the water turnover, the briefer the biological half-time of tritiated water.

**Diuretics.** Diuretics are a commonly prescribed medication for hypertensive people. Treatment of hypertension with diuretics requires that the individual be nearly dehydrated; the turnover of body water in these individuals should be expected to be more rapid than normal. Because of their dehydrated state, the total body water content for hypertensives treated with diuretics may also be less than expected.

**Age.** Butler and Leroy (1965) found the biological half-life of tritiated water to correlate inversely with the individual's age.

### 5.2.3 Incorporation of Tritium into Biomolecules

A small amount of the tritium taken into the body either as water or as elemental gas can be incorporated into biomolecules.

**Tritiated Water.** Tritiated water is assumed to be uniformly distributed throughout the body water (ICRP 1979; IAEA 1987). Within minutes of ingestion, tritiated water attains a uniform concentration in the body (Commerford 1984). Pinson and Langham (1957) determined that soft tissues contain about 70% water, except for fat, which contains about 25% water. A small amount of THO tritium is fixed in organic molecules (IAEA 1987). Virtually all (98% to 99%) inhaled tritiated water is absorbed in the lung and the process takes only a few seconds (Carsten 1979). Absorption of THO through the skin is slower; there is about a 10-minute delay between absorption into the skin and entrance into the blood plasma (Carsten 1979), and

the maximum concentration in the blood will be within 2 hours after absorption through the skin. THO absorption through the GI tract begins in 2 to 9 minutes and is completed in 40 to 45 minutes (Pinson and Langham 1957).

According to Carsten (1979), passage from the blood plasma to extracellular water requires about 10 minutes to reach equilibrium and another 2 minutes to pass from extracellular to intracellular tissues. The World Health Organization assumes that tritiated water reaches equilibrium between blood and extracellular fluid in about 12 minutes. However, in poorly vascularized tissues, such as bone and fat, equilibrium with plasma water may take days to weeks (WHO 1983).

**Elemental Tritium.** Elemental tritium will be brought into the lung during inhalation, when the tritium gas will irradiate the cells of the lung. A small fraction of elemental tritium is dissolved in the blood and distributed throughout the body. The ICRP (1979) assumes that tritium will irradiate only the lung during inhalation.

Of the elemental tritium that is dissolved in the body fluids, much less than 1% will be oxidized to tritiated water. The mechanism for oxidation is not well established in humans, but in rats the role of the intestinal flora seems clear (Pinson and Langham 1957). Tritiated water formed by oxidation in the body behaves identically to tritiated water that is taken into the body by other means.

Pinson and Langham (1957) report that THO appeared at equivalent rates in the body fluids of men after they inhaled TH and THO when the specific activity of TH in ambient air was ~15,000 times that of THO. This seems to imply that about 0.007% of the inhaled T<sub>2</sub> is converted to THO. Although Pinson and Langham discuss the possibility of oxidation of TH to THO, they do not quantify this reaction. In rats, the oxidation of TH to THO has been shown to be due to bacteria in the intestinal tract (Pinson and Langham 1957).

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## **6.0 Calculating Doses from Tritium RL Devices**

## 6.0 Calculating Doses from Tritium RL Devices

Radiation dosimetry has two forms: external dosimetry and internal dosimetry. External dosimetry refers to the radiation dose that will result from radiation emitted from sources outside the body. Internal dosimetry refers to the radiation dose that results from radioactive materials inside the body. Because the major hazard from tritium is from internal radiation doses, this chapter primarily discusses internal dosimetry.

### 6.1 External Dosimetry

Tritium generally does not present an external radiation hazard because it emits no x-rays or gamma radiation except an ionizing beta particle, which has very low energy. The range of the tritium beta in air is very short; unless very close to the tritium source, an object will not be irradiated.

Even if a person's skin is in direct contact with tritium, there will be no external dose due to the beta particle. In water, 90% of the beta energy emitted by tritium will be deposited within  $2.19 \cdot 10^{-4}$  cm of the tritium source; essentially all the beta energy emitted will be deposited within  $3.94 \cdot 10^{-4}$  cm (Berger 1971). The dead layers of the skin range in thickness from 4 to 40 mg/cm<sup>2</sup> (ICRP 1975), which is  $4.0 \cdot 10^{-3}$  to  $4.0 \cdot 10^{-2}$  cm; consequently, the beta particle will not deposit energy in the underlying basal layer of the skin, its radiation-sensitive portion (ICRP 1977).

### 6.2 Internal Dosimetry

The major hazard from tritium is from the internal dose, that is, the radiation dose from radioactive material inside the person. There are two major types of internal dose calculations that can be made: prospective dose calculations and retrospective dose calculations.

#### 6.2.1 Prospective Dose Calculations

Prospective dose calculations are employed to estimate the radiation dose that may result from a particular practice. Calculations are performed prior to work being done with the radiation source. The purpose is to estimate the relative safety of certain actions. For example, it may be of interest to know the dose that might occur after the postulated release of a radioactive material.

Prospective dose estimates generally begin with the assumption that a release of radioactive material of a known magnitude has occurred. Then, the quantity of the release that will be taken into the body is computed; the intake is based on assumed characteristics of the person. The final step is the computation of the radiation dose that is due to the material taken into the body. Throughout the dose estimation process, assumptions must be made about the characteristics of the released material and about the characteristics of the human bodies that have been exposed.

The behavior of the radionuclide in the environment is projected from past observations. Because these calculations rely heavily on assumptions, standard models of radionuclide behavior are used extensively.

In making assumptions about the behavior of the radionuclide inside the body, it is conventional practice to compute the radiation dose to a "reference man," as described by the ICRP (1975). Perhaps no one actually matches this reference man in every way, but it provides a standard that may be used for comparison.

In the computation of prospective doses, it is also useful to estimate the committed effective dose equivalent for the intake. For tritiated water, 99% of the committed effective dose equivalent is received in about 2 months (66 days) and 99.9% is received in about 3 months (100 days).

## 6.2.2 Retrospective Dose Calculations

Retrospective dose refers to that received by an actual person; it is calculated after the individual has taken in radioactive material. Retrospective dose calculations generally begin by estimating the amount of radioactive material that has been taken into the body. These estimates are based on bioassay measurements; for tritium, the predominant bioassay measurement is the tritium content of urine. After the radionuclide content of the body has been estimated, the radiation dose received as a result of that content is estimated.

The most common adjustment for dose estimates is the effective half-time of tritium in the individual. The effective half-time of tritium in the body is a function of both the biological half-time (due to excretion of tritium) and the radioactive half-life of tritium. In practice, for tritiated water the effective half-time equals the biological half-time of tritiated water in the body. Another adjustment in dose calculations is *quality factors*, which are used to account for the fact that the harm to an individual, per unit of exposure, will vary according to the type of radiation.

Dose calculations should use as much information about the incident and the individual exposed as possible. That is, the health physicist should determine the individual's unique biological half-time, soft tissue mass, and water volume. However, it is not always possible to determine the specific values for all parameters of the dose calculations. If dose must be estimated without knowledge of the specific individual's data, the data for a reference man is used.

## 6.3 Health Hazards of the Forms of Tritium

For the chemical forms of tritium considered in this report, the tritium may be considered to reside almost exclusively in the water of the body. The total activity of the tritium in the body water is the product of the concentration of tritium in the body water times the total volume of the body water. For tritiated water in the body, the radioactivity in the body is most conveniently computed by measuring the specific concentration of tritium in urine and then multiplying that concentration by the total volume of body water.

Summation of the committed dose equivalent due to tritium gas yields a value that, according to the calculations presented above, is exactly 15,000 times less than that for tritiated water. If the tritiated water content of a postulated release is greater than about 0.01% of the total release, then the dose due to the tritiated water will predominate. Unless the gas has been dried just before a release, tritiated water will almost always predominate because the available tritium gas normally has a water content of about 0.01%. While tritium gas remains encapsulated in light fixtures, some tritiated water forms, with the result that about 1% of the tritium activity is in the form of THO.

## 6.4 Bioassay Measurements of Tritium

There are three categories of bioassay measurements: preparatory, routine, and diagnostic (ANSI 1983).

Individuals involved in tritium research and development and in manufacturing that use tritium should consider participating in preparatory and routine bioassay programs. A preparatory bioassay is used to collect a base-line measurement to determine an individual's normal level of tritium. At least one *preparatory bioassay* sample should be taken from each person who will be working with tritium. Multiple samples taken over a range of time, e.g., one week, will provide a more accurate measurement.

*Routine bioassays* are collected periodically to ensure that tritium intakes have not occurred. Individuals who routinely work with tritium in an industrial or research setting should be included in a routine bioassay program. ANSI standard N13.14-1983 requires weekly bioassays until equipment and procedures have proven satisfactory; bioassays are collected biweekly after that. Individuals working with tritium in a research or industrial setting can be expected to show elevated levels in the urine because it is difficult, if not impossible, to avoid small intakes.

For users of RL lighting systems, only *diagnostic bioassays* are necessary; in normal use, RL devices should release little tritium. Diagnostic bioassays are performed whenever it is possible that an individual has been exposed to unencapsulated tritium. For users of RL devices, diagnostic bioassays should be performed for those individuals who have been near a broken light. The purpose is to determine whether the person has experienced an intake, and, if so, whether dose reduction measures are warranted.

## 6.5 Basic Dosimetry Concepts

In this section, the basic concepts of dosimetry for tritium are described, followed by the specific equations that may be used for prospective and retrospective dose estimation.

### 6.5.1 Dose Equivalent Rate

The dose equivalent rate to an organ containing a beta-emitting radionuclide may be computed using the following equation:

$$H_T = Q \cdot 51.22 \cdot q(t) \cdot \frac{\epsilon}{M} \quad (6.1)$$

where  $H_T$  = dose equivalent rate, rem/day  
 $Q$  = the quality factor  
 $51.22$  = a conversion factor, (rad-g)/(MeV- $\mu$ Ci-day)  
 $\epsilon$  = radiation energy absorbed in the organ, MeV

$q(t)$  = the activity of the radioactive material in the tissue,  $\mu$ Ci (see Equation [6.22])  
 $M$  = mass of the organ, g.

The retention of tritiated water can be described by a single exponential curve so that

$$q(t) = q(0) \exp(-\lambda_e t) \quad (6.2)$$

where  $q(0)$  = the amount of activity taken into the body,  $\mu$ Ci  
 $\lambda_e$  = the effective half-time of tritiated water in the body, days  
 $t$  = the elapsed time between date of intake and date of urine sample, days.

### 6.5.2 Committed Dose Equivalent

The dose equivalent rate can be integrated over time to calculate the total dose equivalent that may be received following an intake of tritium. Integration of Equation (6.1) with respect to time, following

substitution of Equation (6.2) for  $q(t)$  and 0.00568 MeV for  $\epsilon$ , results in the following equation:

$$H_T = 0.42 \frac{q(0)QT_e}{M} [1 - \exp(-\lambda_e t)] \quad (6.3)$$

where  $H_T$  = integral dose equivalent, rem  
 0.42 = constant (product of 51.22·0.00568·1.44), (rad·g)/(μCi·day)  
 $T_e$  = effective half-time of tritium in the body, days  
 $Q$  = quality factor, dimensionless  
 $q(0)$  = activity taken into the body, μCi  
 $M$  = mass of the soft tissue, g  
 $\lambda_e$  = effective removal constant of tritium from the body, days<sup>-1</sup>  
 $t$  = time since the uptake, days.

To compute the committed dose equivalent, it is necessary to integrate the dose equivalent rate over the 50 years following the intake (ICRP 1977). Although the integration is for 50 years (in accordance with the formality of the ICRP [1977]), virtually all the dose from tritiated water will have been received within 3 months after an intake. Substituting  $t = 50$  years (18,250 days), the above equation becomes:

$$H_{T,50} = 0.42 \frac{q(0)QT_e}{M} \quad (6.4)$$

where the variables are as defined above.

The effective half-time of tritium in the body is a function of both the biological half-time and the radioactive half-life of tritium. In practice, for tritiated water the effective half-time equals the biological half-time of tritiated water in the body.

Because the mass of the body is normally given in terms of kilograms, the above equation may be rewritten as:

$$H_{T,50} = 4.20E-4 \frac{q(0)QT_e}{M} \quad (6.5)$$

where  $H_{T,50}$  = the committed dose equivalent, rem  
 4.20E-4 = a constant (0.42·kg/1000 g), (rad·kg)/(μCi·day)  
 $T_e$  = the effective half-time of tritium in the body, days  
 $Q$  = the quality factor, dimensionless  
 $q(0)$  = the activity taken into the body, μCi  
 $M$  = the mass of the soft tissue, kg.

The following paragraphs describe the values that may be used in the above equations.

### 6.5.3 Quality Factor

Quality factors are used to account for the fact that the harm to an individual, per rad, will vary according to the type of radiation. The quality factors for tritium range over a value from 1 to 2, as shown in Table 6.1. Because different regulatory agencies might require different values for the quality factor, the calculations in this report are based on rad, with a notation to multiply by the appropriate quality factor. The NRC requires that a quality factor of 1.7 be used by its licensees; however, recent NRC documents (see Scherpelz et al. 1986a; NRC 1987) have employed  $Q = 1$  in their calculations. In this report, we explicitly list the quality factor symbol; this serves to emphasize the fact that quality factor may vary depending on regulatory considerations. In those instances where regulations do not require the use of  $Q = 1.7$ , a quality factor of unity should be employed to mirror the current recommendations of the ICRP and NCRP.

### 6.5.4 Tritium Radioactivity in the Body

For the chemical forms of tritium considered in this report, the tritium may be considered to reside almost exclusively in the water of the body. The total activity of the tritium in the body water is the product

**Table 6.1.** Published Values of the Quality Factor for Tritium

Quality Factor	Reference
1	ICRP (1977, 1979a); NCRP (1987); DOE (1987)
1.7	Brodsky (1983); NRC (1983) <sup>(a)</sup>
2	ICRU (1986)

(a) U.S. Nuclear Regulatory Commission (NRC 1983), "Applications of Bioassay for Tritium," in *Draft Regulatory Guide and Value/Impact Statement Task OP 713-4*. U.S. Nuclear Regulatory Commission, Washington, D.C.

of the concentration of tritium in the body water times the total volume of the body water. For tritiated water in the body, the activity in the body,  $q(t)$ , is most conveniently computed by measuring the specific concentration of tritium in the urine and then multiplying that concentration by the total volume of body water. Specific examples showing how to estimate the activity in the body are given in later sections of this report.

### 6.5.5 Mass of Target Material

The ICRP recommendations (1979a) indicate that the irradiated materials are the soft tissues of the body. Table 6.2 shows the values that can be used to compute the dose equivalent rate to the body from tritium contained in the body. Note that the values for the "reference female" were estimated for this report, not by the ICRP.

When performing prospective dose calculations, the soft tissue mass for ICRP reference man (ICRP 1975) should be used.

ICRP Publication 30 (ICRP 1979) combines  $\epsilon$ ,  $Q$ , and  $M$  of Equation (6.1) to form the specific effective energy (SEE). The SEE is computed for a person known as reference man, described in ICRP 23

(ICRP 1975). The dose equivalent to a reference man from tritiated water is given by:

$$H = Q \text{ 51.22 SEE } q(t) \quad (6.6)$$

where the values are as previously described. In the following discussions, the SEE and the constant 51.22 are combined.

## 6.6 Prospective Dose Calculations

In this section, the equations are derived that may be used to calculate prospective doses. Throughout this section, we show  $Q$  explicitly in the equations. In some cases, the addition of  $Q$  to the equation is not correct because the formalism employed by the ICRP includes a quality factor of 1. The  $Q$  is included in the equations to remind the reader that some regulatory agencies may require a  $Q$  greater than 1 (see Brodsky 1983).

Prospective dose calculations are often used to estimate the radiation dose to an individual who may be exposed to the radioactive material. Because the dose estimations are hypothetical, they are computed for an individual who has the characteristics of the ICRP reference man (ICRP 1975).

The prospective dose calculations described here are for ingestion of tritiated water, submersion in tritiated water vapor (along with subsequent inhalation), inhalation of tritium gas, and submersion of an individual in tritiated water.

### 6.6.1 Ingestion of Tritiated Water

The committed dose equivalent to reference man from ingestion of tritiated water is from Equation (6.5) and substitution for assumed values for  $M$  (63 kg) and  $T_e$  (10 days) as follows:

**Table 6.2.** Anatomical and Health Physics Values for Reference Persons

Item	Male	Female	Reference
Total mass	70 kg	58 kg	ICRP (1975, p. 13)
Body water content	42 L	29 L	
	0.6 L/kg	0.5 L/kg	ICRP (1975, p. 29)
Soft tissue mass	63 kg	54 kg <sup>(a)</sup>	ICRP (1979)
Skeletal mass	7 kg	4.2 kg	ICRP (1975, p. 74)
SEE SI units	9.0E-8 MeV/(g-trans)	1.1E-7 MeV/(g-trans)	(b)
Traditional units	1.9E-1 rad/(Ci-h)	2.2E-1 rad/(Ci-h)	

(a) Female value calculated by subtracting skeleton mass (ICRP 1975, p. 74) less the bone marrow mass (ICRP 1975, p. 90) from the total mass.

(b) Computed by dividing <sup>3</sup>H beta transition energy (5.68E-3 MeV) (ICRP 1983).

$$H_{E,50} = Q \cdot 66.6E-6 \cdot q(0) \quad (6.7)$$

where  $H_{E,50}$  = the committed effective dose equivalent, rem

$Q$  = the quality factor, dimensionless  
 $66.6E-6$  = conversion factor ( $420E-6 \cdot 10/63$   
 $= 66.6E-6$ ), rad/ $\mu$ Ci

$q(0)$  = the quantity of tritiated water ingested,  $\mu$ Ci.

where  $C$  is the air concentration of tritiated water in  $\mu$ Ci/mL;  $T$  is the exposure time in minutes; and  $20.0E3$  is the "light work" breathing rate in mL/min. Substitution into Equation (6.8) yields

$$H_{E,50} = Q \cdot 1.33 \cdot C \cdot T \quad (6.9)$$

where  $H_{E,50}$  = the committed dose equivalent, rem

$Q$  = the quality factor, dimensionless  
 $1.33$  = conversion factor ( $66.62E-6 \cdot 20.0E3 = 1.33$ ),  
 mL-rad/ $\mu$ Ci-min

$C$  = the air concentration of tritiated water,  $\mu$ Ci/mL

$T$  = the exposure time, min.

### 6.6.2 Submersion in Tritiated Water Vapor

When an individual is submerged in tritiated water vapor, there are two primary sources of radiation dose: 1) from inhaled tritiated water vapor and 2) from tritiated water absorbed through the skin.

**Inhaled Water Vapor.** All inhaled water is taken up by the body (see discussion on intake mechanisms). Thus, the previous equation may be used by computing the intake,  $q(0)$ , from the equation:

$$q(0) = 20.0E3 \cdot C \cdot T \quad (6.8)$$

### Percutaneous Absorption of Tritiated Water Vapor.

The quantity of tritiated water that may be absorbed through intact skin can be estimated using the information given in the section that described intake mechanisms. However, for estimating prospective dose, it is convenient to employ the ICRP assumption (ICRP 1977) that total percutaneous absorption will equal about one-half the intake due to inhalation at the "light work" breathing rate. NRC licensees, however, should be aware that a

footnote to 10 CFR 20.103(a)(1) indicates that the assumption employed to prepare the tables of maximum permissible concentrations was that intakes by the two routes were assumed to be equal. The committed dose equivalent from percutaneous absorption of tritium is then:

$$H_{E,50} = Q 0.63 C T \quad (6.10)$$

where  $H_{E,50}$  is the committed dose equivalent in rem and 0.63 is the conversion factor (1.33/2) in mL-rad/ $\mu$ Ci-min and other variables are as previously defined.

Thus, the total committed dose equivalent due to submersion in tritiated water vapor is the sum of that due to inhalation and percutaneous absorption:

$$H_{E,50} = Q 2.0 C T \quad (6.11)$$

where the variables are as previously described.

### 6.6.3 Inhalation of Tritium Gas

Following inhalation of tritium gas, radiation dose may be accrued from three sources: dose to the lung from tritium in the lung, dose to the whole body from elemental tritium absorbed in the blood, and dose to the whole body from tritiated water formed from the inhaled tritiated gas.

#### Dose to Lung from Inhalation of $^3\text{H}$ Gas.

The ICRP (ICRP 1979, Supplement to Part 1) lists the dose equivalent rate to the lung due to tritium gas in the air as  $611.2\text{E-}6$  rem-mL/min- $\mu$ Ci ( $9.9\text{E-}15$  Sv- $\text{m}^3/\text{Bq-h}$ ). The dose to the lung from exposure to elemental tritium at an air concentration of  $C$   $\mu$ Ci/mL for time  $T$  minutes is then

$$H_T = Q 611.2\text{E-}6 C T \quad (6.12)$$

where  $H_T$  is the dose equivalent to the lungs, in rem, and other variables are as previously defined.

Because the dose to the lung will cease when the source is removed, the above dose also represents the committed dose equivalent to the lung from elemental tritium in the lung. The effective dose equivalent from the lung dose is computed by multiplying Equation (6.12) by the lung weighting factor of 0.12 (ICRP 1977). The equation for the effective dose equivalent in the lung is then:

$$H_{E,50} = Q 73.33\text{E-}6 C T \quad (6.13)$$

where  $H_{E,50}$  is the committed effective dose equivalent, in rem, due to irradiation of the lung by elemental tritium in the inhaled air.

#### Elemental Tritium Dissolved in the

**Blood.** To compute the dose to the whole body from gaseous tritium dissolved in the blood, the following assumptions are made. First, elemental tritium in blood will uniformly irradiate the whole body. Second, the solubility of tritium in blood is equal to that of hydrogen in water at the same temperature. Third, the dose to the whole body from  $^3\text{H}$  dissolved in body fluids is equal to the dose from tritiated water. Based on this last assumption, the dose equivalent rate conversion factor is  $1.9\text{E-}7$  rad/ $\mu$ Ci-day.

The dose equivalent to the body from tritium dissolved in the blood is then given by the equation:

$$H_E = \frac{(3.2\text{E-}9) (1.713\text{E}9) C T}{2.37\text{E}6} \quad (6.14)$$

where  $3.2\text{E-}9$  = a conversion factor, rad/ $\mu$ Ci  
 $1.713\text{E}9$  = the quantity of  $\text{T}_2$  dissolved in blood at saturation,  $\mu$ Ci  
 $2.37\text{E}6$  = the maximal air concentration of tritium gas,  $\mu$ Ci/mL  
 $T$  = the exposure time, min  
 $C$  = air concentration of  $\text{T}_2$  gas,  $\mu$ Ci/mL.

Equation (6.14) is equivalent to

$$H_E = Q \cdot 2.32E-6 \cdot C \cdot T \quad (6.15)$$

where the units are as defined above. The dose to the whole body from dissolved tritium gas is a small fraction of the effective dose equivalent received due to gas in the lung.

### Tritiated Water Converted from Inhaled

**Tritium Gas.** About 0.003% to 0.004% of the inhaled tritium gas oxidizes to tritiated water in the body (Brodsky 1983). The dose to the whole body from this route of intake is then:

$$H_{E,50} = Q \cdot C \cdot (20,000T) \quad (6.16)$$

$$(4E-5) (66.6E-6)$$

where  $4E-5 = 0.004\%$  (percentage of  $^3\text{H}$  gas oxidized to tritiated water)  
 $20,000 =$  the "light work" breathing rate, mL/min  
 $66.6E-6 =$  a conversion factor in rad/ $\mu\text{Ci}$  (see Equation [6.7]).

Equation (6.16) is equivalent to

$$H_{E,50} = Q \cdot 53.28E-6 \cdot C \cdot T \quad (6.17)$$

where  $H_{E,50}$  is the committed dose equivalent in rem and  $C$  is the air concentration in  $\mu\text{Ci}/\text{mL}$  and other variables are as previously defined.

In summary, the committed effective dose equivalent due to the conversion of tritium gas to tritiated water is nearly equal to that due to irradiation of the lung by tritium gas. In this report, the committed effective dose equivalent due to inhalation of tritium gas includes all three components of dose; it is the sum of the three preceding equations and is given by the equation:

$$H_{E,50} = Q \cdot 128.8E-6 \cdot C \cdot T \quad (6.18)$$

where the variables are as previously described.

Note: Approximately 5% to 10% differences can be obtained for the constants in the above equations, depending on whether one uses the integrated values listed in ICRP 30 (1977) or uses the dose rate values and then integrates. These differences are rounding errors.

## 6.6.4 Discussion of Prospective Dose Calculation

Summation of the committed dose equivalents for tritium gas yields a value that, according to the calculations presented above, is exactly 15,000 times less than that for tritiated water. If the tritiated water content of a postulated release is greater than about 0.01% of the total release, then the dose due to the tritiated water will predominate. Unless the gas has been dried just before a release, tritiated water will almost always predominate because the available tritium gas normally has a water content of about 0.01% (see Table 2.2). While tritium gas remains encapsulated in light fixtures, some tritiated water forms, with the result that about 1% of the activity is THO.

In the above equations we have shown the air activity concentration and the time of exposure. These two components together can be called the integral air concentration, expressed in  $\mu\text{Ci}\cdot\text{min}/\text{mL}$ .

When performing calculations based on air concentration of TH or  $T_2$ , assumptions must be made with care. Humans require a partial pressure of oxygen ( $p_{O_2}$ ) of at least 50-mm Hg for survival; depression of the respiratory center occurs at a  $p_{O_2}$  of about 64-mm Hg (Jensen 1976), but the exact values will depend on the individual's physical condition. The  $p_{O_2}$  at sea level is about 159-mm Hg (Jensen 1976); thus, dilution of the air by hydrogen to about 40% of normal levels will result in asphyxiation. This will occur when the  $T_2$  concentration is about  $1.4E6 \mu\text{Ci}/\text{mL}$ . Assumptions of air concentrations exceeding 100 or 200  $\text{mCi}\cdot T_2/\text{mL}$  air should be carefully checked to ensure that they are reasonable.

Hydrogen in air at concentrations of 4% to 75% by volume are flammable/explosive (Olishifski and

McElroy 1971). These concentrations equate to 0.0948 to 1.778 Ci/cm<sup>3</sup> air for T<sub>2</sub> and 0.0474 to 0.889 Ci/cm<sup>3</sup> for TH. Flammability and/or explosive conditions are another reason to carefully check for reasonableness of any assumptions about air concentrations of tritiated gas.

## 6.7 Retrospective Dose Calculations

Retrospective dose calculations are made for an individual who has been exposed to radioactive materials. Such calculations should use as much information about the individual as possible. That is, the health physicist should determine the individual's unique biological half-time, soft tissue mass, and water volume. If dose must be estimated without knowledge of the specific individual's data, the data for reference man (or woman) should be used.

### 6.7.1 Dose Equivalent Rate

The equations to use are those in Section 6.5.1 with one modification. These equations assumed that all the tritium remains in the body water; this assumption is not entirely correct. Tritium exchanges with the hydrogen in the organic constituents of the body. Hence, the specific concentration in the body water is lower than it should be for accurate dose estimates. It was assumed also that tritiated water is eliminated from the body in a manner that can be described by a single exponential curve. This assumption too is not correct. There is some transfer of tritium to the biological molecules of the body, and the excretion pattern has been observed to follow a three-exponential decay curve. For these reasons, the computed dose based on a single exponent should be increased by 10% (ICRP 1979; Commerford 1984; Health and Welfare Canada 1982 [n.b. the regulatory guidance of Canada states 20%, but the example calculations are consistent with 10%]). The equations in the section below have been corrected for this increase.

Equation (6.19) is used to estimate the dose equivalent rate to an individual:

$$H_E = 3.2E-4 \frac{q(0)Q}{M} \exp(-\lambda_e t) \quad (6.19)$$

where  $H_E$  = the dose equivalent rate following an intake of tritiated water, rem/day  
 $3.2E-4$  = a constant (product of 51.22·0.00568 MeV·1 kg/1000g), (rad·kg)/(μCi·day)  
 $Q$  = the quality factor, dimensionless  
 $q(0)$  = the activity in the body at the time of intake, μCi  
 $M$  = the mass of the soft tissue, kg  
 $\lambda_e$  = the effective removal constant of tritium from the body, days<sup>-1</sup>  
 $t$  = the time since the uptake, days.

### 6.7.2 Committed Dose Equivalent

The previous dose rate equation is then integrated over time to yield:

$$H_E = 4.6E-4 \frac{q(0)QT_e}{M} (1 - \exp(-\lambda_e t)) \quad (6.20)$$

where  $H_E$  = the committed DE, rem  
 $4.6E-4$  = a constant (product of 3.2E-4 · 1.44), (rad·g)/(μCi·day) (1.44 = 1/ln 2)  
 $T_e$  = the effective half-time of tritium in the body, days  
 $Q$  = the quality factor, dimensionless  
 $q(0)$  = the activity taken into the body, μCi  
 $M$  = the mass of the soft tissue, g  
 $\lambda_e$  = the effective removal constant of tritium from the body, days<sup>-1</sup>  
 $t$  = the time since the uptake, days.

To compute the committed dose equivalent, it is necessary to integrate the dose equivalent rate over the 50 years following the intake as recommended by the ICRP (ICRP 1979). If  $t = 50$  years, this equation becomes:

$$H_{E,50} = 4.6E-4 \frac{q(0)QT_e}{M} \quad (6.21)$$

where the variables are as defined above.

### 6.7.3 Personalization of Variables

To permit employing as much individual-specific information as possible, values for the above equations are defined below.

The most common adjustment for dose estimates is the individual's biological half-time. It is not always possible to determine the specific values for all parameters of the equations; consequently, default values that can be used are also shown.

### 6.7.4 Tritium Activity in the Body

The activity of tritium in the body is the only value that must be known for dose assessment. This activity is the product of the specific concentration of tritium in the body water and the volume of the water in the body. The equation to compute activity in the body is:

$$q(t) = C_t V \quad (6.22)$$

where  $q(t)$  = the activity of tritium in the body at the time of measurement,  $\mu\text{Ci}$   
 $C_t$  = the specific concentration of tritium in the body water at the time of measurement,  $\mu\text{Ci/L}$   
 $V$  = the volume of the total body water, L.

#### Concentration of Tritium in Body Fluids.

The specific concentration of tritium in body water must be estimated by direct measurement. All body fluids--water, intracellular water, lymph, blood, tears, perspiration, urine, etc.--are assumed to have the same, or equal, concentration. Measurement of the tritium concentration of any will provide the concentration in all. Because urine can be collected without causing trauma, it is most commonly used to estimate

the tritium concentration of body water. The specific concentration of urine can be determined by one of the methods described in Section 7.0.

**Volume of Body Fluids.** The total body water (TBW) content of the individual may be estimated, or a default value may be employed. Most published procedures for dose calculations do not require that the TBW volume should be estimated. Equations (6.23) and (6.24) can be used to compute the TBW content for males and females, respectively (from ICRP 1975, p. 29):

$$TBW_m = \frac{W[79.45 - 0.24W - 0.12A]}{100} \quad (6.23)$$

$$TBW_f = \frac{W[69.81 - 0.26W - 0.15A]}{100} \quad (6.24)$$

where  $TBW_m$  = TBW content for males  
 $TBW_f$  = TBW content for females  
 $W$  = the weight of the individual, kilograms  
 $A$  = the individual's age, years.

If the information required for Equations (6.23) and (6.24) is not available, reference man or woman values can be used: 42 L (male) or 29 L (female).

### 6.7.5 Mass of the Target Tissue

The individual's soft tissue mass may be estimated by subtracting the skeletal mass from the individual's total mass. The skeletal mass that may be used is 7 kg for males and 4.2 kg for females. The authors have not found an acceptable method for modifying the estimate of skeletal mass for age, size, or other factors.

Some authors recommend that the soft tissue mass be estimated by subtracting both the mass of the skeleton (see Table 6.2) and the mass of fat from the individual's total mass. The fat mass can be determined by the difference between the individual's mass and ideal weight because generally at ideal

weight there is little excess body fat. Ideal weight is determined from tables such as those in the Geigy Scientific Tables (Diem and Lentner 1970). These tables require knowledge of the individual's frame size.

One way to determine frame size is to measure the height and the wrist circumference. Then, compute the ratio of the height divided by the wrist size, and compare the ratio to the values in Table 6.3. If the individual is muscular, this method will probably overestimate fat and underestimate lean body tissue, resulting in overestimate of dose. More accurate methods for determining lean body mass exist but may require considerable efforts (such as submersion of the person in water). If dose calculations are made for both the "fat included" and "fat excluded" cases, the dose to the individual should be well bounded.

The default values for mass of the soft tissue should be 63 kg for males and 54 kg for females (ICRP 1975).

### 6.7.6 Effective Half-Time

The effective half-time of tritium in the body is a function of both the biological half-time and the radioactive half-life of tritium. In practice, the effective

half-time for tritiated water in the body is equal to the biological half-time of tritiated water in the body.

The effective half-time of tritiated water may be determined by direct measurement. If two measurements are available, the effective half-time may be determined by one of two methods, graphical or numerical. The graphical method requires that the health physicist plot the measured specific concentration against time on semi-log graph paper (specific concentration is on the log axis, time on the linear axis). The effective half-time (the time required for the specific concentration to be reduced by one-half) can be determined readily by observing and evaluating the data in the graph. The effective half-time may also be determined from the following equation:

$$T_e = \frac{(t_2 - t_1) \log(2)}{\log(Ct_2/Ct_1)} \quad (6.25)$$

- where
- $T_e$  = the effective half-time, days
  - $t_1$  = the time of the first bioassay measurement, days
  - $t_2$  = the time of the second bioassay measurement, days
  - $Ct_1$  = the specific concentration of the second bioassay measurement,  $\mu\text{Ci/L}$
  - $Ct_2$  = the specific concentration of the first bioassay measurement,  $\mu\text{Ci/L}$ .

The log function may be either natural or common logs, but they must be consistent.

## 6.8 Bioassay Measurements

American National Standards Institute Standard N13.14-1983 (ANSI 1983) defines three categories of bioassay measurements: preparatory, routine, and diagnostic. For users of RL lighting systems, only the diagnostic measurements are necessary; in routine use, the devices should release little tritium.

Individuals involved in tritium research/development

**Table 6.3.** Determination of Frame Size<sup>(a)</sup>

	Ratio	Frame Size
Men	10.4 or greater	Small
	9.6-10.4	Medium
	less than 9.6	Large
Women	11 or greater	Small
	10.1-11	Medium
	less than 10.1	Large

(a) From Grant (1980).

and in manufacturing should consider whether to participate in preparatory and routine bioassay programs.

A preparatory bioassay is used to collect a baseline measurement to determine the individual's normal level of tritium. At least one preparatory bioassay sample should be taken from each person who will be working with tritium. Multiple samples taken over, say, one week will provide a more accurate measure.

Routine bioassays are collected periodically to ensure that tritium intakes have not occurred. Individuals who routinely work with tritium in an industrial or research setting should be included in a routine bioassay program. ANSI N13.14-1983 requires weekly bioassays until equipment and procedures have been proven satisfactory; bioassays are collected biweekly after that. Individuals working with tritium in a research or industrial setting can be expected to show elevated levels in the urine because it is difficult, if not impossible, to avoid small intakes.

### **6.8.1 Diagnostic Bioassay Program**

Diagnostic bioassays are performed whenever it is possible that an individual has been exposed to unencapsulated tritium. For users of RL devices, diagnostic bioassays should be performed for those individuals who have been near a broken light. The purpose is to determine whether the person has experienced an intake and, if so, to quantify the intake and to determine whether dose reduction measures are warranted. ANSI N13.14-1983 requires that samples for diagnostic bioassays be collected within one day of the incident. Diagnostic measurements should be made if the first sample results in a projected dose greater than 1 rem. The diagnostic bioassay program should continue at the rate of two samples per week until the committed dose equivalent after the last bioassay is less than 0.3 rem (ANSI 1983).

The tritium released immediately after a light breaks will most likely be elemental hydrogen, in which case

the greatest dose will be to the lung; the very small uptake will make it difficult to quantify the dose to the lung. With time, tritiated water that may be associated with the light will also be volatilized.

As it may be some time before the tritiated water volatilizes (Niemeyer 1969), total exposure to tritium can be minimized by placing the light in a shipping container as soon as the damage is noticed. Persons who handle a broken device should wear gloves and wash their hands immediately after handling it.

The individuals in a room where an RL device is damaged should be monitored for exposure to tritium. In case of an accident outdoors, individuals near the damaged device, especially those downwind, should be monitored for tritium exposure. The individual who handles the damaged light should also be monitored. Tritium exposure to the person who handles the damaged light can be minimized if plastic gloves are worn.

In the event of an accident, a sample should be collected within one day of the possible exposure (ANSI 1983).

### **6.8.2 Bioassay Sample Collection Techniques**

The collection of bioassay samples for tritium assays is relatively simple because, after a short time for equilibration, all body water contains nearly the same tritium concentration. Thus, any type of body water can be collected; urine is convenient. Collection of urine for tritium bioassay should follow these steps:

1. Obtain a clean sample bottle, one that will hold at least 100 mL.
2. Clean the hands and genital area.
3. Collect the sample some distance from the radiation zone, where it will not be contaminated.

4. Label the bottle.
5. If the sample is to be stored more than a few days or if it is to be shipped for measurement, a preservative may be added, e.g., Thymol (ANSI 1983) or thimerosal (Moghissi and Lieberman 1970). If a preservative is added, its name should be stated on the sample bottle.
6. After an acute intake, collect the sample not less than 1 h after the end of the possible exposure, and if the sample is urine, void at least 1 h after the possible exposure and produce a sample thereafter. Some agencies recommend that 4 h be allowed for equilibrium to be established in blood and urine (Health and Welfare Canada 1982).

Chiswell and Dancer (1969) report that some individuals may object to providing urine samples. They say that exhaled water vapor may be collected as an alternative, but they do not recommend the technique. They found that tritium concentration in exhaled water vapor differed from that in urine. The amount of the variation depended on the collection method; silica gel and molecular sieves yielded the greatest variation, while cold traps resulted in small variations. The differences were attributed to residual moisture in the desiccants.

### 6.8.3 Measurement Techniques

The bioassay sample may be measured according to any method discussed in Section 7.0. The American National Standards Institute (1983) disallows distillation of the bioassay sample unless the exposure is known to be only elemental tritium or tritiated water and oxidative distillation is employed. If the exposure is so large that the dose equivalent may exceed 1 rem, the time elapsed between sample collection and calculation of results shall be within 2 days (ANSI 1983).

### 6.8.4 Bioassay Interpretation and Dose Assessment

The interpretation of bioassay assumes a urine sample is collected after the tritium has been distributed throughout the body.

Tritium concentration in the urine may be estimated using any method described in Section 7.0. The units of the data received from the bioassay laboratory may vary. In the following equations, it is assumed that the urine concentration is in terms of  $\mu\text{Ci/mL}$ .

The quantity of tritiated water in the person may be assumed to equal the product of the urine concentration times the body water content, as shown in Equation (6.26). The dose rate to the individual at the time of sampling can then be computed using Equation (6.19).

If the date of intake is known, or can be estimated to within a day or two, then the amount of tritiated water taken into the body is estimated by back-calculating using the following equation:

$$q(0) = q(t)\exp(T_e t) \quad (6.26)$$

where  $q(0)$  = the activity taken into the body,  $\mu\text{Ci}$   
 $q(t)$  = the activity calculated for time =  $t$ ,  $\mu\text{Ci}$ , computed using Equation (6.22)  
 $T_e$  = the effective half-time of tritiated water in the body, days  
 $t$  = the elapsed time between the date of intake and the date the urine sample was taken, days.

The next step is to estimate the committed effective dose equivalent for the individual using Equation (6.20). If the date of intake was known and the original intake was computed as described above, then compute the  $H_{E,50}$  based on the presumed intake; otherwise, compute the  $H_{E,50}$  based on the observed activity in the body. Compare the

computed  $H_{E,50}$  with the various action levels given below; if the value is less than 0.3 rem, additional calculations are not necessary.

If the estimated  $H_{E,50}$  exceeds 0.3 rem, then the appropriate actions described below should be taken. One suggested action is to determine the person's individual excretion rate constant, which will require at least two samples per week. As a minimum, two urine samples are required to estimate the individual's excretion half-time.

### 6.8.5 Action/Emergency Levels

Table 6.4 describes the actions that should be taken once the magnitude of an intake is determined. In some cases, the actions are suggestions of the authors of this manual; in other cases, they are those that should be taken if the health physicist wants to be in compliance with the ANSI standard. Lastly, there are requirements of regulatory authorities. In some cases, the regulations require immediate notification. Because it is impossible to have performed a bioassay within such required time limits, the health physicist needs to have already determined what type of accident requires immediate notification (see Section 8.0, "Emergency Planning").

The action levels described in Table 6.4 are based on the effective dose equivalent to the person who inhales the radioactive material. More complete regulatory action levels, including those based on air concentrations and damage to facilities, are given in Section 8.0.

### 6.8.6 Dose Reduction Measures Applied After Intakes

The NCRP suggests that forced water is a therapy that should be considered immediately after an intake (NCRP 1980). Forcing fluids to tolerance, at least 3 to 4 L/day, reduces biological half-time to about 1/3 to 1/2 of normal (NCRP 1980). Pinson and Langham (1957) observed a biological half-life of 10 days in an individual who consumed 2.7 L/day; when the water intake was increased to 12.8 L/day in the same

subject, the biological half-time was reduced to 2.4 days. Pinson and Langham (1957) postulated that forcing liquids for 24 h/day would reduce the half-time to 1.5 days, but that was not verified experimentally. The dose is reduced proportionately to the effective half-time.

In an attempt to demonstrate the principle of reduced biological half-time following forced intake of water, Akaishi (1982) has shown data for which excess water intake apparently caused no change in biological half-time. This occurred because forcing extra water was voluntary for the exposed individual, who probably did not comply. Thus, although forcing water may be a good method in theory for reducing radiation dose, its actual achievement may be difficult.

Diuretics can also increase the body's water turnover, but should be used with care and only under supervision of a physician who has considered the possibility of electrolyte imbalances (NCRP 1980).

## 6.9 Dose Projections

This section provides general methods for calculating dose projections. Appendix B contains more detailed scenarios for two occupational doses (that is, for exposed individuals within 10 m of the tritium source) and for two environmental doses (for exposed individuals between 10 and 100 m from the source).

When the amount of tritium inhaled by a potentially exposed individual is not known, techniques of dose projection must be used. Air concentrations of tritium at the time of the exposure must be estimated, as must the amount of tritium released and the extent of diffusion.

Dose projections are distinguished by who might receive the radiation dose. *Occupational dose projections* are used to describe potential accidents in which the individual who receives the dose and the damaged device are in close proximity to each other, i.e., within a few meters. *Environmental dose*

**Table 6.4.** Action Levels

Total Effective Dose Equivalent	Emergency Actions
1. $H_E \leq 0.05$ rem	No actions are required.
2. $0.05 < H_E \leq 0.3$ rem	Determine cause of the release. Take remedial actions to prevent similar incidents. Determine whether other individuals could have been exposed to tritium also and perform bioassay measurements for them.
3. $0.3 < H_E \leq 1$ rem	Take actions as described above. Confirm the dose estimate with additional bioassay measurements; continue confirmatory bioassay measurements twice weekly until the remaining $H_E$ estimate is less than 0.3 rem (ANSI 1983).
4. $1 < H_E \leq 3$ rem	Take actions as described above. Restrict the worker's exposure to tritium until remedial actions (to preclude further exposures) have been established. The health physicist should consult a physician to determine whether dose reduction methods are appropriate (ANSI 1983).
5. $3 < H_E \leq 5$ rem	Take actions as described above. When the total effective dose equivalent (sum of internal and external) to an individual exceeds 3 rem in a quarter, a written report must be sent to the appropriate NRC regional office within 30 days (10 CFR 20.403). DOE 5484.1 requires notification to Headquarters by memo whenever the quarterly radiation exposure to an individual exceeds 3 rem.
6. $5 < H_E \leq 25$ rem	Take actions as described above. When the total effective dose equivalent (sum of internal and external) to an individual exceeds 5 rem, a report must be submitted to the appropriate NRC regional office by telephone or telegraph within 24 h of the incident [10 CFR 20.403(b)(1)]. DOE 5484.1 requires that DOE Headquarters be notified of this type of accident within 72 h, and calls for a Type B investigation of the incident (DOE 1981). At this exposure level, the appropriate Canadian authorities must be notified for Canadian exposures (Health and Welfare Canada 1982). The health physicist should consult a physician about whether or not to initiate dose reduction measures. This action is suggested by Canadian authorities also (Health and Welfare Canada 1982), but is a requirement if the health physicist wants to comply with ANSI N13.14-1983 (ANSI 1983). Other actions include the following:
7. $H_E > 25$ rem	<ul style="list-style-type: none"> <li>• Protect the individual from any further intakes.</li> <li>• Protect the individual from any further external exposure.</li> <li>• Determine the individual's retention function.</li> </ul> <p>This represents a very serious incident. An exposure of 25 rem or more requires immediate notification of the appropriate NRC regional office by telephone, telegraph, or facsimile [(10 CFR 20.403(a)(1)]. DOE 5484.1 also requires immediate notification to DOE Headquarters of this accident, and calls for a Type A investigation of this accident (DOE 1981).</p> <p>Other actions include the following:</p> <ul style="list-style-type: none"> <li>• Remove the individual from work immediately.</li> <li>• Determine the individual's retention function.</li> </ul>

*projections* are used to describe potential accidents in which the individual who receives the radiation dose and the damaged lights are separated by a larger distance, i.e., 100 m or more. The type of accident is irrelevant to the assumptions and methods used to project doses; that is, the assumptions for dose projections would be the same for lights broken by a road grader and those dropped from a fork lift.

Dose projections are computed with a three-step method:

1. Compute the integral air concentration for the area.
2. Multiply the integral air concentration by the breathing rate in order to estimate the total intake.
3. Estimate the dose by using the appropriate internal dose equations.

The calculations are performed with the Gaussian plume atmospheric dispersion model (Slade 1968). This model is the one most widely used for estimating airborne radionuclide exposures within 80 km of the release point and is recommended by the U.S. NRC in the regulatory guides (e.g., see NRC 1977).

In the event of an accident, the hazardous radioactivity in the lights would be contained in the volume of the tritiated water and would not be dispersed. Thus, following an accident that destroyed the integrity of the light's containment, it should be placed in a leak-proof container. Those who handle broken lights should wear gloves and wash their hands as soon as possible.

### 6.9.1 Air Concentrations in a Room

The air concentration at any time,  $t$ , following the release of radioactivity into a room can be estimated from Equation (6.27) by assuming instantaneous and complete mixing following release of activity:

$$C_t = C_o \exp(-\lambda_e t) \quad (6.27)$$

where  $C_t$  = air concentration at any time,  $\mu\text{Ci}/\text{mL}$   
 $C_o$  = original air concentration,  $\mu\text{Ci}/\text{mL}$   
 $= \frac{\text{Activity released, } \mu\text{Ci}}{\text{room air volume, mL}}$   
 $\lambda_e$  = effective removal constant,  $\text{time}^{-1}$ ,  
 where  $\lambda_e = \lambda_p + \lambda_1$  for  
 $\lambda_p$  = physical decay constant,  $\text{time}^{-1}$   
 and  
 $\lambda_1$  = exfiltration removal constant,  
 $\text{time}^{-1}$   
 $t$  = time at which the activity is to be  
 calculated.

The cumulative air concentration is the time integral of Equation (6.1):

$$\tilde{C} = \int_0^t C_o \exp(-\lambda_e t) dt \quad (6.28)$$

which is:

$$\tilde{C} = \frac{C_o}{\lambda_e} [1 - \exp(-\lambda_e t)] \quad (6.29)$$

where  $C$  = cumulative air concentration,  $(\mu\text{Ci-h})/\text{mL}$   
 $\lambda_e$  = the effective removal constant,  $\text{time}^{-1}$ ,  
 where  $\lambda_e = \lambda_p + \lambda_1$  for  
 $\lambda_p$  = physical decay constant,  $\text{time}^{-1}$   
 and  
 $\lambda_1$  = exfiltration removal constant,  
 $\text{time}^{-1}$   
 $t$  = the time at which the activity is to be  
 calculated.

If possible, the ventilation rate of the actual storage building should be used. If knowledge of the actual air turnover rates is lacking, the information in Table 6.5 can be used. The data in the table were developed for purposes that are different than the requirements of this document, namely to calculate

**Table 6.5.** Air Turnover in Buildings

Building Type	Turnover rate, h <sup>-1</sup>	Reference
Wood Frame	0.5 - 9	Johnson et al. 1973, p. 15f
Research Lab Buildings	8 - 10	Personal Communication, PNL Building Managers
Sealed Control Room	0.06	Murphy and Campe 1974
Reactor Building	0.00114	NRC 1989, p. 3.9
Glove Boxes	10-30	Robins, Bartoszek and Wordall 1984
Laboratories	1-10	Robins, Bartoszek and Wordall 1984
Reactor Halls	0.25 - 1	Robins, Bartoszek and Wordall 1984
<u>Suggested Rates</u>		
Buildings, wood frame	1 - 5	
Buildings - airtight (range)	0.015 - 0.06	
Buildings - airtight with circ	0.015	
Buildings - airtight sans circ	0.001	

releases to the environment. The tabulated values are nonetheless useful indicators of reasonable air turnover rates. Table 6.6 shows the effects of differing stay times on committed effective dose requirements.

### 6.9.2 Diffusion Equations

The diffusion equations are used to compute an integral air concentration at a particular point downwind from the release. The advantage of the integral air concentration is that we are not required to know the rate at which the radioactive material was released. The target individual is assumed to be downwind for the full duration. This feature will maximize the computed dose.

For releases of long duration (greater than 8 hours), occupancy factors may be used (NRC 1974b); occupancy factors are not discussed in this report.

Equation (6.30) is used to compute the  $\chi/Q$  value for a point source and a point receptor (NRC 1974a; NRC 1974b):

$$\chi/Q = \frac{1}{U\pi\sigma_z\sigma_y} \quad (6.30)$$

where  $\chi/Q$  = relative integral air concentration, s/m<sup>3</sup>  
 $U$  = the wind speed (source to receptor), m/s  
 $\sigma_z$  = vertical dispersion coefficient, m<sup>-1</sup>  
 $\sigma_y$  = horizontal dispersion coefficient, m<sup>-1</sup>.

The wind speed is often taken as 1 m/s (see, for example, NRC 1974a and NRC 1974b); calm winds are taken to have a wind speed of 0.5 m/s (Hanna, Briggs, and Hosker 1982, p. 25). For distances less than 100 m, the  $\sigma_z$  and  $\sigma_y$  values of Bowne (1974)

**Table 6.6.** Committed Effective Dose Equivalents From Indoor Occupational Accidents

Activity Released, Ci	Room Size, ft <sup>3</sup>	Percent Activity as HTO	Ventilation Rate, h	Stay Time, h	DE <sup>(a)</sup> , rem
50	1000	1	1	1	1.3
50	1000	1	1	4	2.0

(a) Committed effective dose equivalent.

are used. To be conservative, the least turbulent wind conditions are used. Tables 6.7 and 6.8 show  $\sigma_y$  and  $\sigma_z$  values for the Pasquill conditions for plumes in a rural area.

The quantity inhaled following a release is the product of the quantity released, the  $c/Q$ , and the breathing rate of the individual. The inhaled intake is determined by Equation (6.31):

$$\text{Intake} = (\text{BR})(\chi/Q)(\text{release}) \quad (6.31)$$

where BR = the breathing rate, m<sup>3</sup>/s  
 $\chi/Q$  = the relative integral air concentration, s/m<sup>3</sup>  
 release = the activity released.

The equation may be used twice, once for the tritiated water activity that is released and again for the elemental hydrogen that is released. The effective dose equivalent from tritiated gas is about 4 orders of magnitude less than that from tritiated water. Thus,

**Table 6.7.** Rural  $\sigma_y$  Values Used for Plume Calculations<sup>(a)</sup>

Distance, m	Pasquill Condition					
	A	B	C	D	E	F
1	1.16 m	1.17 m	1.16 m	1.19 m	1.19 m	1.17 m
2	1.68	1.60	1.49	1.39	1.34	1.29
3	2.09	1.92	1.71	1.52	1.44	1.37
4	2.44	2.19	1.89	1.62	1.51	1.42
5	2.74	2.42	2.05	1.70	1.57	1.46
6	3.03	2.62	2.18	1.77	1.62	1.50
7	3.28	2.80	2.30	1.84	1.67	1.53
8	3.52	2.98	2.41	1.89	1.71	1.56
9	3.76	3.15	2.51	1.94	1.74	1.59
10	4.05	3.44	2.71	2.05	1.77	1.61

(a) Interpolation of Figure 6 from Bowne (1974).

**Table 6.8.** Rural  $\sigma_z$  Values Used for Plume Calculations<sup>(a)</sup>

Distance, m	Pasquill Condition					
	A	B	C	D	E	F
1	1.0 m	1.0 m	1.0 m	1.0 m	1.0 m	1.0 m
2	1.40	1.20	1.18	1.17	1.15	1.13
3	1.75	1.41	1.37	1.32	1.24	1.24
4	2.08	1.67	1.48	1.45	1.33	1.33
5	2.34	1.89	1.68	1.58	1.35	1.35
6	2.57	2.07	1.77	1.70	1.43	1.37
7	2.79	2.23	1.85	1.75	1.46	1.41
8	2.95	2.36	1.94	1.83	1.51	1.45
9	3.27	2.60	2.12	1.91	1.54	1.47
10	3.36	2.74	2.24	1.94	1.58	1.51

(a) Interpolation of Figure 8 from Bowne (1974).

unless the tritiated water content is very small, it appears common to ignore the dose contribution from the elemental form.

Table 6.9 shows the effect on committed effective dose equivalents from different wind speeds.

### 6.9.3 Effect of Rain

Material may be removed from a plume by the actions of rain or snow. In general, the process is assumed to remove material uniformly throughout the vertical extent of the plume. The air concentration at any distance may be computed by using a modified source strength (IAEA 1982, Section 3.4.2):

$$f_w = \exp(-\Lambda x/u) \quad (6.32)$$

where  $\Lambda$  = washout coefficient,  $s^{-1}$   
 $x$  = the downwind distance, m;  
 $u$  = the wind speed, m/s.

There is little data on washout coefficient available. The data that exist are inconsistent; however, they are presented in Table B.3 in Appendix B. The washout coefficient ( $\Lambda$ ) is assumed to be approximately proportional to the precipitation rate. Konig et al. (1984) assumed  $\Lambda$  is proportional to the rainfall rate  $\theta$  (mm/s) or

$$\Lambda = C\theta \quad (6.33)$$

He found that  $C = 3 \cdot 10^{-9}$  a/mm·s by comparing theoretical and measured values of ground activity following precipitation events near Karlsruhe Nuclear Research Center, Germany. Davis (private communication P. A. Davis AECL to G. A. Jensen PNL, December 1991) points out that the washout coefficient  $\Lambda$  will be generally proportional to rainfall rate and will equal  $10^{-4}/s$  for  $\theta = 1$  mm/h.

**Table 6.9.** Committed Effective Dose Equivalents From Outdoor Occupational Accidents

Activity Released, Ci	Stability Class	Wind Speed	Percent Activity as HTO	Downwind Distribution, m	Duration of Inhalation	DE <sup>(a)</sup> , mrem
50	F	1 m/s	1	5	Duration of release	2.7
50	F	calm	1	5	Duration of release	5.4
50	F	1 m/s	1	10	Duration of release	2.2
50	F	calm	1	10	Duration of release	4.4

(a) Committed effective dose equivalent.

## 6.10 References

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## **7.0 Tritium Measurement and Sampling Methods**

## 7.0 Tritium Measurement and Sampling Methods

Various methods can be employed to measure tritium on surfaces and in air, water, urine, and environmental samples. These methods can be divided into two categories: monitoring and sampling.

Because of the nature of monitoring instruments used, monitoring methods often combine sample collection, preparation, and measurement to give their results in real time. A drawback of monitoring methods is that the sensitivity of monitors is not as great as that of samplers.

Sampling methods also consist of sample collection, preparation, and measurement as three distinct stages in the sampling process, each of which may be separated both spatially and temporally. However, unlike monitoring, sampling can involve collecting a sample over a long period of time, possibly concentrating the tritium content. The sample is then analyzed for tritium content. One advantage of these methods and devices is that they are able to measure low tritium concentrations because of their inherent integrating capability. Sampling methods do not require counting the sample in the environment and thus are not affected by external radiation levels. Also, most methods can distinguish between elemental tritium and tritiated water. The major disadvantage of sampling methods, compared with air monitoring, is that there will be a time lag between collection and analysis of the sample; consequently, there may be a significant time lag between the release and knowledge of the release. Also, the exact time of the release may not be known.

### 7.1 Air Measurements

Air measurements are usually performed in areas where airborne tritium is considered a distinct possibility, including manufacturing areas and regions near tritium recovery operations, such as stack monitors. The user of RL devices will probably have no need for monitoring tritium in air because permeation of tritium through glass is very slow and tritium will not build up in storage areas to any appreciable extent.

Air measurements may be taken by monitoring or sampling. Real-time monitoring for tritium may be based on air ionization or scintillators, or the air may be sampled and its tritium content determined apart from the environment in which it is being sampled.

### 7.2 Measurement of Tritium in Water

The two major methods for measuring tritium in water, one recommended by the American Public Health Association (APHA) and another by the American Society for Testing and Materials (ASTM), differ in that the APHA recommends distilling the collected water, while the ASTM recommends filtering the water. Both methods call for dark-adaptation of samples and then counting their tritium content.

### 7.3 Surface Surveys

A wipe sample (a "swipe") is taken of a surface using moist filter paper. The tritium concentration is then found with a gas-flow proportional counter or liquid scintillation counter.

## 7.4 Bioassay

The bioassay method most commonly employed for tritium analysis is assay of the tritium content of urine. The tritium concentration in urine may be counted directly, by placing a sample in a liquid scintillation counter, or by distilling the urine, collecting the water, and counting the distilled water in a liquid scintillation counter.

## 7.5 Counting Tritium in Environmental Samples

Of greatest interest is the amount of tritiated water present in a sample of soil, animal tissue, or plants. A common method of determining tritium content is by azeotropic distillation with toluene.

## 7.6 Air Monitoring Systems

In the following discussion, "memory effects" will be mentioned. Memory effects means that the radioactive material may interact in some way with, and be retained by, the detector. The detector may respond to tritium that is retained, thus providing a response even though the air currently being monitored contains no tritium. (The detector "remembers" the tritium activity in the previous air.)

### 7.6.1 Air Ionization Chambers

Air ionization chambers collect the ions that are produced by radioactive materials in the air. The chamber consists of an anode and a cathode, which have a sufficient potential difference to prevent recombination of the ions and collect the ions at the electrodes; no multiplication of ions takes place. The sample to be measured is drawn into the chamber by a partial vacuum caused by a pump at the exit of the device.

Two different types of air ionization chambers are used for monitoring tritium in air. Kanne chambers are large ionization chambers reported to have active volumes of 18.5 to 51.5 L (Kelley and Phillips 1973). External factors such as cigarette smoke, aerosols and ions, moisture and condensation, ambient gamma-ray field changes, radon, clogged filters, fission product gases, absorption, and memory effects will all cause errors in the readings from the chambers. To reduce the effect of these external factors, a filter and ion trap are commonly placed at the input. Gamma

compensation, using a similar but closed chamber that negates response to external gamma radiation is also used. Reported detection levels for Kanne chambers range from 2 to 8 pCi/mL (Kelley and Phillips 1973).

Smaller ionization chambers are also used. Commercially available instruments use ionization chambers for real-time measurements of tritium in air. One such device has a sensitive volume of 2.6 L.

### 7.6.2 Proportional Counters

Proportional counters may be used to measure tritium in air. Proportional counters are similar to ionization chambers; however, proportional counters operate at higher voltages so that gas multiplication will occur. The devices operate in a pulse mode. Proportional counters require that the air be mixed with a counting gas (methane or P-10 gas) before it enters into the sensitive region of the counter. An advantage of the proportional counter is that the proportional response of the detector allows the use of electronic circuitry that can distinguish between tritium and other radionuclides. A disadvantage of the proportional counter is that sampling at high rates requires large amounts of the counting gas. Some proportional counters separate the air (containing the tritium) from the counting region by a thin metal-covered plastic membrane; the beta particle of the tritium penetrates the membrane and is counted, but mixing of air and counting gas does not occur. Tritium monitors based on proportional counters are commercially available.

### 7.6.3 Scintillator, Photomultiplier Tube

Plastic scintillators can be used to monitor air and water streams that contain tritium. Absorption of radiation energy by a plastic scintillator causes the scintillator to emit a flash of light that is proportional in intensity to the energy deposited. A photomultiplier tube and associated electronic circuitry quantifies the light output. The plastic scintillator can be in the form of rods or sheets. An advantage of the plastic scintillator is that different radionuclides can be distinguished based on the energy deposited in the scintillator. A disadvantage of this type of detector is related to memory effects.

### 7.6.4 Air Sampling Systems

Tritium air sampling devices may be partitioned into three major groups: 1) methods that collect all the tritium in air and do not distinguish between tritium in its elemental and its oxidized states, 2) methods that collect only the airborne tritiated water (with and without mechanical devices), and 3) methods that distinguish between elemental and oxidized tritium. These three groups are discussed in the following sections, along with the need for air sampler calibration.

**Nondistinguishing Methods.** The method for sampling tritium in air described by the American Society for Testing and Materials (ASTM method D 3442-75) does not distinguish between tritium in the form of water or elemental gas (ASTM 1981). Essentially, the method consists of drawing air at a rate of 100 to 500 cm<sup>3</sup>/min through a tube furnace. The tube furnace is packed with either palladium catalyst heated to 500°C or copper oxide heated to 600° to 650°C. The heated catalyst converts the elemental tritium to tritiated water. The air is then passed over silica gel, which absorbs the water in the air. After collection (the recommended collection time is 1 week), the silica gel is heated for 24 hours at 300°C. The heating drives off the water, which is collected and then counted with a liquid scintillation counter (Method D 2476 in ASTM 1982). If the air

that will be collected contains radioiodine and radioactive particulate material, the air is filtered through a silver zeolite cartridge before it enters the tube furnace.

**Methods That Collect Only Water.** An air sampling system that collects only airborne tritiated water draws air through an apparatus that removes the water vapor. The water is then collected and counted, often by means of a liquid scintillation counter. The observed activity is then divided by the volume of the air that yielded the counted sample to give the concentration of tritiated water in the air. The results may be recorded in units of µCi/mL of air and compared with regulatory limits, such as those in 10 CFR 20.

The water may be removed from the air by one of several means described here:

1. *Passing the air through a desiccant.* The air may be drawn through columns of silica gel (IAEA 1979; McLain and Shamblin 1985; McLain and Lee 1987), a molecular sieve (alumino-silicates), calcium sulphate, or activated alumina (IAEA 1979). The tritiated water may be quantified following distillation of the silica gel (McLain and Shamblin 1985) or by directly counting the silica gel (McLain and Lee 1987). The collection efficiency may approach 99% (IAEA 1979).

The collected water is then removed from the desiccant. For the molecular sieve, the removal method consists of heating the molecular sieve in a distillation apparatus. The water that is driven off is collected and counted in a liquid scintillation counter. If the desiccant is silica gel, it is immersed in a known amount of water, and after equilibrium has been achieved (approximately 40 minutes), a water sample is taken for counting by liquid scintillation counting (IAEA 1979).

2. *Condensing the water.* The air may be passed through a series of cold traps that will freeze out the water vapor. The cold traps may be cooled

by liquid nitrogen (IAEA 1979; Iyengar et al. 1965), alcohol and dry ice (Chiswell and Dancer 1969), electrically cooled baths, or some other means. After an appropriate collection time, the water is thawed and counted by liquid scintillation counting. Care must be taken to avoid an ice buildup in the cold trap because the ice could cause a reduction in the air flow of the detector (IAEA 1979).

3. *Drawing the air through an absorbing liquid.* Air may be drawn through a series of gas washing bottles that contain distilled water (IAEA 1979) or ethylene glycol. A sample of the absorbing liquid is then counted in a liquid scintillation counter. For tritium gas, this method has a low efficiency, but it is appreciably more efficient for tritiated water vapor. If the HTO/H<sub>2</sub>O in the absorbing water is low compared to that in the incoming air, collection efficiencies of 90% to 98% can be achieved (IAEA 1979). This method has a drawback in that the tritium in the water will be diluted by the volume of water in the collection bottle.

### Measuring Tritium Concentration

**Without Mechanical Devices.** The tritium concentration of tritiated water vapor may also be measured without mechanical devices. These methods remove the moisture from the air using silica gel and "cold fingers" (metal strips partially inserted into a container filled with dry ice or liquid hydrogen). Because no mechanical device is employed to measure air volumes, the relative humidity of the air must be known in order to determine the air concentration of the tritiated water. With the cold fingers technique, water condenses on the metal strip. When the cooling media have been exhausted, the water will melt and run into the container.

The amount of water vapor in the air may be determined by measuring the relative humidity of air and then multiplying the relative humidity (as a fraction) by the saturation content of water in air at the air temperature. The relative humidity may be measured by measuring the dew point (Graham and Roselius

1986) or by using relative humidity meters based on the resistance or capacitance of hydrophilic polymer membranes (McLain and Shamblin 1985). Other methods are based on the use of wet and dry bulb thermometers and on the conduction of hygroscopic salts. Devices for measuring relative humidity are readily available from laboratory supply houses.

### Methods That Distinguish Between Elemental and Oxidized Tritium.

Air sampling methods that distinguish between tritiated water and elemental tritium can be generically described as beginning with drawing the tritium-laden air into a device. Once in the device, the water in the air is removed and collected using one of the methods described in the previous section on "water only" collection. The water-free air is then treated to convert the elemental hydrogen in the air to water, which is then removed from the air stream. The method employed to oxidize the elemental tritium is usually combustion. The two different waters are then counted for their tritium content. The tritium air concentration is estimated by dividing the tritium activity by the total air volume that passed through the collection device.

Tritium in the form of tritiated water may be selectively removed from air in many different ways. Some methods can directly yield the tritiated water content of the air; others will yield only the specific concentration of tritium in water vapor of the air. In the latter methods, the relative humidity must be known in order to estimate the specific concentration of tritium in air.

### 7.6.5 Air Volume Determination

When the air volume of the sample must be known, the air samplers should be calibrated (IAEA 1979). Calibration standards are divided into two categories: primary and secondary. Primary standards for air flow are a) the spirometer, b) the Mariotte bottle, and c) the bubble meter. Secondary standards include a) dry gas meters, b) wet test meters, c) precision rotameters, d) orifice meters, and e) Venturi meters. The IAEA (1979) recommends that all field

instruments be calibrated before use and at 3-to-6-month intervals thereafter.

## 7.7 Water Monitoring/Sampling

Three standard compendia describe the methods for measuring the tritium activity in water. The American Public Health Association (APHA 1985) recommends that the collected water be distilled to remove interfering radionuclides and materials that might interfere with the liquid scintillation process. The ASTM (1982) does not require distillation of water; however, it recommends that if the samples or scintillator stock solutions were exposed to daylight or fluorescent lighting, they be dark-adapted for a minimum of 2 hours.

In HASL-300 (Volchok and de Planque 1983), two methods are described (E-H-01 and E-H-02) for determination of tritium content in water; both methods use electrolysis to concentrate the tritium in the water. This method is used primarily for environmental samples, especially those environmental samples that are expected to contain tritium at or near normal quantities. The APHA and ASTM methods for quantifying the tritium content of water are described briefly in the following subsections. In most cases, the source of the water is immaterial; it may be water collected in an air sampler, urine from a person who may have been exposed to tritium, or water from an environmental sample.

### 7.7.1 APHA Method

Using the APHA method to quantify tritium in water, the water sample (a minimum 100 mL) is treated with alkaline permanganate distillation to hold back quenching and chemoluminescent materials (APHA 1985). A 4-mL sample of the distilled water is then pipetted into a vial for liquid scintillation counting, and 16 mL of a dioxane-based counting cocktail (or its commercial equivalent) is added. The vials are dark-adapted for 3 hours and then counted. Samples that contain more than 200 pCi/mL are counted for

50 minutes; those that contain less than 200 pCi/mL are counted for 100 minutes.

### 7.7.2 ASTM Method

The method described by the ASTM is similar to the APHA method (ASTM 1982). The ASTM method suggests that turbid water may be filtered through a membrane filter. Distillation is suggested if radioactive materials other than tritium are present; redistillation of the sample may be required in some cases. The ASTM method is silent on the addition of material to the water sample (such as to make an alkaline permanganate solution) prior to distillation. The method calls for counting a 10-mL sample of the water with 10 mL of scintillator. The recommended time for dark adaptation is 2 hours.

## 7.8 Surface Surveys

The most common method for estimating the activity on a surface is to take a wipe sample (often called a swipe) and count the sample at a later time. The wipe sample is taken with a filter paper; moistening the swipe with water or other liquid increases the sampling efficiency for tritium. The activity on the wipe sample is then quantified by counting the swipe in a gas-flow proportional counter or in a liquid scintillation counter.

## 7.9 Bioassay

Urine samples from persons suspected of receiving a tritium dose are collected in clean containers, preferably disposable plastic containers. Before the individual collects the sample, he should wash his hands in order to eliminate any tritium contamination that may be on them. The container is equipped with a tightly fitting cover to prevent evaporation of the sample. If the sample will not be analyzed for some time, a preservative, such as an aqueous solution of ethyl(2-mercaptobenzoato-S)mercury sodium salt (thimerosal) may be added (Moghissi and Lieberman 1970).

There are two major methods for counting tritium in urine: direct counting of urine using liquid scintillation counting and distilling the urine before counting. These methods are discussed in the following subsections, along with mention of a method for determining the content of tritium in organic materials. The tritium content of urine is best estimated by means of a liquid scintillation counter.

### 7.9.1 Direct Counting of Urine

An aliquot of the urine is pipetted directly into a vial for liquid scintillation counting, scintillator solution is added, and the mixture is counted. (This method is described by Gautier [1983] as method R230-1 and by the ASTM [1982] as method D2476-81.) The advantages of this method are that the preparation time is minimal and no sample preparation equipment is required. The disadvantages are that the color inherent to urine may cause color quenching of the sample and a quench correction curve must be employed. (Correction for color quenching requires a different curve than does chemical quenching.)

There may also be some chemical quenching, and chemiluminescence can be also a problem. The classic method for eliminating the problem of chemiluminescence is to store the urine samples in the dark for a few hours and then transfer the samples to the counter in a darkened room. A small amount of  $\text{SnCl}_2$  solution (freshly prepared, saturated) will eliminate chemiluminescence.

### 7.9.2 Distillation of Urine

The second major method for assaying the tritium activity in urine is to distill the urine, collect the water, and count the activity in the water using liquid scintillation counting. (This is the method described by Gautier [1983] as method R240-1 and the method described by the APHA [1985].) This method eliminates the problems associated with quenching and chemiluminescence; although water will cause a dilution quench, it can be easily corrected and should be fairly constant for all samples. An additional advantage is that interfering radionuclides will be removed from the water. The disadvantage of this

method is that there is a considerable preparation time, and the preparation equipment must be cleaned. This method will require more time for analysis because of the preparation time, but there will be fewer interferences with the counting process.

### 7.9.3 Determination of Tritium in Organic Materials

In HASL-300 (Volchok and de Planque 1983), a method (E-H-03) for determining the tritium content of organic materials is described. The method employs a combustion apparatus that is described in the text.

## 7.10 Determination of Tritium in Environmental Samples

Tritiated water may be recovered from samples in soil, tissue, and plants by azeotropic distillation with toluene (see, for example, Brown and Workman 1986). The method is relatively rapid and may be performed with little attention. The material to be analyzed is placed in a distillation flask with toluene. A distillation trap, similar to those employed for determining the moisture content of petroleum products, is used to collect the water. Distillation of the mixture is allowed to continue until all water has been removed from the sample (toluene-water will come off at about  $85^\circ\text{C}$  and the temperature rises to about  $110^\circ\text{C}$  after the water is removed). The water is removed from the collection device and counted in a liquid scintillation counter. A disadvantage of this method is that the dried organic material must be freed of toluene before the tritium content of the organic constituents may be determined.

The following steps describe the procedures that should be followed to obtain environmental samples, especially those samples that are collected after an accident. The material presented here is condensed from NUREG/CR-5212 (Stoetzel and Lynch 1988), to which the reader is referred for further information.

1. *Field Survey Techniques.* Field teams must be able to document accurately the positions where environmental samples have been taken. Without this information, the data obtained are much less valuable. In the case of the locations for semipermanent devices (such as a runway), a map of the area should be drawn and kept for use in emergencies. The map should contain locations of prominent landmarks. Grid patterns may be laid over the map to help assure a systematic approach for locating environmental samples.

During the sampling, survey teams should proceed from less contaminated areas to the more highly contaminated areas while collecting the samples to minimize the possibility of contaminating samples. The location of each sampling location should be clearly indicated on the map.

2. *Sample Collection Procedures.* The environmental sampling should collect vegetation, water, snow, and possibly soil samples. The samples should be collected in glass containers, if possible. Tritium can diffuse through plastics and a single, highly contaminated sample may contaminate all other samples that are packed together.

For soil samples, only the uppermost layers of soil need be sampled. Soil may be collected because it is within the upper layers of the soil that tritium gas may be converted to tritiated water by microorganisms. An acceptable method is to collect soil samples from a 1-ft<sup>2</sup> area down to a depth of approximately 1/2 in. Samples are collected using a shovel or a trowel and then they are placed in a glass container. Another method is to collect soil samples from an area approximately 5 yards on a side. Within this area, five separate samples, each about 4 in. on a side are collected and analyzed as a single sample.

For vegetation sampling, samples are commonly obtained by clipping the vegetation close to the soil. The amount of soil collected with the vegetation should be minimized. Samples

typically weigh 1 to 2 lb (wet weight) and are taken from a unit area of from 1 to 9 ft<sup>2</sup>. Some general considerations for collecting vegetation samples include the following:

- Samples are clipped as close to the surface as possible without getting roots and soil in the sample.
  - For leafy vegetation samples, only the leaf portion is collected, leaving the stems and roots behind.
  - Snow samples should be collected from an area of approximately 1 yd<sup>2</sup> to a depth of about 1/2 in. This will result in a water sample of approximately 1 quart, which will be adequate for analysis. If snow was falling during the release, the fresh snow may contain the highest concentration of tritium. If snow has fallen after the release, field teams must sample deep enough to collect the contaminated layer. Another concern during snow sampling is drifting snow. If possible, snow samples should be collected in areas that are not prone to drifting.
  - Water sampling done by field teams would typically include water samples from any streams, rivers, ponds, drainage ditches, or standing water locations in the area affected by the release. Such surface water samples would be collected by dipping sample containers in the water until full. If the body of water being sampled is not deep enough to use the sample container, a dipper is used to pour the sample into the container.
3. *Recordkeeping.* Good records should be maintained from the onset of an emergency. Field teams should keep a detailed log of all samples collected. The log should include information such as where and when the sample is collected. Preprinted data sheets provide an excellent format and should be available to field teams.

Recordkeeping also involves careful and complete labeling of the samples. Again, preprinted forms (labels) are recommended. The labels should contain information such as the names of the team members collecting the sample, sample location, sample type, and sample size. The label should be filled out with a water-proof pen or indelible pencil.

## 7.11 References

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## **8.0 Emergency Planning**

## 8.0 Emergency Planning

Emergency planning is a necessary requirement for the use of RL lights as runway markers and ship exit signs. Radiological contingency plans will assist in assuring that the following conditions exist at radioactive material facilities:

- Facilities are properly configured to limit releases of radioactive materials and radiation exposures in the event of an accident.
- Capability is provided for measuring and assessing the significance of the release.
- Appropriate equipment and procedures are provided onsite to protect workers against radiation hazards during an accident.
- Notifications are made promptly to Federal, state, and local government agencies in an emergency.
- Recovery actions are taken in a timely fashion to return the facility to a safe condition following an accident.

The plans should contain descriptions of the following: the plant, engineered safety features for abnormal events, emergency classes, emergency organization, radiological contingency measures, equipment and facilities, maintenance of emergency preparedness, records and reports, and recovery actions.

### 8.1 General Emergency Planning for Runway Lights

For users of RL runway lights, a caretaker should check the lights periodically for breakage. The caretaker should be trained in basic radiation safety, including the use of radiation detection equipment, and initial emergency actions required to stabilize an accident situation until other radiation safety assistance arrives. The caretaker should also have current radiation safety contacts available in case the lights are broken. Caretakers should have emergency response kits that include radiation detection instruments, bioassay kits, rubber gloves, protective clothing, and other equipment needed to contain an area in which RL lights have been broken. Caretakers should have written emergency procedures to follow in case of an accident.

Wherever RL lights (such as runway markers) are stored or used, a caretaker should periodically check the physical integrity of the lights. The caretaker should be trained in basic radiation safety, as well as in how to determine if the RL device tubes are intact, how to use radiation detection equipment to determine if the tubes are leaking, who to contact in case of tube breaches or accidents involving the RL lights, and which initial emergency actions are required after an accident until assistance arrives.

Although tube breakage would be obvious, a small crack and subsequent leak in a tube would be more difficult to detect. The first symptom would probably be dimming of the light. If a caretaker finds either a tube break or leak, he should move upwind of the panel containing the affected tube and notify his supervisor. The supervisor should then obtain radiological assistance (e.g., notify the radiation protection officer [RPO]). If the runway is located on a military base, it is possible that an RPO could be dispatched to the location to handle the situation. For a remote runway, the RPO would instruct the caretaker on how to handle the broken unit. The unit should be placed in a

plastic bag and put in a specially built aluminum box for shipment to the manufacturer or a disposal site. Rubber gloves should be worn when handling broken or leaking RL tubes. After handling the panel, the caretaker should give a urine sample for bioassay.

For more severe accidents, such as a fire in an RL panel storage area, caretakers must be trained in initial actions to take. A list of initial actions is presented below:

1. Perform lifesaving rescues and emergency first aid.
2. Secure the area, keeping all unauthorized personnel away from the affected area. All authorized personnel should be kept upwind of the affected area.
3. Notify any offsite support agencies, such as the fire department or ambulance, if necessary. Injured personnel who may be contaminated should be taken to the nearest hospital. Inform ambulance and hospital personnel of the potential for contamination. If the hospital has a radiation safety officer (RSO), the RSO would be responsible for assuring that the ambulance and ambulance attendants are free of contamination. If the hospital does not have an RSO, assistance would have to be obtained from a state or Federal support agency.
4. Obtain assistance in radiological support. If the accident involves runway markers on a military base, the base RPO should be notified. If the accident occurs on a runway at a remote location, the caretaker should be provided with the phone number of individuals who could provide this support. Such individuals could be from state agencies or the Federal government.
5. Do not attempt to move or clean up any of the radioactive material involved in the accident.
6. Obtain the names of all persons involved in the accident. Consider collecting urine samples from the individuals involved. Potentially contaminated individuals should not be allowed to leave the accident scene until cleared by a qualified health physicist.

In addition to obtaining assistance in the radiological support area, the caretaker or health physicist (e.g., RSO) should also notify the regulatory authority responsible for licensing the RL lights. This could be an Agreement State, the NRC, or the DOD. Caretakers or health physicists should be prepared to provide the following information to the regulatory agency:

- their names
- who they are representing
- location of the accident
- approximate area affected by the accident
- radioactive material involved and approximate amount
- information on the type of accident

- time of accident
- status of accident--whether the accident scene has been stabilized
- any persons injured and status of injured personnel, whether or not any workers or members of the public were affected by the release, and any emergency response crews (e.g., fire, ambulance, police) that have provided assistance at the accident scene.

Caretakers for remote runways should be provided with emergency response kits. Kits should contain radiation detection instrumentation, bioassay kits for approximately six personnel, a supply of plastic gloves, several sets of protective clothing, rope to use in roping off an area, and plastic bags in which to place contaminated objects. The caretakers would also benefit from having a written emergency procedure that contains information such as:

- names and phone numbers of key individuals to contact for assistance in an emergency (radiological assistance, regulatory agencies, ambulance services, local fire departments, and hospitals)
- guidance on when to collect bioassay samples.

## **8.2 General Emergency Planning for Exit Signs**

For users of RL exit signs, no formal emergency procedures are needed except that in case of a broken sign, the area should be restricted and a health physicist called to check for contamination.

Emergency response for users of exit signs follows the practice of common sense. If the user suspects that a sign has been breached, and (e.g., sign drops to floor from an overhead location), the area around the sign should be restricted until a health physicist is summoned to check for contamination. If the exit sign is in a remote location (e.g., on a ship at sea), then the user should again try to isolate the area and observe if there is any loss of luminescence.

## **8.3 General Accident Prevention Methods and Procedures**

Users of RL lights should have a safety program identifying good practices when handling the lights. Even though RL lights are required to meet or exceed ANSI N540-1975 (ANSI 1975) standards for self-luminous light sources, the lights should not be treated roughly or placed on a stove or heated surface. The safety plan should establish an annual testing for all signs for possible tritium leakage. Good housekeeping practices should be followed in light-storage areas.

## **8.4 General Emergency Planning for Manufacturers of RL Lights**

For manufacturers of RL lights, an emergency plan must include the following information: facility description, types of accidents, classification of accidents, detection of accidents, mitigation of accident consequences, assessment of releases, responsibilities of emergency personnel, notifications and coordination, information to be communicated, training, safe shutdown, exercises and audits, and hazardous chemicals. Facilities must also have the capability to implement their emergency plans; written procedures are normally kept and updated for this purpose. Emergency planning should include having established emergency response facilities (from which actions can be

coordinated during an emergency) and multiple locations for key equipment, in case primary locations are not accessible during an ongoing accident.

## 8.5 Nuclear Regulatory Commission Requirements and Guidance

Current requirements and guidance for emergency planning at facilities that receive or possess by-product radioactive material can be found in 10 CFR 30.32.

The NRC requires that each application to possess quantities of radioactive material in excess of those quantities identified in 10 CFR 32.72, Schedule C, "Quantities of Radioactive Materials Requiring Consideration of the Need for an Emergency Plan for Responding to a Release," must contain either:

- an evaluation showing that the maximum dose to a person offsite due to a release of radioactive materials would not exceed 1 rem effective dose equivalent or 5 rem to the thyroid, or
- an emergency plan for responding to a release of radioactive material.

Factors that may be used to support an evaluation showing that the above doses are not exceeded (10 CFR 30.32 (g)(2)) include the following:

- The radioactive material is physically separated so that only a portion could be involved in an accident.
- All or part of the radioactive material is not subject to release during an accident because of the way it is packaged.
- The release fraction in the respirable size range would be lower than the release fraction shown in 10 CFR 30.72, Appendix C, due to the chemical and physical form of the material.

- The solubility of the radioactive material would reduce the dose received.
- Facility design or engineered safety features in the facility would cause the release fraction to be lower than shown in 10 CFR 30.72, Appendix C.
- Operating restrictions or procedures would prevent a release fraction as large as that shown in 10 CFR 30.72, Appendix C.
- Other mitigating factors appropriate for the specific facility are present.

Emergency plans for responding to a release of radioactive material must include: facility description, potential types of accidents, classifications of accidents, detection of accidents, mitigation of consequences of accidents, assessment of releases (not real-time assessment but pre-determined actions based on projected accidents), organizational responsibilities, notification and coordination, information to be communicated, emergency training, recovery, exercises and audits, and hazardous chemicals.

Quantities of radioactive materials at facilities requiring consideration of an emergency plan are found in the 10 CFR 30.72, Appendix C. For RL light manufacturers or users, more than 20,000 Ci for  $^3\text{H}$ ,  $6 \cdot 10^6$  Ci for  $^{85}\text{Kr}$ , 4000 Ci for  $^{147}\text{Pm}$ , and 100 Ci for  $^{226}\text{Ra}$  would require the consideration of an emergency plan. As discussed, licensees have the option of either showing that potential accidents at their facility could not exceed the dose guidelines discussed above or reducing their radioactive material inventory so they would not exceed the quantities identified above. Either situation would exempt the licensee from providing and maintaining an emergency plan; however, the licensee still may be required to have and maintain a radiological contingency plan.

NUREG-1140 (NRC 1985) identifies the classes of fuel cycle and other radioactive material licensees that could have accidents that might result in radiation doses to the public exceeding EPA protective action guides (EPA 1980). This analysis was based on past accident histories and offsite dose calculations based on worst-case type of accidents at the facilities. It was used as the basis for determining which facilities would be required to submit and maintain an emergency plan.

The offsite dose calculations in NUREG-1140 were calculated by assuming release fractions for radioactive materials, assuming a standard Gaussian plume model, and calculating doses from three pathways: inhalation, external dose from the passing cloud, and dose from ground deposition. Effective dose equivalent and child thyroid dose were calculated. The following assumptions were used in performing the dose calculations:

- 8-hour ground exposure time and a shielding factor of 0.7
- two sets of meteorological conditions: F stability class with 1 m/s wind speed, and D stability class with 4.5 m/s wind speed
- intercept fraction for inhalation  $1 \cdot 10^{-6}$
- plume centerline doses calculated
- breathing rate of  $2.66 \cdot 10^{-4} \text{ m}^3/\text{s}$
- CRAC2 computer code (Ritchie et al. 1983) for the calculations
- exposed individuals in a fire assumed to be on the plume centerline for 30 minutes
- doses calculated at 100 m from the release point
- noble gases assigned a release fraction of 1.0, tritium a release fraction of 0.5,  $^{147}\text{Pm}$  a release fraction of 0.01, and  $^{226}\text{Ra}$  a release fraction of 0.001.

Calculations for a by-product facility with a maximum licensed possession limit of 150,000 Ci of tritium showed an effective dose equivalent range of 0.1 to 10 rem. The upper dose is the worst-case situation assuming worst-case meteorology. The lower dose is the upper dose divided by 100. Calculations for a by-product facility with a maximum licensed possession limit of 10,000 Ci of  $^{85}\text{Kr}$  showed an effective dose equivalent range of 0 to 0.002 rem. Similar calculations for a facility with a maximum licensed possession limit of 3500 Ci of  $^{147}\text{Pm}$  showed an effective dose equivalent range of 0.008 to 0.8 rem. Only the by-product facility handling 150,000 Ci of tritium would have a potential of exceeding the EPA Protective Action Guidelines and thus require an emergency plan.

## 8.6 Emergency Planning Requirements for RL Lights

Emergency planning requirements for RL lights can be divided into the requirements for the facility that manufactures and initially transfers the lights and the requirements for the users of the lights. The following discussion concentrates on the requirements for the user.

### 8.6.1 Manufacturers

The manufacturers of RL lights need to comply with 10 CFR 30.32. For example, a manufacturer of RL lights having a maximum licensed possession limit of 100,000 Ci of tritium would possess a quantity of tritium which exceeds the 20,000-Ci possession limit in 10 CFR 30.72. Therefore, a radiological contingency plan would be required and the manufacturer would be required to evaluate the need for an emergency plan based on exceeding an inventory of 20,000 Ci of tritium. Unless the licensee can show that doses to the public would not exceed the lower end of the EPA Protective Action Guidelines for potential accidents, an emergency plan would need to be provided and maintained.

## 8.6.2 Users

As mentioned, 10 CFR 30.32 (g)(1) states that each application to possess radioactive material sealed in glass in excess of the quantities in 10 CFR 30.72, Schedule C, must contain either an evaluation showing that the maximum dose to a person offsite due to a release of radioactive material under reasonable and plausible circumstances would not exceed 1 rem effective dose equivalent or 5 rem to the thyroid, or an emergency plan for responding to a release of radioactive material. Schedule C (entitled "Quantities of Radioactive Materials Requiring Consideration of the Need for an Emergency Plan for Responding to a Release") lists a value of 20,000 Ci for tritium,  $6 \cdot 10^6$  Ci for  $^{85}\text{Kr}$ , and 4000 Ci for  $^{147}\text{Pm}$ . The same seven factors presented earlier from Section 30.32 (g)(2) are to be used in supporting an evaluation to show that the lower limit of EPA Protective Action Guidelines would not be exceeded:

1. The radioactive material is physically separated so that only a portion could be involved in an accident.
2. All or part of the radioactive material is not subject to release during an accident because of the way it is stored or packaged.
3. The release fraction in the respirable size range would be lower than the release used in calculating 10 CFR 30.72, Schedule C, due to the chemical or physical form of the material.
4. Solubility of the radioactive material would reduce the dose received.
5. Facility design or engineered safety features in the facility would cause the release fraction to be lower than in 10 CFR 30.72, Schedule C.
6. Operating restrictions or procedures would prevent a release fraction as large as that shown in 10 CFR 30.72, Schedule C.

These factors apply to users of RL lights. Therefore, users would have to apply to the NRC for an exemption. Users should be able to obtain an exemption by showing that the maximum dose to an offsite person could not exceed 1 rem effective dose equivalent. Appendix B of this report presents a review of dose projections for accident scenarios with none of the scenarios resulting in offsite doses above 1 rem. Thus, it does not appear that there would be any requirement for providing and maintaining an emergency plan for RL runway markers.

Runway markers consist of multiple  $^{166}\text{Ci}$  tritium sources, each sealed in a glass tube. The USAF lights developed through Oak Ridge consist of six  $^{166}\text{Ci}$  tubes per panel assembly (996 Ci). Each panel assembly is 15 in. long by 15 in. wide by 5-1/2 in. high and is sealed in a Lexan cover. Two of the panel assemblies then comprise a rack (1992 Ci), and approximately 132 racks (255,000 Ci) would be needed for a runway.

## 8.7 Developing an Emergency Plan

The following discussion assumes that the manufacturer is required to submit and maintain an emergency plan in accordance with the 10 CFR 30.32 (3). Although there probably will be no specific NRC requirements for an emergency plan for users of RL lights, it is prudent for users to give some thought to how they would respond in an emergency. Good practices that should be followed by RL light users are discussed.

### 8.7.1 Emergency Plan for the Manufacturer

Guidance to licensees on the preparation of emergency plans is provided in 10 CFR 30.32.

It is clear that facilities must have the capability to implement any developed emergency plans. Procedures for implementing an emergency plan are typically written and maintained for this purpose. Procedures contain more in-depth information on

how to perform specific emergency actions. For example, implementation procedures may be written for emergency classification, notification of offsite agencies and onsite emergency personnel, emergency monitoring and sampling, activation of emergency facilities, personnel accountability, onsite evacuation, handling of medical emergencies, search and rescue operations, training, exercises, and maintenance of emergency equipment.

Facilities must also have established emergency response facilities from which response actions can be coordinated during an emergency. Title 10 CFR, Part 30.32 does not discuss emergency response facilities in any detail other than to say that a control point must be established from which offsite notifications can be made. This control point could also be the location for controlling mitigating actions. Key members of the onsite emergency organization should be located at the control point. Provisions should also be provided for members of regulatory agencies such as the NRC or Agreement State officials. The control point should be located in an area that will be habitable during the postulated accidents at the facility. If habitability is a concern during certain accidents, a backup location should be identified in the emergency plan. The licensee should also identify a location near the plant where a media briefing could be held.

The facility should also have multiple locations for key emergency equipment, in case primary locations are not accessible because of accident conditions. This was a lesson learned from the UF<sub>6</sub> cylinder rupture accident at the Sequoyah Fuels Corporation conversion facility in Gore, Oklahoma, as discussed in NUREG-1198 (NRC 1986).

### **8.7.2 Emergency Plan for the User**

Generally, there should be no requirement for users of RL lights to have an emergency plan because users should be able to meet one or more of the exemptions discussed in the 10 CFR 30.32. However, if an accident occurs (e.g., a fire in a storage building), the user needs to have preplanned actions to follow. The

extent of the preplanned actions will depend on the type and quantity of RL lights a user possesses.

### **8.7.3 Emergency Plan for Users of Runway Markers**

Emergency planning considerations for users of runway markers depends on the location of the runway. Personnel at a runway located in a remote area in Alaska will respond to accidents in a different manner than if the runway were located on a military base near a populated area. Health physics and other support personnel (such as fire fighters and medical teams) would be able to respond more quickly if the runway were located near a populated area. Designated RL lights caretakers would have a key role in the initial emergency response at remote runway locations.

Caretakers' basic radiation safety training should include information on the amount of radioactive material in the lights, the type of radiation emitted by the material in the lights, the health impact of the exposure to the material if the tubes are breached, and the radiation detection equipment that can be used to measure radioactive material released. Information should be provided on how to use the detection equipment and what concentrations of tritium or other RL material are of health concern. The training could be performed by the manufacturer. The material listed above could be covered in a 2-hour to 4-hour course.

For initial emergency actions, training should cover the full range of emergency situations, from the most severe (involving a fire in a RL light storage area or a plane crash into runway panels) to the breakage or leakage of a single RL tube.

## **8.8 References**

American National Standards Institute (ANSI). 1975. *Classification of Radioactive Self-Luminous Light Sources*. ANSI N540-1975, American National Standards Institute, New York.

Ritchie, L. T., et al. 1983. *CRAC2 Model Description*. NUREG/CR-2522, U.S. Nuclear Regulatory Commission, Washington, D.C.

U.S. Environmental Protection Agency (EPA). 1980. *Manual of Protective Action Guides and Protective Actions for Nuclear Incidents*. EPA-520/1-75-001, Office of Radiation Programs, Washington, D.C.

U.S. Nuclear Regulatory Commission (NRC). 1985. *A Regulatory Analysis on Emergency Preparedness for Fuel Cycle and Other Radioactive Material Licensees-Draft Report for Comment*. NUREG-1140, Office of Nuclear Regulatory Research, Washington, D.C.

## **9.0 U.S. Government Safety Requirements for Tritium RL Devices**

## 9.0 U.S. Government Safety Requirements for Tritium RL Devices

Tritium,  $^{85}\text{Kr}$ , and  $^{147}\text{Pm}$  are by-product materials that may be used in radioluminescent lights. By-product materials are defined as any material made radioactive by exposure to the radiation associated with a given process or by use of special nuclear material. Domestic licensing requirements for by-product material are found in Title 10 Parts 30, 31 and 32 of the U.S. Code of Federal Regulations (1993d, e, and f). Depending on the amount and type of radioluminescent material, requirements range from nothing (exempt) to a license (either a general or a specific license).

### 9.1 Nuclear Regulatory Commission Requirements for General Licenses for RL Lights

A general license can be issued by the U.S. Nuclear Regulatory Commission (NRC) to private industry, research facilities, educational and medical facilities, and Federal, state, or local government agencies for the receipt, possession, or use of by-product material contained in devices for the purpose of producing light. The general license requires that the licensee shall do the following:

- assure that all labels affixed to the device are not removed
- test each device for leakage of radioactive materials at no longer than 6-month intervals (except for devices containing krypton or tritium, which need not be tested for any purpose)
- immediately suspend operation when the device is damaged or 0.005  $\mu\text{Ci}$  or more of removable radioactive material is detected
- maintain control of the device (that is, do not abandon it)
- dispose of the device through the holder of a specific license
- transfer the device to another general licensee only when the device remains in use at a particular location or when the device is held in storage in the original shipping container at its intended location of use.

### 9.2 Nuclear Regulatory Commission General License for Aircraft Lights

A general license can be obtained for using luminous safety devices in aircraft. Regulations require that each device contain not more than 10 Ci of tritium or 300 mCi of  $^{147}\text{Pm}$ . The licensing requirements for using RL devices in aircraft are less stringent than the requirements for RL light devices used as exit signs in buildings or aboard ships. For example, RL light devices in aircraft have no requirement for being labeled or leak-tested.

Radioluminescent exit signs are typically composed of multiple tubes with a maximum amount of tritium in each tube less than 10 Ci. However, the sign may contain more than 10 Ci. Depending on the interpretation by the NRC, these exit signs may or may not be covered under a general license. In addition, each device must have been

manufactured, assembled, or initially transferred in accordance with a license issued under general licensing requirements, or manufactured or assembled in accordance with a specific license issued by an Agreement State. The general license does not authorize the manufacture, assembly, repair, import, or export of luminous safety devices containing tritium or  $^{147}\text{Pm}$ .

### **9.3 Requirements for Nuclear Regulatory Commission Specific Licenses for Manufacturing Tritium Devices**

To obtain a specific license to manufacture products containing tritium, the applicant must satisfy the following requirements:

- demonstrate the adequacy of proposed equipment and facilities, training, and experience to protect health and minimize danger to life and property
- submit sufficient information relating to design, prototype testing, quality control procedures, labeling or marking, and conditions of handling, storage, use, and disposal of product to meet safety criteria
- provide assurance that the product will be designed and manufactured such that dose to the maximally exposed individual shall not exceed the following limits:
  - 1 mrem whole body, 15 mrem extremities, and 3 mrem to the other organs during normal use and disposal of a single exempt unit
  - 10 mrem whole body, 150 mrem extremities, and 30 mrem to the other organs during normal handling and storage of exempt units that would typically accumulate in one location during marketing, distribution, installation, and servicing the product
  - accidents involving exempt units resulting in a low probability of exceeding 0.5 rem to the whole body, 7.5 rem to the extremities, or 1.5 rem to other organs, and a negligible probability of exceeding 1.5 rem to the whole body, 200 rem to the extremities, and 50 rem to other organs.

### **9.4 Nuclear Regulatory Commission Specific License Requirements for Supplying RL Devices to Others**

In order to supply by-product material devices to a general licensee, the applicant for a specific license must meet the following requirements:

- satisfy the requirements in 10 CFR 30.33 concerning equipment and facilities to protect health and minimize danger to life and property and provide adequate training and experience to use the radioactive materials
- submit adequate information with regard to the design and testing of the device
- provide assurance that the device shall have durable, legible, clearly visible labels approved by the NRC

- maintain for a period of 5 years records of all general-licensed persons to which devices were transferred. (It is required that these transfers be reported to the NRC.)

## **9.5 Nuclear Regulatory Commission License Requirements for Manufacturing Aircraft RL Devices**

To hold a specific license for manufacturing RL devices for use in an aircraft, the applicant must satisfy the following requirements:

- demonstrate adequate proposed equipment and facilities, training, and experience to protect health and minimize danger to life and property
- provide sufficient information regarding each device to evaluate the potential exposure from the device
- provide assurance that each device will contain no more than 10 Ci of tritium or 300 mCi of  $^{147}\text{Pm}$  (The dose rate from a  $^{147}\text{Pm}$  device will not exceed 0.5 mrad/h at 10 cm from any surface when measured through 50 mg/cm<sup>2</sup> of absorber.)
- have bound tritium and  $^{147}\text{Pm}$  in the device in such a manner that they will not be released under normal handling
- enclose the tritium and  $^{147}\text{Pm}$  to preclude direct physical contact by any person
- design the device so it is not easily disassembled
- ensure that the device passes the required prototype tests
- label the devices for use by persons with general licenses (Labelling includes model number, serial number, radioactive material and quantity, and sign reading "CAUTION-RADIOACTIVE MATERIAL," and the name of the manufacturer.)
- produce required material transfer reports.

## **9.6 Agreement State Licensing Requirements**

If states are part of the NRC's Agreement States Program, they can regulate the design, manufacture, distribution, and use of by-product material. State laws and regulations and local ordinances may add additional requirements on shipments and shippers of radioactive materials.

## **9.7 U.S. Military Licensing Requirements**

Although certain uses of radioactive materials by military agencies may be exempt, the use of RL lights as runway markers typically would require a license. Military agencies would need to submit to an NRC sealed-source review (see Regulatory Guide 10.11 [NRC 1987b]). The U.S. Air Force has a broad-scope license and issues permits off the broad-scope license. The U.S. Army would license each unit individually.

The following sections discuss in more detail the regulations for the U.S. civilian and military agencies.

## 9.8 Civilian Agencies

U.S. civilian agencies responsible for licensing RL lights include the NRC and Agreement States. Agreement States will follow the same licensing and regulatory requirements as the NRC. (The U.S. Department of Energy [DOE] does not license the use of by-product material.)

### 9.8.1 Nuclear Regulatory Commission

Domestic licensing requirements for by-product material are found in Title 10 Parts 30, 31, and 32 of the U.S. Code of Federal Regulations (CFR 1993d, e, and f). Depending on the amount and type of RL material, requirements range from nothing (exempt) to the need for a specific or general licensee. Radiation protection requirements are found in 10 CFR 20 (CFR 1993b).

General licenses are effective without filing applications with the NRC. Requirements for general licenses for RL light devices used as exit signs can be found in 10 CFR 31.5 and 10 CFR 31.7. Specific licenses for manufacturers and distributors of self-luminous products are issued to applicants pursuant to the regulations in 10 CFR 32.22. Specific licenses for users are issued to applicants pursuant to regulations in 10 CFR 30.32 and 30.33 and 10 CFR 32.210(c).

### 9.8.2 Exemptions

According to 10 CFR 30.19, a user of a self-luminous product containing tritium,  $^{85}\text{Kr}$ , or  $^{147}\text{Pm}$  is exempt from requirements for a license when the products are manufactured, processed, produced, or initially transferred in accordance with a specific license issued according to 10 CFR 32.22. The exemptions discussed in 10 CFR 30.19 are meant to address small quantities (i.e., microcurie amounts) of tritium,

$^{85}\text{Kr}$ , and  $^{147}\text{Pm}$ . Persons using by-product material under certain DOE and NRC contracts are exempt from licensing requirements.

### 9.8.3 General License

Title 10, Parts 30 and 31 of the CFR (CFR 1993d and e) provides requirements for general domestic licenses for by-product material; 10 CFR 30.19 and 10 CFR 31.7 both relate to RL lights. Title 10, Part 30.19 covers RL light exit signs for buildings and ships, while 10 CFR 31.7 covers luminous safety devices for use in aircraft.

Title 10, Part 30.19 of the CFR discusses the conditions under which a general license can be issued to private industry, research facilities, educational and medical facilities, and Federal, state, or local government agencies for the receipt, possession, or use of by-product material contained in devices for the purpose of producing light. The devices must have been manufactured in accordance with the requirements in 10 CFR 32.22. The requirements for the general licenses are set out in Section 9.1 (above).

Title 10, Part 31.7 of the CFR provides a general license for use of luminous safety devices in aircraft. A person can own, receive, acquire, possess, and use tritium or  $^{147}\text{Pm}$  contained in luminous safety devices for use in aircraft. Each device shall contain not more than 10 Ci of tritium or 300 mCi of  $^{147}\text{Pm}$ . The licensing requirements for RL light device usage in aircraft is less stringent than the requirements for RL light devices used as exit signs in buildings or aboard ships. For example, RL light devices in aircraft have no requirement for being labeled or leak tested.

Radioluminescent exit signs are typically composed of multiple tubes with a maximum amount of tritium in each tube that is less than 10 Ci; however, the total radioactivity in the sign may be greater than 10 Ci. Therefore, depending on the interpretation by the NRC, these exit signs may or may not be covered under a general license. In addition, each device shall have been manufactured, assembled, or initially

transferred in accordance with a license issued under the provisions of 10 CFR 32.22 or 10 CFR 32.53, or manufactured or assembled in accordance with a specific license issued by an Agreement State.

Under a general license, the user is exempt from requirements in 10 CFR 19, "Notices, Instructions, and Reports to Workers; Inspections" (CFR 1993a); from 10 CFR 20, "Standards for Protection Against Radiation" (CFR 1993b); and from 10 CFR 21, "Reporting of Defects and Noncompliance" (CFR 1993c). There are two exceptions: the user shall comply with the provisions of 10 CFR 20.402 ("Reports of Theft or Loss of Licensed Materials") and 10 CFR 20.403 ("Notification of Incidents").

The general license does not authorize the manufacturing, assembly, repair, import, or export of luminous safety devices containing tritium or  $^{147}\text{Pm}$ .

#### **9.8.4 License to Manufacture or Transfer By-product Material**

Specific domestic licenses to manufacture or transfer certain items containing by-product material are found in 10 CFR 32 (CFR 1993f). Requirements for a license to manufacture, process, produce, or initially transfer self-luminous products containing tritium,  $^{85}\text{Kr}$ , and  $^{147}\text{Pm}$  are found in 10 CFR 32.22. Requirements for a specific license to manufacture or initially transfer devices containing by-product material to persons generally licensed under 10 CFR 31.5 are found in 10 CFR 32.51. The requirements for a specific license to manufacture, process, and produce luminous safety devices for use in aircraft are found in 10 CFR 32.53.

The requirements for a specific license to use RL light devices can be found in 10 CFR 30.32 and 10 CFR 30.33.

Holders of specific licenses shall comply with the provisions of 10 CFR 19 ("Notices, Instructions and

Reports to Worker; Inspections" [CFR 1993a]) and 10 CFR 20 ("Standards for Radiation Protection" [CFR 1993b]).

To obtain a specific license to manufacture or transfer by-product material, the license applicant must meet the requirements in 10 CFR 32.22, 10 CFR 32.51, and 10 CFR 32.53. These requirements are listed below:

1. 10 CFR 32.22, "Self-Luminous Products Containing Tritium,  $^{85}\text{Kr}$ , or  $^{147}\text{Pm}$ ." According to 10 CFR 32.22, the applicant for a specific license must satisfy the requirements listed in Section 9.3.

The conditions of the license require adequate control procedures in the manufacturing of the product to assure that quality control standards approved by the NRC are met. In addition, labeling and marking each unit is required. Records and file reports must also be maintained with the Director of the Office of Nuclear Material Safety and Safeguards (NMSS). File reports must include information on products transferred to persons according to 10 CFR 30.19. Filing times are discussed in 10 CFR 32.25.

2. 10 CFR 32.51, "Supplying By-product Material Devices to General Licensed." According to 10 CFR 32.51, the applicant for a specific license must meet the requirements listed in Section 9.4 (above).
3. 10 CFR 32.52, Same: Material transfer reports and records.
  - Maintain for a period of 5 years records of all general licensed persons to which devices were transferred. These transfers shall be reported to the NRC.
4. 10 CFR 32.53, "Luminous Safety Devices for Use in Aircraft." According to 10 CFR 32.53, the applicant for specific license must satisfy the requirements listed in Section 9.5 (above).

5. 10 CFR 32.54. Same: Labeling of devices.
  - Label the devices for use by persons generally licensed under 10 CFR 31.7. Labelling shall include model number, serial number, radioactive material and quantity, and sign reading "CAUTION-RADIOACTIVE MATERIAL," and the name of the manufacturer.
6. 10 CFR 32.56. Same: Material transfer reports.
  - Annual material transfer reports shall be submitted annually.

### **9.8.5 License to Use By-product Material**

Title 10, Section 30.32 (10 CFR 30.32) states that an application for a specific license to use by-product material in the form of a sealed source or in a device that contains the sealed source (RL light devices would fall into this category) must either 1) identify the source or device by the manufacturer and model number as registered with the NRC under 10 CFR 32.210, or 2) contain the information identified in 10 CFR 32.210(c), which requires that any manufacturer or initial distributor of a sealed source or device containing a sealed source whose product is intended for use under a specific license may submit a request to NRC for evaluation of radiation safety information about its product and for its registration. Such a request must contain sufficient design and testing information to provide reasonable assurance that the radiation safety properties of the source or device are adequate to protect health and minimize danger to life and property. The NRC will evaluate the request and can issue a certificate of registration.

Guidance for the preparation of applications for specific licenses involving laboratory and industrial use of small quantities of by-product material is found in Regulatory Guide 10.7, Rev. 1 (NRC 1979). Regulatory Guide 10.7 is intended for applicants

using millicurie amounts of by-product material; therefore, it is not applicable to licensing of RL devices.

### **9.8.6 Nuclear Regulatory Commission Radiation Safety Evaluation and Registration**

The NRC determines the adequacy of the radiation safety properties of devices and sealed sources containing by-product material before authorizing the distribution and use of by-product material in a device or sealed source. Regulatory Guide 10.10 (NRC 1987a) provides assistance to manufacturers and distributors of by-product material devices when they submit requests for the NRC radiation safety evaluation and registration of devices containing by-product material. Regulatory Guide 10.11 (NRC 1987b) provides the same sort of guidance for manufacturers and distributors of sealed sources containing by-product material. The application submitted to the NRC should indicate whether the by-product material will be used under an exemption from regulation or under a general or specific license. The application should also contain information on the sealed source type, model, radionuclide and maximum activity, details of construction, labeling, radiation levels, and testing of prototypes. ANSI N540-1975 (1975), Classification of Radioactive Self-Luminous Light Sources, establishes minimum prototype testing requirements for radioactive self-luminous light sources aimed at assuring adequate containment of the radioactive material. As defined in ANSI N540-1975, the maximum activity of gaseous tritium in an individual self-luminous light source is 50 Ci.

Upon approval of the application, the NRC will issue a numbered certificate of registration that identifies the sealed source by type and model, series, or drawing number. The user of a sealed source should identify the source exactly as it is identified in the manufacturer's or distributor's registration certificate when applying for a license. This will allow the licensing reviewers of the NRC or Agreement State

to relate the user's license application to the information in the registration certificate.

### 9.8.7 Transportation Requirements

Transportation requirements for radioactive materials including RL devices are found in 49 CFR 100-178 (U.S. Department of Transportation) and 10 CFR 71 (NRC). The NRC follows the requirements in 49 CFR 100-178. Regulatory requirements are directed towards the assurance of safety through proper packaging for the specific types and quantities of materials to be transported. The majority of the requirements are directed toward the shipper.

State laws and regulations and local ordinances may add additional requirements on shipments and shippers of radioactive materials (Pike and O'Reilly 1982). NUREG/CR-1263 (1980) provides a compilation of state laws and regulations on the transport of radioactive materials.

## 9.9 Military Agencies

The use of RL lights as runway markers typically would require an NRC license. Military agencies would need to submit an application for an NRC sealed-source review as defined in Regulatory Guide 10.11 (NRC 1987b). The U.S. Air Force and U.S. Navy have a broad-scope license, and they issue permits off the broad-scope license. The U.S. Army would license each unit individually.

In a letter dated October 16, 1981, from Joseph C. Wang (NRC, Material Licensing Branch, Division of Fuel Cycle and Material Safety) to the U.S. Department of Air Force, the NRC stated that prior to issuing a license, all devices containing radioactive material must be registered and evaluated for health and safety by the NRC or Agreement State in accordance with the guide, Standard Format and Content of Applications for Health and Safety Review and Registration of Devices Containing Radioactive Material or Content of Application for Custom Licensing of Devices.

## 9.10 References

- 10 CFR 19. 1993a. U.S. Nuclear Regulatory Commission, "Notices, Instructions, and Reports to Workers." U.S. Code of Federal Regulations.
- 10 CFR 20. 1993b. U.S. Nuclear Regulatory Commission, "Standards for Protection Against Radiation." U.S. Code of Federal Regulations.
- 10 CFR 21. 1993c. U.S. Nuclear Regulatory Commission, "Reporting of Defects and Noncompliance." U.S. Code of Federal Regulations.
- 10 CFR 30. 1993d. U.S. Nuclear Regulatory Commission, "Rules of General Applicability to Domestic Licensing of By-product Material." U.S. Code of Federal Regulations.
- 10 CFR 31. 1993e. U.S. Nuclear Regulatory Commission, "General Domestic Licenses for By-products Material." U.S. Code of Federal Regulations.
- 10 CFR 32. 1993f. U.S. Nuclear Regulatory Commission, "Specific Domestic Licenses to Manufacture or Transfer Certain Items Containing By-product Material." U.S. Code of Federal Regulations.
- 10 CFR 71. 1994. U.S. Nuclear Regulatory Commission, "Packaging and Transportation of Radioactive Material." U.S. Code of Federal Regulations.
- 49 CFR 100-178. 1993. U.S. Department of Transportation, "Research and Special Programs." U.S. Code of Federal Regulations.
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## **Appendix A**

### **Dosimetric Terms**

# Appendix A

## Dosimetric Terms

The definitions of the dosimetric terms provided here are based on available sources (IAEA 1986, ANSI 1976, ICRP 1984, ICRU 1980). The definitions are given in the order of increased abstractness rather than alphabetically.

1. Dose. Dose is a term used in radiation protection that serves as a measure of the radiation energy "received" or "absorbed" by a target. The concept of "receiving" dose by a target is usually expressed as "absorbed dose" or "dose equivalent." In common usage, the word "dose" is often used to mean either absorbed dose or dose equivalent, depending on the context.
2. Absorbed Dose. Absorbed dose is defined as the mean energy imparted by ionizing radiation to matter. Mathematically, absorbed dose is defined as

$$D = \frac{d\epsilon}{dm} \quad (\text{A.1})$$

where  $D$  is the absorbed dose and  $d\epsilon$  is the mean energy imparted. The traditional unit of absorbed dose is the rad which is the deposition of 100 ergs/gram of material. Under the International System of Units (SI), the SI unit for absorbed dose is the gray, which is the deposition of 1 joule/kg of material. One gray = 100 rad.

3. Dose Equivalent. Some radiations are more effective at producing radiation damage than others; consequently, equal absorbed doses of different radiations are not always associated with the same degree of harm. In radiation

protection, the difference in effectiveness has been reflected by the quality factor,  $Q$ . The quality factor is a type of weighted average of the effectiveness of radiation to produce an effect as compared to a standard type of radiation (x or gamma rays).

The dose equivalent is the product of the absorbed dose and the quality factor; when absorbed dose is in terms of rad, the corresponding dose equivalent term is rem; when absorbed dose is in terms of gray, dose equivalent is expressed in terms of sievert. Dose equivalent allows for the addition of the effects of different types of radiation; for radiation protection purposes, 1 rem of alpha radiation is equivalent to 1 rem of beta radiation.

In this report, the dose equivalent is computed according to the following equation:

$$H = DQ \quad (\text{A.2})$$

where  $H$  is the dose equivalent,  $D$  is the absorbed dose, and  $Q$  is the quality factor.

The quality factor for tritium may range from 1 to 2 depending on the source of the information. The national and international advisory agencies (e.g., National Council on Radiation Protection and Measurements [NCRP] and International Commission on Radiological Protection [ICRP]) both recommend the use of unity (1) for the tritium quality factor. The U.S. Nuclear Regulatory Commission (NRC) requires a quality factor of 1.7 (NRC 1989) (however,

discussions with NRC staff reveal that these individuals use  $Q = 1$  in their own calculations). Also, the weighting factors published in 10 CFR 20.1004 (NRC 1993) list the weighting factors for beta radiation as 1.0. Because the quality factor used may depend on regulatory concerns, in this report computation of dose equivalent from absorbed dose requires an explicit step. The reader should be aware that computational formulas presented in the literature often contain assumptions about the quality factor (and other factors) that are not often explicitly stated. All calculations performed for this report will use  $Q = 1$ .

4. Committed Dose Equivalent. The ICRP has suggested (ICRP 1977) that, when considering the consequences of an intake of radioactive material, the total dose equivalent received for 50 years after the intake be computed and recorded. This 50-year dose equivalent is the committed dose equivalent. In the cases of tritiated water and elemental tritium, the vast majority of the committed dose is received within 3 months of an intake. Thus, for tritium the committed dose equivalent is identical to the annual dose equivalent (the dose equivalent received the first year after the intake). In this report, equations provide for computation of dose equivalent for any time after an intake of tritium. The symbol for the committed dose equivalent is  $H_{50,T}$ .

5. Committed Effective Dose Equivalent. The ICRP in its Publication 26 (ICRP 1977) has recommended that the dose equivalents from external and internal radiation be summed. To facilitate this summation, the ICRP has introduced a weighting system for the dose equivalent received by individual organs. The purpose is to compare the risk received by an organ to the risk that would be encountered by an individual uniformly irradiated over the whole body. The weighting factors are shown in Table A.1. In those instances where only a few organs are irradiated or the dose equivalent varies greatly between organs, the dose equivalent received by each organ is multiplied by the weighting factor for that organ. The sum of all such products is the effective dose equivalent for the irradiation. In the case of tritiated water, which is uniformly distributed throughout the body following an intake, it is sufficient to compute a dose equivalent for the whole body and use a weighting factor of unity (1). However, in the case of inhalation of elemental tritium, a large fraction of the dose equivalent is received by the lung to the virtual exclusion of other organs. In this case, the dose equivalent to the lung is multiplied by the weighting factor for the lung to provide the effective dose equivalent. (This type of calculation is discussed in Section 4.0). The symbol for the committed effective dose equivalent is  $H_{E,50}$ .

**Table A.1.** Weighting Factors Recommended by the ICRP (1977)

Tissue	Weighting Factor
Gonads	0.25
Breast	0.15
Red Bone Marrow	0.12
Lung	0.12
Thyroid	0.03
Bone Surfaces	0.03
Remainder	0.30

The chemical forms of tritium emphasized in this report are tritium gas (HT or T<sub>2</sub>) and tritiated water (HTO or T<sub>2</sub>O). The dose from an intake of tritiated water will be evenly distributed throughout the body. In this case, the weighting factor is 1 and the effective dose equivalent is numerically equal to the dose equivalent to the whole body. In contrast, an intake of tritium gas will irradiate the lung to a much greater extent than the rest of the body. In this case, the effective dose equivalent, due to irradiation of the lung, is the product of the dose equivalent to the lung times the weighting factor of 0.12. Other chemical forms of tritium, not discussed to any extent in this review, may concentrate in specific organs, in which case the weighting factors for these other organs should be employed.

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## **Appendix B**

### **Examples of Dose Projections**

## Appendix B

### Examples of Dose Projections

The dose projections considered below include occupational dose projections for both indoor and outdoor accidents and environmental dose projections, illustrated by examples of a runway fire and a warehouse incident.

#### B.1 Occupational Dose Projections

In this section, two kinds of incidents are described: incidents that occur indoors and those that occur outdoors. The first incident describes an incident that could occur in an enclosed area, e.g., a warehouse. In this situation, the worker is exposed to a gas cloud that slowly diffuses. The second incident is one that occurs outside. In this situation, the gas will be rapidly diluted and dispersed by wind currents.

##### B.1.1 Indoor Accidents

The hypothetical indoor doses are divided, for purposes of analysis, into 1) accidents that involve the breaking of RL lights in either ventilated or unventilated areas and 2) the slow release of tritium through the glass walls of the RL device.

**Ventilated Area.** The dose calculation for an indoor ventilated area has the following underlying assumptions: a light breaks while an individual is in the area, the tritiated water is instantly dispersed throughout the area, the tritiated water is removed from the air volume in a manner defined by the air turnovers of the area, and the individual remains in the area until all of the tritium has dissipated.

The radiation dose to a worker who is exposed to tritium in an enclosed area is a function of the initial concentration of the material in the air, the rate at which the material is vented from the area, and the total time that the individual remains in the area.

The air concentration at any time,  $t$ , following the release of radioactivity into a room can be estimated from the equation:

$$C(t) = C_o \exp(-\lambda_e t) \quad (\text{B.1})$$

where  $C(t)$  = air concentration at any time,  $\mu\text{Ci}/\text{mL}$   
 $C_o$  = original air concentration,  $\mu\text{Ci}/\text{mL}$ , or (activity released,  $\mu\text{Ci}$ )/(room air volume,  $\text{mL}$ )  
 $e$  = base of the natural logarithms  
 $\lambda_e$  = effective removal constant,  $\text{time}^{-1}$ , for  $\lambda_e = \lambda_p + \lambda_l$  where  
 $\lambda_p$  = physical decay constant,  $\text{time}^{-1}$  and  
 $\lambda_l$  = exfiltration removal constant,  $\text{time}^{-1}$   
 $t$  = time at which the activity is to be calculated.

The cumulative air concentration is the time integral of Equation (B.1):

$$\bar{C} = \int_0^t C_o \exp(-\lambda_e t) dt \quad (\text{B.2})$$

which is:

$$\tilde{C} = C_o \frac{1}{\lambda_e} \left[ 1 - \exp(-\lambda_e t) \right] \quad (\text{B.3})$$

where  $\tilde{C}$  = cumulative air concentration, (μCi-h)/mL  
 $\lambda_e$  = the effective removal constant, time<sup>-1</sup>  
for  $\lambda_e = \lambda_p + \lambda_q$  where  
 $\lambda_p$  = physical decay constant, time<sup>-1</sup>  
and  
 $\lambda_q$  = exfiltration removal constant, time<sup>-1</sup>  
t = the time at which the activity is to be calculated.

If possible, the ventilation rate of the actual storage building should be used. If knowledge of the actual air turnover rates is lacking, the information in Table B.1 can be used. The data in the table were developed for purposes that are different than the requirements of this document. The tabulated values are, nonetheless, useful indicators of reasonable air turnover rates.

**Unventilated Area.** In the previous incident, the activity in the area gradually decreased over time as the air was changed by the ventilation system. In this incident, the assumptions are that an individual is in a large enclosed area and that the lights are broken, immediately releasing their entire contents, which are completely and uniformly distributed throughout the building. The individual inhales the radioactive material for the duration of time that he is working in the area. The air concentration of tritiated water is assumed to remain constant. (Note: The individual may reduce the air concentration by inhaling the tritiated water but this effect is ignored for these calculations. The reduction of the air concentration is expected to be small except where a room is small and the individual occupies the room for a relatively long time. However, if the situation were such that the tritiated water concentration would be affected, the oxygen level would also be affected).

**Table B.1.** Air Turnover in Buildings

Building Type	Turnover Rate, h <sup>-1</sup>	References
Wood Frame	0.5 - 9	Johnson et al. 1973, p. 15ff.
Research Lab Buildings	8 - 10	Personal communication, PNL building managers
Sealed Control Room	0.06	Murphy and Campe 1974
Reactor Building	0.00114	NRC 1989, p. 3.7
Glove Boxes	10-30	Robins, Bartoszek and Wordall 1984
Laboratories	1-10	"
Reactor Halls	0.25 - 1	"
Suggested Rates - This Document		
Buildings, wood frame	1 - 5	
Buildings - air tight (range)	0.015 - 0.06	
Buildings - airtight with circulation	0.015	
Buildings - airtight without circulation	0.001	

**Buildup of Tritium in a Room from Diffusion Through Glass.**

Tritium can diffuse through glass at a very slow rate. If the RL devices are contained in a sealed area, the tritium activity will build up with time. Simultaneous with the buildup, from diffusion through glass out of the light sources, the tritium will decrease due to radioactive decay and exfiltration from the room.

Herman and Jones (1978) described experiments in which tritium-activated compasses were stored in three locations representing semi-arid, moderate-, and high-humidity regions. The compasses were stored in semi-open shed, a well-ventilated brick warehouse, a humidity-controlled warehouse, a quonset hut, and a sealed warehouse section. They reported that no airborne tritium was found in any storage locations except the sealed warehouse where the activity was found to be on the order of 1.5 µCi/mL. Unfortunately, the activity stored and the volume of the storage area were not reported. The tritium concentration is high because the compasses were not sealed units. (The tritium was incorporated into a paint.) Although the air contained tritium, no "significant removable contamination" was found on the cartons, shelving, or areas surrounding the cartons.

A factor in the buildup of tritium gas in sealed warehouses is the quality of the manufacture of the device. Very small amounts of tritium will be expected to diffuse through glass. However, Herman and Jones (1978) report leakage rates of up to 1.6E-2 µCi/day for sealed glass tubes (the activity in the leaking tube was not stated but similar sources contained up to 450 mCi).

Ristagno (1978) reports that tubes containing tritium for use in digital watches (having an area of 21 to 64 mm<sup>2</sup>) leaked at a rate of less than 0.01 µCi/24 h per watch (the number of tubes per watch was not stated).

Equation (B.4) describes the buildup of tritium with time:

$$C(t) = \int_0^t L \exp[-\lambda_e(t-\tau)] d\tau \quad (B.4)$$

- where
- C(t) = the total activity in the area, Ci
  - L = the leach rate from the tubes, Ci/unit time
  - λ<sub>e</sub> = the effective removal constant, time<sup>-1</sup> for λ<sub>e</sub> = λ<sub>p</sub> + λ<sub>q</sub> where
    - λ<sub>p</sub> = physical decay constant, time<sup>-1</sup>
    - λ<sub>q</sub> = exfiltration removal constant, time<sup>-1</sup>
  - t = the time at which the activity is to be calculated
  - τ = the elapsed time between when the <sup>3</sup>H was leached and the time for estimating the <sup>3</sup>H activity

The time units may be in terms of any convenient time scale, i.e., seconds, minutes, hours, days, etc., but they must be compatible. The above equation is readily solved:

$$C(t) = L \frac{1}{\lambda_e} (1 - \exp(-\lambda_e t)) \quad (B.5)$$

The cumulative air concentration is the integral of Equation (B.5):

$$\tilde{C} = \int_0^t \frac{1}{\lambda_e} L (1 - \exp(-\lambda_e t)) dt \quad (B.6)$$

which is:

$$\tilde{C} = L \frac{t}{\lambda_e} - \frac{L}{\lambda_e^2} (1 - \exp(-\lambda_e t)) \quad (B.7)$$

- where
- ~C = the integral air concentration, µCi-h/ft<sup>3</sup>

$L$  = the rate of diffusion of tritium through the walls of the tubes, Ci/unit time  
 $\lambda_e$  = the effective removal constant,  $\text{time}^{-1}$   
 for  $\lambda_e = \lambda_p + \lambda_l$  where  
 $\lambda_p$  = physical decay constant,  $\text{time}^{-1}$   
 $\lambda_l$  = exfiltration removal constant,  $\text{time}^{-1}$   
 $t$  = the time at which the activity is to be calculated.

The diffusion rate of tritium through the walls of the tubes can be computed using equations developed by Easterly, Noguchi, and Bennett (1985), who investigated the ingrowth of THO in the absence of ventilation. Under conditions of no ventilation, the reaction rate can be described by the equation:

$$\frac{dA}{dt} \text{THO} = \lambda A_{\text{TH}} \quad (\text{B.8})$$

where  $\lambda$  is the THO buildup factor. This equation can be integrated over time to give

$$A_{\text{THO}} = A_0(1 - e^{-\lambda t}) \quad (\text{B.9})$$

When using the above equation, the diffusion rate may be assumed to be a constant if the calculation is carried out for short time intervals; in 1 year, the tritium activity in the devices will decrease by only about 5%.

### B.1.2 Outdoor Accident

In order to compute the dose from the outdoor accident, it is necessary to determine the extent to which the radioactive material has dispersed between the point of the release and the individual who inhales the material. To compute the dispersion, standard atmospheric diffusion equations are used.

Descriptions and derivations of the equations can be found in Hanna, Briggs, and Hosker (1982), Miller (1984), Slade (1968), and NRC Regulatory Guides 1.3 and 1.4 (NRC 1974a; NRC 1974b). The atmo-

spheric diffusion equations use tables and charts that show the numerical values for the variables  $\sigma_y$  and  $\sigma_z$  that are shown in the equations. These tabulated variables change value based on the distance between the source and the receptor; they are generally not valid for distances less than 100 m. For the short distances involved for occupational accidents (on the order of 10 m or less), we have used the charts published by Bowne (1974).

The diffusion equations are used to compute an integral air concentration at a particular point downwind from the release. The advantage of the integral air concentration is that we are not required to know the rate at which the radioactive material was released. The target individual is assumed to be downwind for the full duration. This feature will maximize the computed dose. If the release takes place over long periods of time, the calculated dose will be unreasonably large. For releases of long duration (greater than 8 hours), occupancy factors may be used (NRC 1974b); occupancy factors are not discussed in this report.

The equation used to compute the  $\chi/Q$  value for a point source and a point receptor is as follows (NRC 1974a; NRC 1974b):

$$\chi/Q = \frac{1}{U\pi\sigma_z\sigma_y} \frac{s}{m^3} \quad (\text{B.10})$$

where  $\chi/Q$  = relative integral air concentration,  $\text{s/m}^3$   
 $U$  = the wind speed (source to receptor),  $\text{m/s}$   
 $\sigma_z$  = vertical dispersion coefficient,  $\text{m}^{-1}$   
 $\sigma_y$  = horizontal dispersion coefficient,  $\text{m}^{-1}$ .

The wind speed is often taken as 1 m/s (see, for example, NRC 1974a and NRC 1974b); calm winds are taken to have a wind speed of 0.5 m/s (Hanna, Briggs, and Hosker 1982, p. 25). For distances less than 100 m,  $\sigma_z$  and  $\sigma_y$  values of Bowne (1974) are

used. To be conservative, the least turbulent wind conditions are used.

The quantity inhaled following a release is the product of the quantity released, the  $X/Q$ , and the breathing rate of the individual. The equation that may be used is:

$$\text{Intake} = (\text{BR})(\chi/Q)(\text{release}) \quad (\text{B.11})$$

where BR = the breathing rate,  $\text{m}^3/\text{s}$   
 $\chi/Q$  = the relative integral air concentration,  $\text{s}/\text{m}^3$   
 release = the activity released.

The equation may be used twice, once for the tritiated water activity released and again for the elemental hydrogen released. The effective dose equivalent from tritiated gas is about 4 orders of magnitude less than that from tritiated water; thus, unless the tritiated water content is very small, it appears common to ignore the dose contribution from the elemental form.

There have been several estimates of the radiation dose that will occur as a result of an outdoor incident. The results of these calculations are shown in Table 6.6. The assumption used for these calculations is that the individual is working in close proximity to a device that breaks, releasing all of its contents to the air. We have used the equations used for plume calculations with the assumption that the person is inhaling the activity of the most concentrated part of the plume. The sigma values used for these calculations are those plotted by Bowne (1974) for small distances between the source and the person (see Tables 6.7 and 6.8). These calculations show that for up to about 1000 Ci of released tritium, the radiation dose is fairly small, on the order of tens of mrad. The calculations that were performed ignore the radiation dose from the elemental form of tritium and from any tritiated water that might migrate to the outside surface of the broken lights and be absorbed through the skin.

**Effect of Rain.** Material may be removed from a plume by the actions of rain or snow. In general, the process is assumed to remove material uniformly throughout the vertical extent of the plume. The air concentration at any distance may be computed by using a modified source strength (IAEA 1982, Sect. 3.4.2):

$$f_w = \exp(\Lambda x/u) \quad (\text{B.12})$$

where  $\Lambda$  is the washout coefficient,  $\text{s}^{-1}$ ;  $x$  is the downwind distance, m; and  $u$  is the wind speed, m/s.

Little data on washout coefficient are available. The data that exist are inconsistent, but are presented in Table B.2. The washout coefficient ( $\Lambda$ ) is assumed to be approximately proportional to the precipitation rate.

## B.2 Environmental Dose Projections

To demonstrate how to calculate environmental dose projections, two incidents are described. In the first incident, the source of the tritium is assumed to be very diffuse; tritium-activated runway markers are assumed to be destroyed in place.

The second set of calculations assumes that tritium is released from a small, localized area, e.g., a warehouse. For this incident, two situations are modeled. The first case is that the plume from the release travels along the ground. The second case is that the plume is elevated to a height of 50 m before it begins to travel downwind and disperse.

It should be kept in mind that, for the ground plume situations, the concentration will increase as the affected individual gets closer to the point of release. The worst calculated radiation dose was about 60 rad to the affected individual at a distance of 100 m from the source. For environmental dose projections, the calculations do not estimate the

**Table B.2.** Available Washout Coefficients ( $\Lambda$ ) for Rain or Snow\*,  $\Lambda = C\theta$

Precipitation		Comments
$\Lambda \text{ s}^{-1}$	mm/h	
$10^{-4}$	--	Theoretical
$4.0 \cdot 10^{-4}$	4 rain	Semi-empirical
$2.6 \cdot 10^{-5}$	1 rain	Theoretical and measured
$10^{-4}$	4 rain	Theoretical and measured
$2.4 \cdot 10^{-4}$	5 rain	Calculation
$2.6 \cdot 10^{-5}$	1 snow	Theoretical and measured
$3.0 \cdot 10^{-5}$	1 snow	Measured

$C = 3 \cdot 10^{-9} \text{ (a/(mm}\cdot\text{s))}$  and  $\theta = \text{rainfall rate(mm/a)}$

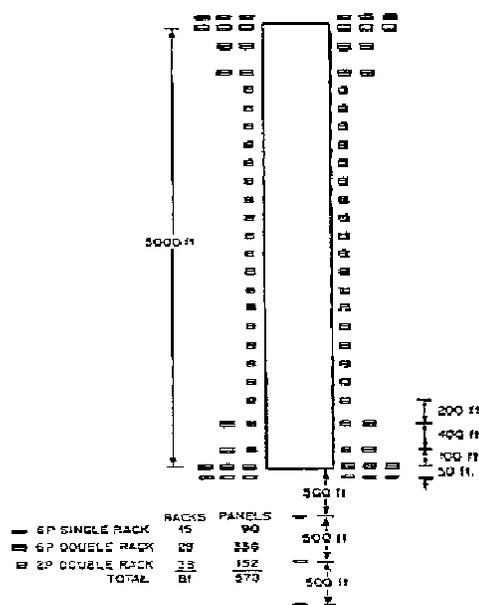
\* Data supplied by private communication: P. A. Davis, Atomic Energy Canada, Ltd. (AECL Research).

radiation doses at distances less than 100 m, but clearly the radiation doses will be greater for an individual who is closer to the incident.

The integral air concentrations were computed via computer codes based on the computer code WRAITH (Scherpelz et al. 1986a). The equations used by these codes were the straight-line Gaussian plume dispersion equations. In the case of the elevated release incident, the equations were modified to account for the elevated release point.

### B.2.1 Runway Fire

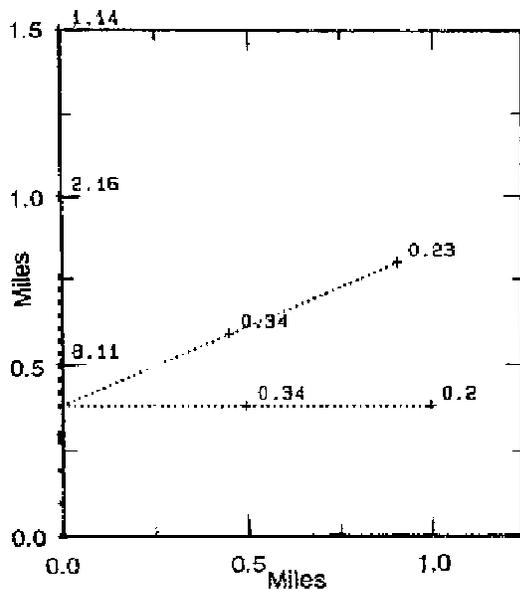
In this incident, it was assumed that tritium-activated runway markers were deployed in the same configuration as the SALTY DEMO 85 layout, shown in Figure B.1. The accident scenario involved the catastrophic destruction of 120 RL panels, approximately one-half of the markers on one side of the runway. The wind conditions modeled were wind blowing down the runway, wind normal to the runway, and wind  $65^\circ$  to the runway. Integral air concentrations were calculated at one-half-mile intervals. The largest doses are associated with the "wind blowing down the runway" situation.



**Figure B.1.** Airfield Lighting Layout, SALTY DEMO 85. The integral air concentrations for this accident were computed with a computer code that implemented Equation (B.10).

This situation yields the largest doses because the plume is more concentrated; it is not so wide and spread out as for the other situations that were modeled.

The tritiated water air concentration for each light was computed and then the contribution from each light was summed to obtain the values shown in Figure B.2. The values in the figure are in terms of  $\text{Ci}\cdot\text{s}/\text{m}^3$ , which is numerically equal to integral air concentrations in terms of  $\mu\text{Ci}\cdot\text{s}/\text{mL}$ , the units required for the equations used in computation of health physics units (see Section 6.0). The greatest air concentration shown in Figure B.2 is equivalent to  $8.11 \mu\text{Ci}\cdot\text{s}/\text{mL}$ , which resulted in a committed effective dose equivalent of about 15.3 rem. This dose would require that regulatory authorities be notified within 24 hours.



**Figure B.2.** Calculated Accumulated Tritiated Water Air Concentrations for the SALTY DEMO 85 (Point 0.0 is the location of the lead runway light; the dotted lines indicate wind direction; values are tritium air concentrations ( $\text{Ci}\cdot\text{m}^3$ )).

## B.2.2 Warehouse Incident

In this incident, it was assumed that the tritium-activated devices were in close proximity to each other when the contents were released. Two different cases were considered: a ground level release and an elevated release. The ground level release would model the situation where lights are stored in a small area, the containment becomes destroyed, and the contents of the lights are released and blown along the ground by the wind. In the second situation, the elevated release, it was assumed that the tritium was released and rose to an elevation of 50 m before it was blown downwind. The calculations for these two incidents were performed using the computer code DUTAD (Scherpelz et al. 1986b).

**Ground Level Release.** The ground level release can be used as a surrogate for any incident in which the activity is released from a small area and is blown downwind without appreciable plume rise. These calculations would also be valid for any area that has few trees or other objects on the ground; the terrain is assumed to be smooth and gently rolling. Table B.3 shows the results of the calculations for different atmospheric conditions at ground level. Isoleths that show the downwind dose estimates are shown in Figures B.3 to B.8.

**Elevated Release.** The elevated release can be used as a surrogate for any incident in which the activity is elevated to a height of 50 m before it is blown downwind and dispersed. The value of 50 m is a conservative value for plume rise of hydrogen following a fire. A fire may elevate the plume to heights that may range from 46 to 293 meters (Selander 1981). The calculations presented here would also be valid for a release in an enclosed area that was vented through a stack having a height of 50 m. Table B.4 shows the results of the calculations for different atmospheric conditions at a 50-m height. Isoleths that show the dose estimates are shown in Figures B.9 to B.14.

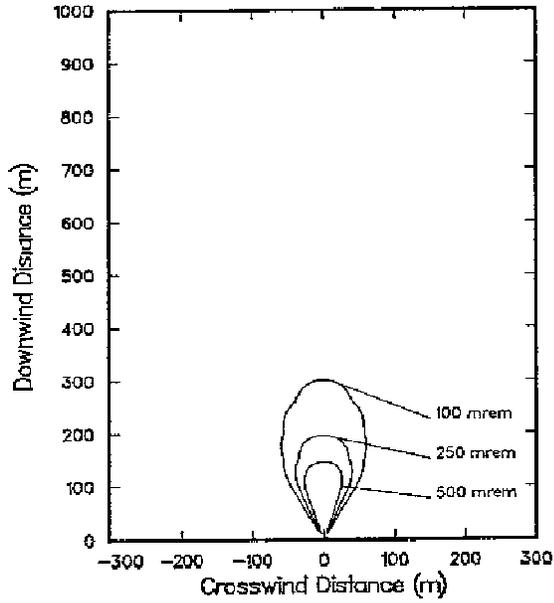
**Table B.3.** Warehouse Incident Calculations for Ground-Level Release Under Different Atmospheric Conditions (Average Wind Speed = 2.0 m/s)

Distance from Release Point, m	Pasquill Stability Class ( $\chi/Q$ , s/m <sup>3</sup> )					
	A	B	C	D	E	F
100	5.503E-04	9.947E-04	1.700E-03	4.233E-03	8.842E-02	2.915E-02
150	2.210E-04	4.421E-04	8.268E-04	1.950E-03	4.113E-03	1.206E-02
200	1.130E-05	2.456E-04	4.856E-04	1.168E-03	2.736E-03	6.499E-03
260	6.208E-05	1.439E-04	2.957E-04	7.262E-04	1.443E-03	3.789E-03
360	2.781E-05	7.361E-05	1.574E-05	4.082E-04	8.041E-04	2.110E-03
500	1.123E-05	3.674E-05	8.511E-05	2.264E-04	4.372E-04	1.132E-03
675	4.577E-06	1.890E-05	4.874E-05	1.333E-04	2.689E-04	6.663E-04
900	1.641E-06	1.001E-05	2.858E-05	8.072E-05	1.665E-04	3.971E-04
1250	4.866E-07	4.677E-06	1.584E-05	4.759E-05	9.829E-05	2.319E-04
1700	2.471E-07	2.153E-06	9.253E-06	2.951E-05	6.034E-05	1.437E-04
2350	1.868E-07	9.365E-07	5.286E-06	1.775E-05	3.585E-05	8.686E-05
3200	1.416E-07	3.702E-07	3.002E-06	1.079E-05	2.221E-05	5.513E-05
4400	1.072E-07	1.686E-07	1.706E-06	0.598E-06	1.420E-05	3.551E-05
6000	8.162E-08	1.083E-07	1.001E-06	4.153E-06	9.205E-06	2.308E-05
8300	6.092E-08	7.990E-08	5.815E-07	2.608E-06	5.930E-06	1.519E-05
11300	4.629E-08	6.007E-08	3.512E-07	1.776E-06	4.009E-06	1.042E-05
15500	3.521E-08	4.611E-08	2.103E-07	1.252E-06	2.721E-06	6.992E-06
21000	2.725E-08	3.565E-08	1.303E-07	7.696E-07	1.917E-06	5.010E-06
29000	2.072E-08	2.688E-08	7.728E-08	4.676E-07	1.302E-06	3.413E-06
40000	1.571E-08	2.040E-08	4.634E-08	2.956E-07	9.043E-07	2.327E-06

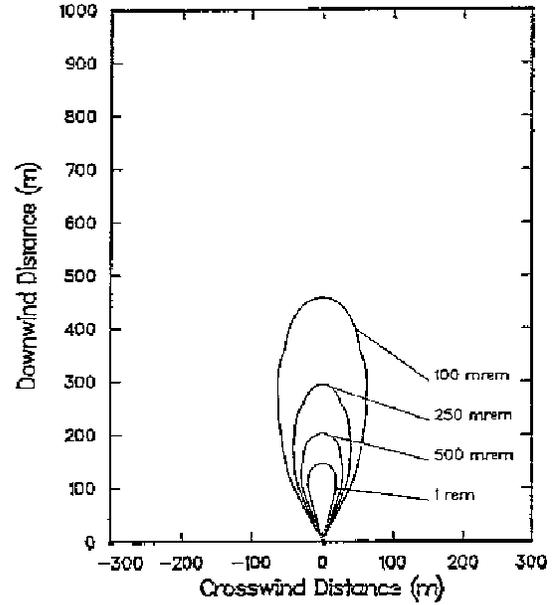
In Figures B.15 and B.16 the downwind dose estimations are plotted against the downwind distance for the warehouse incident.

Also shown on these figures are the actions required by the U.S. Environmental Protection Agency (EPA) and the NRC. In all cases, written notification to the NRC would be required. The ground level release figure warrants some explanation. The evacuation and shelter levels are recommendations of the EPA. If the dose levels to offsite individuals exceed these

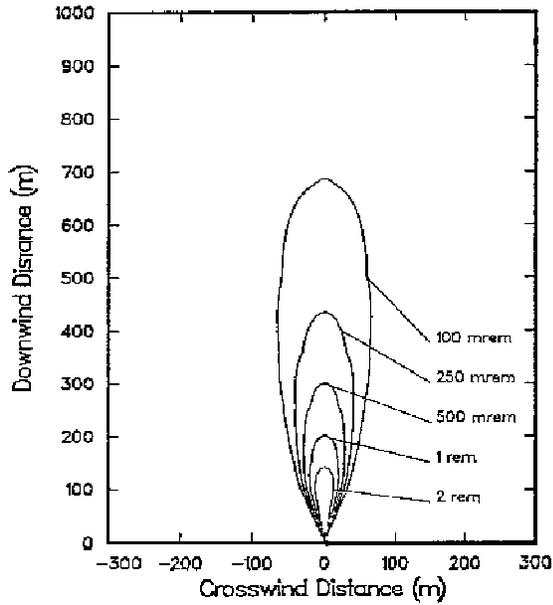
levels, then sheltering, or evacuation, of the population should be considered. These are only recommendations: each state has jurisdiction within its boundaries. Evacuation would be recommended only if the non-occupational population lives within about 1 km of the release point and the atmospheric conditions were very stable; otherwise, sheltering would be recommended. The plume will generally traverse the site in less time than would be required for evacuation; thus, sheltering would be the most useful recommendation.



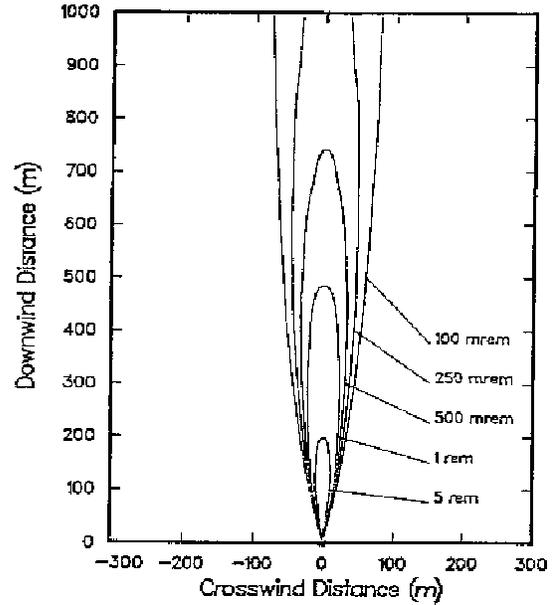
**Figure B.3.** Dose Profiles Downwind of a Ground Level Release of Tritium, Pasquill Stability Class A



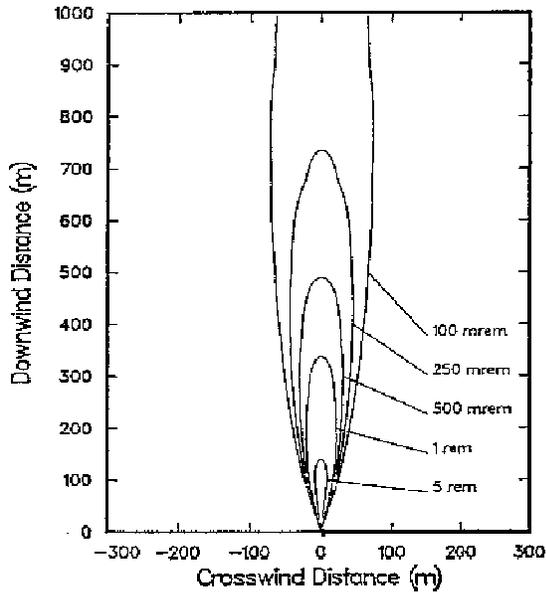
**Figure B.5** Dose Profiles Downwind of a Ground Level Release of Tritium, Pasquill Stability Class C



**Figure B.4.** Dose Profiles Downwind of a Ground Level Release of Tritium, Pasquill Stability Class B



**Figure B.6.** Dose Profiles Downwind of a Ground Level Release of Tritium, Pasquill Stability Class D

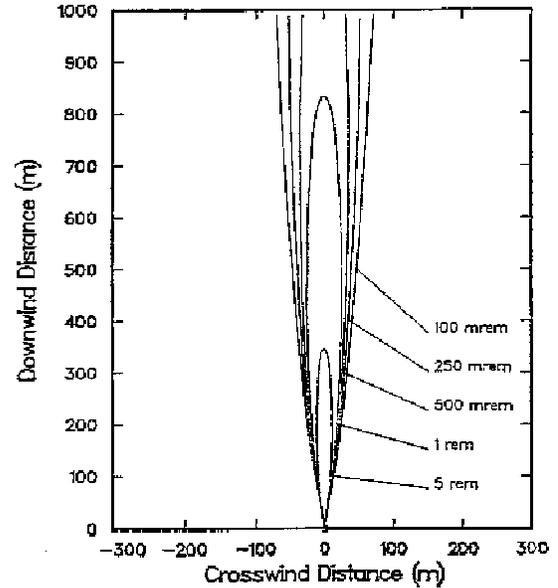


**Figure B.7.** Dose Profiles Downwind of a Ground Level Release of Tritium, Pasquill Stability Class E

The figures show that 24-hour notification of the NRC would be required under most instances. It should be kept in mind when looking at these curves that a ground-level release of this type is very unlikely. For the tritium to be released as water would imply the presence of fire. As previously discussed, a fire would likely drive the plume to a height of 50 m, in which case Figure B.15 that relates to an elevated release is more applicable. Figure B.15 indicates that the tritium air concentrations do not reach the levels required for sheltering and that 24-hour notification of the NRC would be required if the atmospheric conditions were very stable.

### B.3 References

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**Figure B.8.** Dose Profiles Downwind of a Ground Level Release of Tritium, Pasquill Stability Class F

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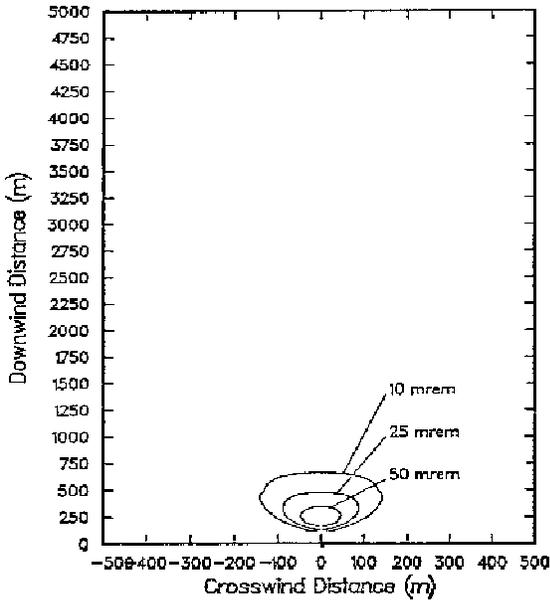
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**Table B.4.** Warehouse Incident Calculations for 50-m Elevated Release Under Different Atmospheric Conditions (Wind Speed = 2 m/s)

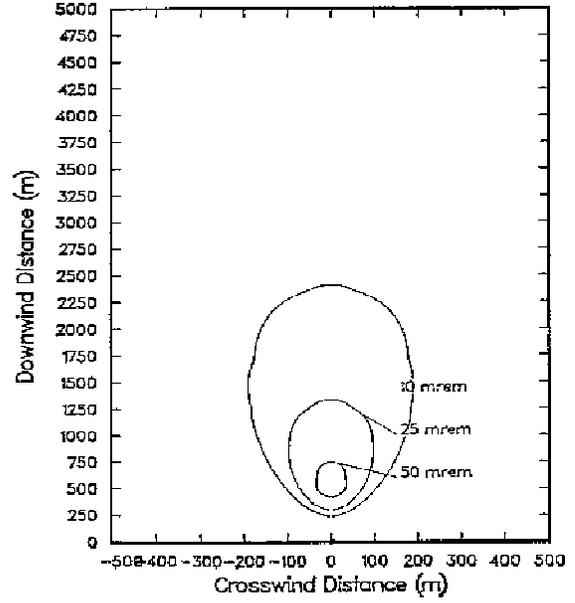
Distance from Release Point, m	Pasquill Stability Class ( $\chi/Q$ , s/m <sup>3</sup> )					
	A	B	C	D	E	F
100	1.953E-06	3.707E-09	2.031E-12	1.125E-27	0.000E+00	0.000E+00
150	1.871E-05	1.709E-06	2.697E-08	3.547E-15	1.795E-32	0.000E+00
200	3.524E-05	1.165E-05	1.030E-06	6.488E-11	4.647E-20	0.000E+03
260	3.412E-05	2.476E-05	6.652E-06	1.777E-08	1.136E-13	5.645E-35
360	2.218E-05	3.144E-05	2.017E-05	9.300E-07	1.158E-09	1.174E-21
500	1.048E-05	2.501E-05	2.886E-05	7.097E-06	2.682E-07	4.520E-13
675	4.489E-06	1.572E-05	2.593E-05	1.591E-05	2.726E-06	1.132E-09
900	1.634E-06	9.159E-06	1.971E-05	2.075E-05	8.100E-06	1.071E-07
1250	4.863E-07	4.513E-06	1.283E-05	2.003E-05	1.438E-05	1.363E-06
1700	2.471E-07	2.125E-06	8.166E-06	1.668E-05	1.698E-05	4.772E-06
2350	1.867E-07	9.324E-07	4.922E-06	1.242E-05	1.589E-05	9.746E-06
3200	1.416E-07	3.697E-07	2.881E-06	8.464E-06	1.270E-05	1.183E-05
4400	1.072E-08	1.685E-07	1.667E-06	5.602E-06	9.613E-06	1.127E-05
6000	8.159E-08	1.082E-07	9.872E-07	3.708E-06	6.939E-06	9.710E-06
8300	6.090E-08	7.987E-08	5.766E-07	2.410E-06	4.808E-06	7.869E-06
11300	4.628E-08	6.005E-08	3.493E-07	1.677E-06	3.410E-06	6.202E-06
15500	3.520E-08	4.609E-08	2.096E-07	1.200E-06	2.401E-06	4.656E-06
21000	2.724E-08	3.564E-08	1.301E-07	7.459E-07	1.728E-06	3.556E-06
29000	2.072E-08	2.688E-08	7.718E-08	4.574E-07	1.198E-06	2.587E-06
40000	1.570E-08	2.040E-08	4.630E-08	2.911E-07	8.429E-07	1.856E-06

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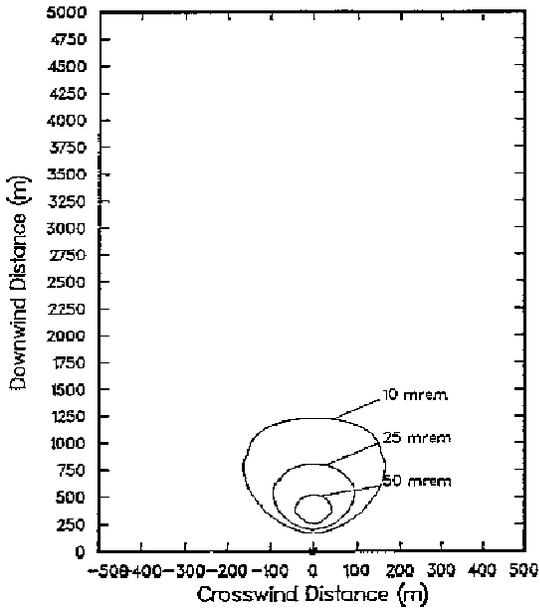
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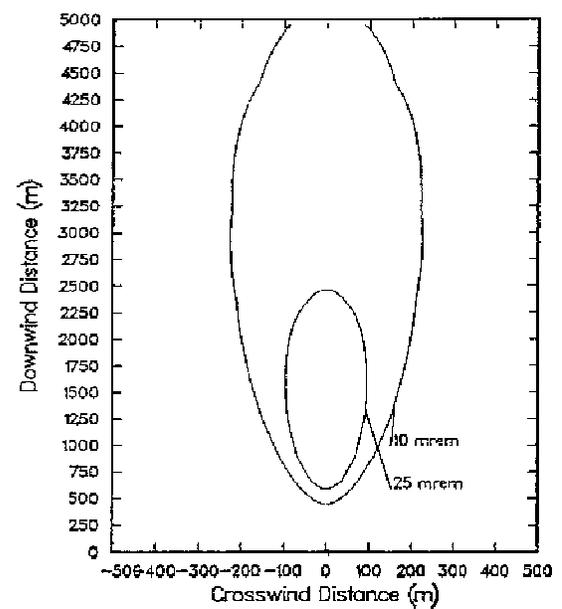
**Figure B.9** Dose Profiles Downwind of a 50-m Elevated Release of Tritium, Pasquill Stability Class A



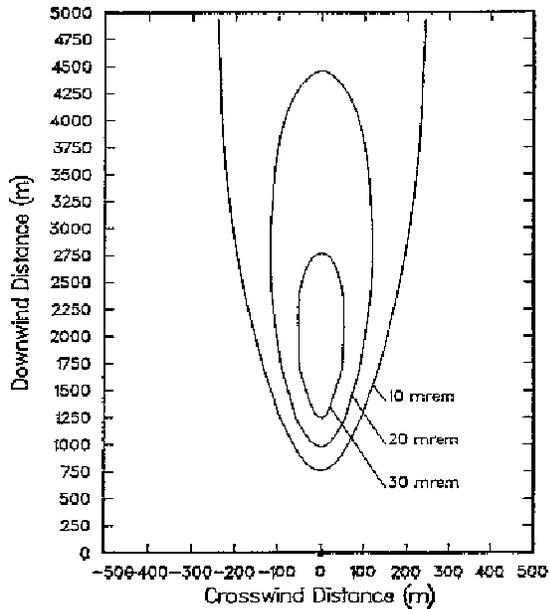
**Figure B.11** Dose Profiles Downwind of a 50-m Elevated Release of Tritium, Pasquill Stability Class C



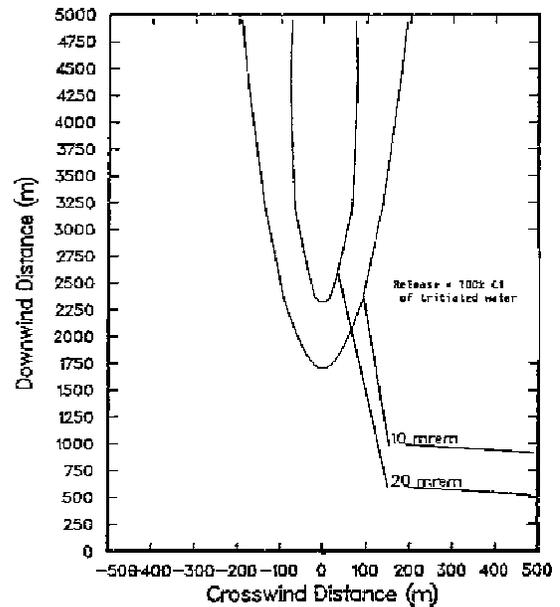
**Figure B.10** Dose Profiles Downwind of a 50-m Elevated Release of Tritium, Pasquill Stability Class B



**Figure B.12** Dose Profiles Downwind of a 50-m Elevated Release of Tritium, Pasquill Stability Class D



**Figure B.13.** Dose Profiles Downwind of a 50-m Elevated Release of Tritium, Pasquill Stability Class E



**Figure B.14.** Dose Profiles Downwind of a 50-m Elevated Release of Tritium, Pasquill Stability Class F

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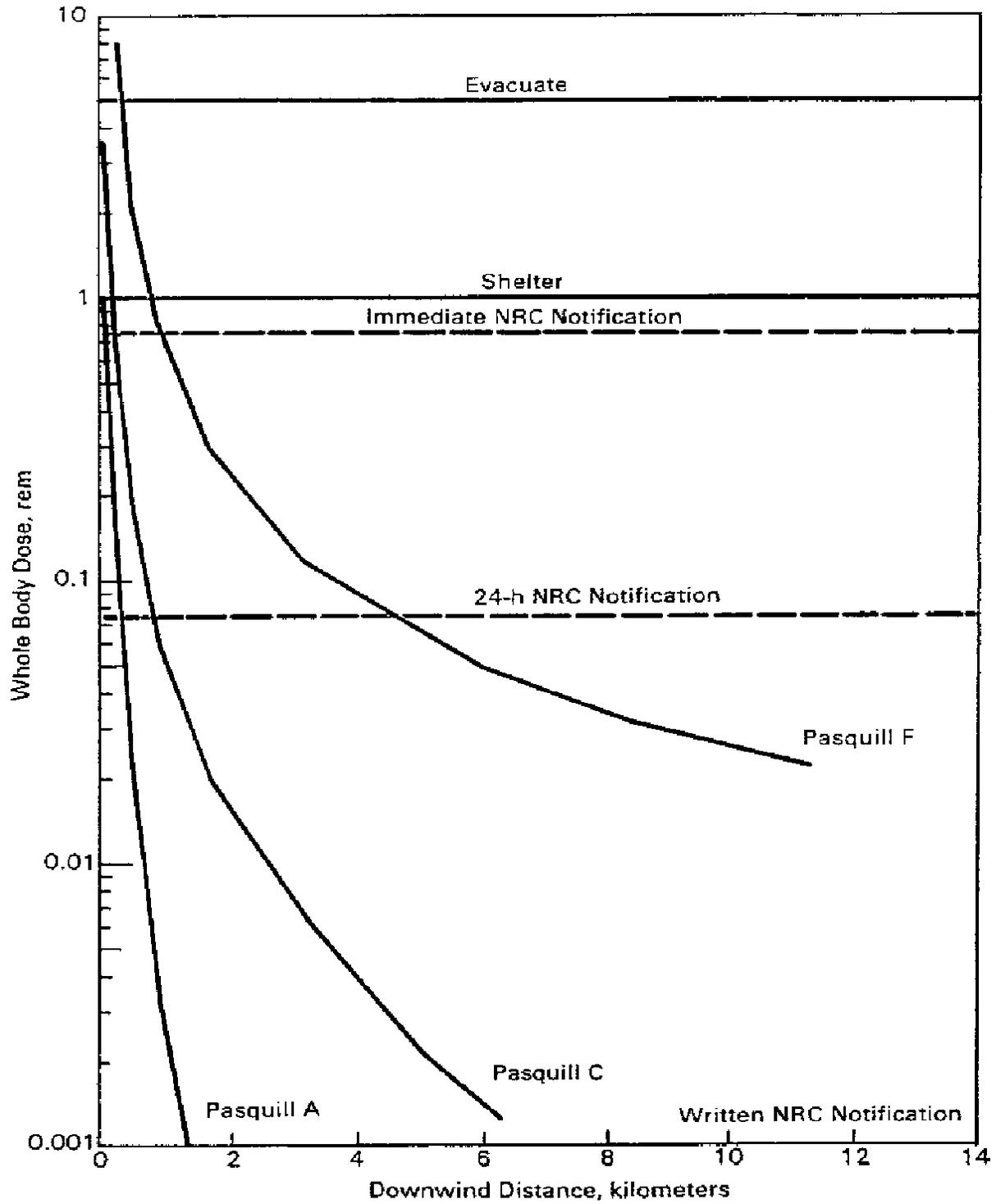


Figure B.15. Ground Level Release of 100,000 Ci of Tritiated Water from Warehouse Incident

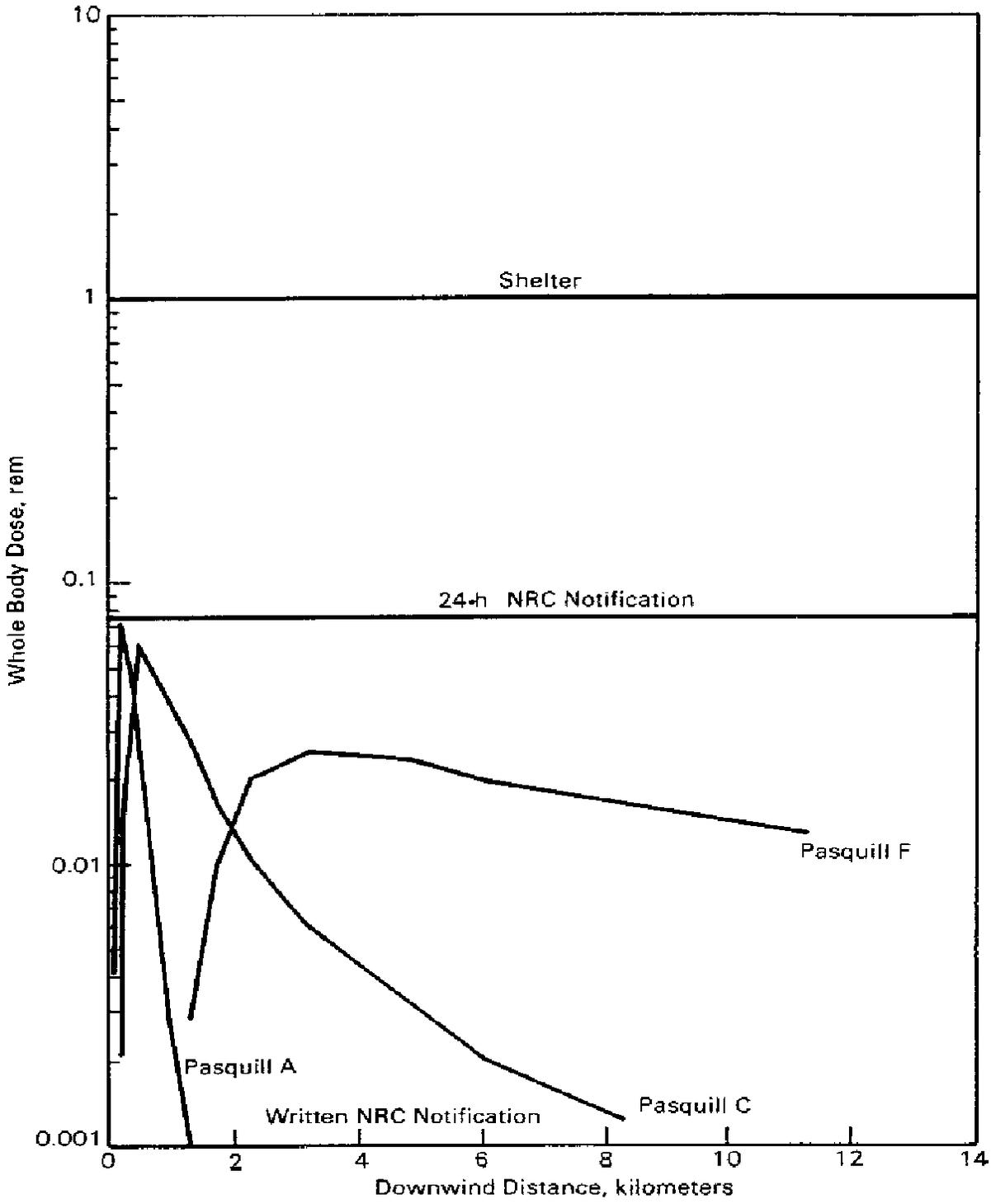


Figure B.16. 50-m Elevated Release of 100,000 Ci of Tritiated Water from Warehouse Incident

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