

Contents

1	Introduction	6
2	Motivation	7
2.1	Nuclear Energy	7
2.2	Tracer	8
3	Background Theory	8
3.1	Tritium in the hydrological cycle	8
3.2	Health Risks	10
3.3	Quantifying the Health Effects of Radioactivity.	11
4	Existing Models	12
5	TERM	14
5.1	Gaussian Plume and Gaussian Puff	16
5.2	Continuous atmospheric release	17
5.3	Discrete atmospheric release	19
5.4	Continuous river release	23
5.5	Discrete river release	24
5.6	Determining the values of the diffusion coefficients (D_{xt} , D_{yt} and D_{zt}) . . .	25
5.7	Continuous coastal release	25
5.8	The contribution of the river and coastal models to dose	26
6	Model Validation	26
6.1	Continuous Atmospheric HTO Release	26
6.2	Discrete Atmospheric HTO Release	30
6.3	Discussion of model validation	30
7	Case Studies	33
7.1	Wylfa	33
7.2	Chalk River Laboratories 2008 release	36
7.3	Case Study Discussion	40
8	Conclusions and Further Work	41
	References	42
A	Discrete Atmospheric Model Plant Transfer Term	45
B	Uniform transfer rate from plant water to plant organic matter.	46

C Activity of pure, undiluted HTO	47
D Input Parameters for Wylfa's Routine Atmospheric Release.	47
E Input Parameters for Chalk River Laboratory Accidental Release	48
F Risk Assessment Retrospective	50

List of Figures

1	Tritium levels of atmospheric water in the northern hemisphere (based on the GNIP database)- from J.R. Gat. <i>Isotope Hydrology, A Study of the Water Cycle</i> , [16]	6
2	Outline of the model	15
3	Image Sources	16
4	Concentrations in a river due to a continuous line source (Bq/L)	25
5	Air concentration assuming zero release height, 2006 meteorological data, and stability probabilities given in Table 4. Air activity concentration (HTO) ($\log_{10}(Bq/m^3)$)	28
6	Comparison of TERM using estimated stability class probabilities with measured values and IMPACT model results.	29
7	Comparison of TERM assuming stability class D and stability class F with measured values.	29
8	12 and 16 sector model comparison	29
9	Model Comparison	29
10	Comparison of TERM and UFOTRI results for a hypothetical release.	30
11	Wind Rose for Valley [32]	33
12	Wylfa Release Results	35
13	Graphic representation of the plane source discrete river release model	37
14	Planar discrete release predicted slug center concentrations	37
15	Results assuming a river width of 1790m and a release of $3 \times 10^{10} Bq$	37
16	The air concentrations predicted by TERM for a discrete release of 4.5TBq. Colourbar in $\log_{10}(Bq/m^3)$	38
17	The soil concentrations in layer 1 predicted by TERM for a discrete release of 4.5TBq (Colourbar in $\log_{10}(Bq/L)$)	39
18	The soil concentrations in layer 2 predicted by TERM for a discrete release of 4.5TBq (Colourbar in $\log_{10}(Bq/L)$)	39
19	The soil concentrations in layer 3 predicted by TERM for a discrete release of 4.5TBq (Colourbar in $\log_{10}(Bq/L)$)	39

20	The plant compartment of the discrete release model [34]	46
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List of Tables

1	Variations in water regulations between countries [4]	11
2	Model Comparison	14
3	Stability Categories of Pasquill [27]	27
4	Estimated probabilities for each stability class in each wind speed class. . .	27
5	Pasquill-Gifford (NRC) diffusion formulae coefficients	32
6	Ottawa Valley Release Statistics [12]	36

Glossary

C_i	concentration in compartment i
Q	the amount released in Bq/s (for continuous models)
x	the radial direction in the continuous atmospheric model
z	the vertical direction in the continuous atmospheric model
$\Delta\theta$	The sector angle for the sector averaged Gaussian Plume model
F_{ijk}	triple joint frequency
u_k	typical wind speed of wind speed class k (continuous model)
h_{eff}	effective release height
σ_x	dispersion coefficient in the x direction
σ_y	dispersion coefficient in the y direction
σ_z	dispersion coefficient in the z direction
RF_{sw}	the ratio of HTO concentration in soil water to that in air moisture (Bq/L soil water per Bq/L air moisture)
H_a	air humidity (L/m^3)
$H_{a,time}$	air humidity averaged over period “time” (L/m^3)
H_{sat}	saturated air humidity (L/m^3)
RF_p	the ratio of HTO concentration in plant water to that in air moisture (Bq/L plant water per Bq/L air moisture).
DW_p	dry to fresh weight ratio for plants
ID_p	isotopic discrimination factor for plant metabolism
WE_p	the water equivalent of plant dry matter
DW_a	dry to fresh weight ratio for animals
ID_a	isotopic discrimination factor for animal metabolism
WE_a	the water equivalent of the animal dry matter

f_{ww}	the fraction of animal water that is derived from ingestion of water
f_{wa}	the fraction of animal water that is derived from inhalation of air
k_{ww}	the fraction of animal water intake from contaminated well water
k_{pw}	the fraction of animal water intake from contaminated pond water
f_{wpw}	the fraction of animal water that is derived from plant water
f_{wdw}	the fraction of animal water that is derived from plant dry matter
k_{fp}	the fraction of animal water intake from contaminated pond water
Bucket_ratio	empirical factor for the concentration in precipitation in a bucket to that in air
<i>Andelman</i>	the Andelman volatilisation factor for radionuclide pollutants
λ	the decay rate of tritium
Z_{tos}	the distance to top of well screen from the soil surface (m)
<i>porosity</i>	effective soil porosity (unitless)
q_{infil}	groundwater infiltration rate through soil (m/s)
R_{HT}	ratio of HTO concentration in air moisture (Bq/L) to HT concentration in air (Bq/m^{-3})
f_{oxid}	fraction of year when oxidation can occur (unitless)
CF_{HT}	a factor equal to the ratio of HTO concentration in plant water to HT concentration in air ($Bq/L_{plantHTO}$ per Bq/m^3_{airHT})
x, y and z	are cartesian coordinates in the discrete atmospheric models
M	amount released in Bq (for discrete models)
U	wind speed in discrete atmospheric release model and river velocity (for river models) (m/s)
u^*	friction velocity (m/s)
U_o	The velocity in the “free stream”. This is taken to be the measured wind speed in this model
ν	kinematic viscosity of air (m^2/s)
St_k	The Stanton number as a function of roughness length
Re^*	Reynolds number based on roughness length and friction velocity
Pr	Prandtl number
v_{HTO}	deposition velocity of HTO (m/s)
$r_{aerodynamic}$	aerodynamic resistance s/m
$r_{boundary.layer}$	boundary layer resistance
r_{soil}	soil resistance
θ_i	the soil moisture volume fraction of layer i
θ_{sat}	the saturated soil moisture volume fraction
θ_{wilt}	the wilting point soil moisture volume fraction
h_{si}	soil layer height i
Λ	washout coefficient (s^{-1})

<i>precipitation</i>	the amount of precipitation in (mm/hr)
α	H/T isotope ratio in liquid and air, assumed to be 1.1
μ	water content per unit area of leaf (g/cm^2)
r_{plant}	total resistance to plant (sum of aerodynamic, boundary layer and canopy resistance)
E_{soil}	evapotranspiration $Lm^{-2}s^{-1}$
α_q, m, β	constants for a particular soil (α_q is noted as α in the documentation[42])
x	distance along the stream-wise direction (river models)
y	distance in the transverse direction (river models)
z	distance in the vertical direction (river models)
A	cross-sectional area of river (river models)
D_x	dispersion coefficient in the stream-wise direction (river models)
D_y	turbulent diffusion coefficient in transverse direction (river models)
D_z	turbulent diffusion coefficient (river models)
D_L	longitudinal dispersion coefficient
t	time elapsed since the beginning of the simulation
y_o	distance of the source form the lower bank (river models)
h	height of channel (m) (in river models)
n	the Manning coefficient
w	river width (m)
z_o	distance of the source from the river bed (river models)
F	the flow rate in a river L/s
R_h	hydraulic radius (m)
x	distance between source and point of interest (coastal) (m)
U_c	the annual average current in the direction towards the point of interest
α_{coast}	the annual average fraction of time that the current direction is towards the point of interest from the source
β_{coast}	annual average recirculation factor for the source
Q_v	annual average volumetric discharge rate of liquid effluent (L/s)
κ	proportionality coefficient
d	average water depth in the reach occupied by the plume (m) (coastal model)
D_o	initial dilution at the point of discharge
$DW_{aq.p}$	dry to fresh weight ratio for aquatic plants
$ID_{aq.p}$	isotopic discrimination factor for aquatic plant metabolism
$WE_{aq.p}$	the water equivalent of the aquatic plant dry matter
DW_{aqua}	dry to fresh weight ratio for aquatic animals

$ID_{aq,a}$	isotopic discrimination factor for aquatic animal metabolism
$WE_{aq,a}$	the water equivalent of the aquatic animal dry matter
$t_{lossOBT}$	the rate of loss of hydrogen from the organic part of the plant (s^{-1})
M_{gHTO}	mass of hydrogen in the grass water (kg)
M_{gOBT}	mass of hydrogen in the grass organic matter (kg)
M_{pHTO}	mass of hydrogen in the plant water (kg)
M_{pOBT}	mass of hydrogen in the plant organic matter (kg)
$t_{gHTOgOBT}$	transfer rate of H from grass water to grass organic matter (s^{-1})
$t_{gOBTgHTO}$	transfer rate of H from grass organic matter to grass water (s^{-1})
$t_{pHTOpOBT}$	transfer rate of H from plant water to plant organic matter (s^{-1})
$t_{pOBTpHTO}$	transfer rate of H from plant organic matter to plant water (s^{-1})

1 Introduction

Tritium (^3H) concentrations in the atmosphere peaked in the northern hemisphere during 1963 (see Figure 1) as a result of testing thermonuclear devices. Concentrations were highest in the mid-continental locations [28]. Since atmospheric testing of thermonuclear devices was banned in 1963, the levels of tritium in the atmosphere, and precipitation, have decreased to pre-bomb concentrations [16]. For this reason interest in tritium declined and research into its behaviour and transport also declined as a result.

However, tritium has recently once again become of interest to the scientific community as the advantages of using tritium as a tracer for water are recognised and its rate of production increased with the expected expansion of the nuclear industry (see subsection 2.1 and subsection 2.2). Thus there is a need to model its transfer through the hydrosphere and atmosphere reliably to ensure that exposures to radioactive contamination are kept to levels that are as low as reasonably practicable (ALARP). Routine and accidental releases must be modelled in order to quantify the risk associated with particular sources.

The aims of this project are to review existing models for tritium releases and produce a new model building on this earlier work. The model should incorporate the advantages of existing models, as well as additional processes.

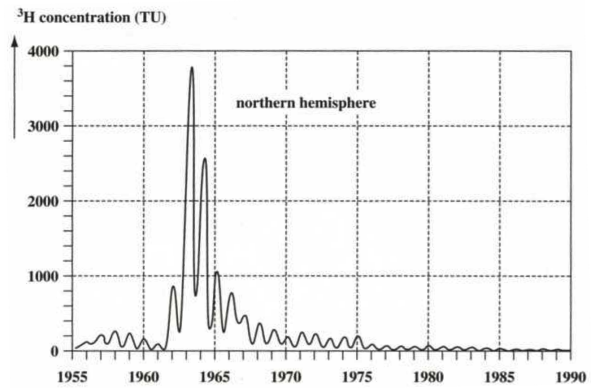


Figure 1: Tritium levels of atmospheric water in the northern hemisphere (based on the GNIP database)- from J.R. Gat. *Isotope Hydrology, A Study of the Water Cycle*, [16]

2 Motivation

The following sections detail the production and/or use of tritium which will lead to an increased global inventory of the isotope.

2.1 Nuclear Energy

In an age where the security of future energy supply is uncertain due to the finite amounts of fossil fuel reserves available and concerns over global warming, safe alternative forms of energy must be considered and investigated. Some of these options (sections 2.1.1 and 2.1.2) involve the use and/or production of radioactive materials. Environmental modelling of potential releases will be an essential part of the safety cases for such developments.

2.1.1 Fission

Design assessments are currently underway on two new pressurised water reactors (PWR): AP1000 submitted by Westinghouse and UK EPR submitted by EDF and AREVA [15] for 8 sites in the UK [30]. These reactors produce tritium via fission. The tritium is released mainly in reprocessing plants, although some may leak through the fuel cladding to the primary coolant during operation. Tritium is a ternary fission product within nuclear fuel and is produced at a rate of $3.7\text{--}7.4 \times 10^{10}$ Bq per year/1,000MW(e) [41]. Tritium is also produced via neutron bombardment of boron and lithium found in the reactor coolant of PWRs which may also result in a release to the atmosphere [21].

2.1.2 Fusion

The technology for the use of fusion reactors as large-scale electricity production facilities is still under development. The reaction responsible for the generation of heat is shown in Equation 2.1. A deuterium and tritium nucleus fuse to produce a helium nucleus and a neutron. Binding energy is released by the process.



Fusion reactors will require a large inventory of tritium if used on a large scale [11] and will breed tritium from the neutron bombardment of lithium [20]. The possible leakage or accidental release of tritium should be modelled in order to ensure low risk quantities of tritium are stored and to aid emergency planning in the event of an accidental release.

2.2 Tracer

Tritium has a number of advantages as a tracer for water. HTO's chemical behaviour is almost exactly the same as non-tritiated water; it is inexpensive; has a convenient radioactive half life; and it is easily measured by liquid scintillation. Also, the slightly larger atomic radius of ^3H compared with ^1H causes only a small isotopic effect [29]. The isotope can be used to trace water flows in the environment, for various purposes such as the ageing of water [36].

3 Background Theory

Tritium is the natural radioactive isotope of hydrogen, which has two neutrons and a proton in its nucleus. Tritium is a weak beta emitter with a half life of 12.32 years [11]:



Tritium is naturally produced as a result of cosmic ray bombardment of the stratosphere [11, 29, 21, 19, 16] but anthropogenic sources of tritium are responsible for the majority of the environment's inventory of the isotope. Sources include: nuclear reactors, fuel reprocessing plants, heavy water production facilities, commercial production for medical diagnostics, radio-pharmaceuticals, luminous paints and many more [11].

3.1 Tritium in the hydrological cycle

The study of tritium requires an understanding of the hydrological cycle as it eventually incorporates all releases of tritium in the form of tritiated water [29]. The movement of tritiated water in the environment is very similar to the cycling of regular water; this is why it is used as a water tracer [28, 16]. There are some differences to take into account in the hydrological cycle such as the effects of radioactive decay energy on the kinetics of reactions. There are also isotopic effects due to the increased mass and size of tritium relative to protium (^1H).

3.1.1 Isotopic effects on evaporation and diffusion.

Diffusion tends to be slower for heavier isotopes [11]. The experimentally obtained ratio of the diffusivity through air of H_2O to that of HDO is 1.0250 [16]. Since HTO is heavier than HDO, this suggests that the diffusivity through air of HTO will also be smaller than that of H_2O .

Molecules with heavier isotopes tend to have higher concentrations in the condensed phase due to higher boiling and condensation temperatures [16]. T_2O has a boiling point $1.51^\circ C$ higher than H_2O [21, 11].

Michel [28] states that tritium follows water's pathway through the hydrological cycle almost exactly, with only small perturbations due to fractionation effects during phase changes. He notes that these fractionation effects can usually be ignored relative to measurement uncertainties and the change resulting from radioactive decay. Where relevant, isotopic factors have been included in the model produced in this project (section 5).

3.1.2 Radioactive Decay

The half life of 12.32 years means that the tritium in the environment will have decreased significantly a few decades after a release event. Since tritiated water traces the motion of water in the hydrological cycle, it can be used to measure the residence time of meteoric waters in a particular compartment [16]. The peak concentration of tritium in the Northern hemisphere from 1963 (section 1) has been used for this purpose.

3.1.3 Isotopic effects on chemical reactions.

Differing atomic masses and the presence of radioactive energy cause isotopic effects in hydrogen isotopes. Atoms with larger atomic masses have lower zero-point energies [41]; therefore the energy required to break a bond with a deuterium or tritium atom will be higher than that required for a bond with a protium atom. Reaction mechanisms requiring this bond to break will therefore have a higher activation energy resulting in slower reaction kinetics. By contrast, the presence of the energy of decay provides energy to fuel reactions [21, 41].

One relevant example of this can be seen in the exchange reaction for deuterium shown below:



Only a very small amount of deuterium exchanges with the hydrogen in water in the absence of a catalyst [21]. Since tritium is a heavier isotope one may expect the rate of the equivalent reaction for tritium to be slower:



However, ionizing radiation [21, 41] from radioactive decay of surrounding tritium isotopes can provide the activation energy for the reaction. With high concentrations of tritium [41] this reaction may occur at a reasonable rate without a catalyst.

3.1.4 Oxidation of HT to HTO

In addition to the isotope exchange reaction (Equation 3.3), tritiated gas can be oxidised to form tritiated water in the soil by the action of bacteria [11]:



It has been suggested that isotope exchange of tritiated gas with water to form tritiated water (Equation 3.3) occurs at a rate of 1% per day [21]. Bacterial oxidation (Equation 3.4) occurs at the soil-air interface at a much shorter half life of 40 minutes to 5 hours. This is the only mechanism considered in previously existing tritium transport models (except for PC-CREAM 08 which does not model HT releases) since it is the most significant.

3.1.5 Effects of precipitation and temperature on tritium in the environment.

The concentration which reaches the ground via precipitation varies depending on a number of factors. *Washout* is where radioactive material is scavenged by rain falling through the plume [38]. *Rainout* is where the radioactivity has been incorporated into the rain-cloud, and is then deposited with the associated precipitation [38]. Light, long-duration rainfall events can yield higher tritium concentrations than heavy short-duration events [11]. Larger rain drops tend to have lower tritium concentrations compared to smaller droplets and the concentration of tritium in raindrops increases with length of travel through the contaminated air mass[11].

Dry deposition values are found to be higher in summer compared to winter, since it occurs via direct vapour exchange with soil water or snow on the ground and the rate of this process increases with increasing temperature. [11]

3.2 Health Risks

As with all radioactive nuclei there are health risks associated with tritium. Because the stochastic health effects of radiation exposure have no threshold value below which they do not occur [18], exposures must be kept as low as reasonably practicable (ALARP).

The form of the tritium affects the health risks associated with exposure. Exposures to HT have less serious biological effects because the probability of retention in the body is much less than for HTO [21]. This is reflected in the higher dose factors used for HTO compared to HT [18].

Table 1: Variations in water regulations between countries [4]

Canada	7,000Bq of tritium per Litre
EU	100Bq of tritium per Litre
California Public Health Goal	14.8Bq of tritium per Litre

Tritium only poses a risk once inside the body; this is due to the low penetrating power of the beta particle that results from its decay [21, 39]. The beta particle can not penetrate through the dead layer of skin [39]. Intakes of HTO via ingestion, inhalation or skin absorption are almost completely absorbed and distributed throughout the body within 1 or 2 hours; HTO has a biological half life of 10 days [24].

The guidelines for safe quantities of tritium in drinking water vary between countries, as seen in Table 1.

3.3 Quantifying the Health Effects of Radioactivity.

The doses to humans are quantified in the following ways:

Absorbed dose (D) is the radioactive energy absorbed per unit mass, given in J/kg or, equivalently, Grays (Gy) [38]. The possible biological effects of radiation however, depend on the type of radiation as well as the quantity of energy deposited. The International Commission on Radiological Protection (ICRP) therefore defined the notion of an *equivalent dose*.

Equivalent dose (H_T) is the absorbed dose modified by a radiation weighting factor (w_R), which takes into account the relative dangers of different types of radiation. It is also given in J/kg, called Sievert (Sv) to indicate that the quantity is the absorbed dose multiplied by the radiation factor [38]. Equivalent dose limits for various organs are defined in order to prevent deterministic health effects as a result of radiation [18].

Different tissues and organs irradiated with radioactivity may result in more detriment to health than others, and this is accounted for in the ICRP *effective dose*.

Effective dose (E): the equivalent dose multiplied by the tissue weighting factor (w_T). The tissue weighting factor takes into account the relative associated harm to health of exposure of a particular organ to an equivalent dose of radiation. As for the equivalent dose the units of effective dose are J/kg and also given the name sievert (Sv) [38]. An effective dose limit of 1mSv/a is defined for members of the public who are not employees of the nuclear industry. Higher limits are used for trainees (6mSv/a) and fully trained employees (20mSv/a) [25].

4 Existing Models

Since hydrogen is a very common element, and a basic element of biological molecules, tritium (^3H) transport through the environment is more complex than for other nuclides. This gives rise to different methodologies for modelling it [18, 38, 13]. There is a need to see what is lacking in existing models of tritium transfer to guide improvement.

Four existing radionuclide transfer models have been investigated in this project:

1. CANDU Owners Group Inc. Derived Release Limits Guidance by D. Hart [18]
2. PC-CREAM 08 [38]
3. GENII-Version 2 [13]
4. UFOTRI [34, 35]

UFOTRI is the only model which is specifically designed for modelling tritium releases. The remaining models model the transport of many different radionuclides. Tritium is a special case in models 1 to 3. Models 1 to 3 do not concentrate on the detail of the transport of tritium through the environment; rather, they use a simple *Specific Activity*¹ model, which is intended to reduce the uncertainty of the results for tritium [18]. Until recently the importance of tritium in most radioactive release models has been low compared to other elements and therefore less data has been collected for model calibration [38]. A summary of what is included in each of the four models investigated is given in Table 2. Details of the models are given below.

CANDU Owners Group Inc. Derived Release Limits Guidance. D. Hart [18]

This model (the “Hart” model) is intended for calculating derived release limits for CANDU Owners Group Inc. facilities. It is applicable to continuous, low-level emissions at a constant rate under steady state environmental conditions [18]. The atmospheric transport is modelled via a sector averaged Gaussian plume model using historical hourly meteorologic data. Reflection from the ground is taken into account in the calculations. Simple models are used for releases to both rivers and coastal waters. Equilibrium conditions are assumed and so all other processes are modelled by equilibrium ratios of specific activity. Hence there is no information about the rates at which transfer processes occur.

PC-CREAM 08 [38]

This was produced by the Health Protection Agency (HPA) based on a methodology given by the European Commission [38]. Like Hart, the software is developed for application to

¹*specific activity* (SA) is the activity per mass of total hydrogen [18]. However, it is usually expressed in units appropriate to the compartment under consideration (Bq/L for water or Bq/kg fresh weight for living organisms). The simplest Specific Activity model assumes the tritium is mixed uniformly, physically and chemically in a compartment. It is assumed, if an organism draws hydrogen from one compartment only, that the SA of the organism will be the same as that compartment. If an organism draws hydrogen from many compartments, the SA of the organism will be a weighted value of the SA of the various compartments it is associated with [18].

routine releases, assuming equilibrium conditions. As in Hart, atmospheric releases are modelled using a sector averaged Gaussian plume dispersion model, but reflections from the top of the mixing layer are taken into account as well as from the ground. The surface water model assumes instantaneous mixing. The terrestrial pathways are modelled via specific activity concepts as in the Hart model.

GENII Version 2 [13]

This was developed for the US department of energy. Chronic releases are modelled as in Hart and PC-CREAM 08. Simple modifications to the equations for tritium have been made to model a discrete release event to the atmosphere, but not to surface waters. Atmospheric releases are modelled using a Gaussian plume model which takes into account the reflection from the ground and mixing layer. Simple models are used for releases to coastal lakes and rivers. As in Hart and PC-CREAM 08 the terrestrial pathways are modelled using specific activity concepts.

UFOTRI [34, 35]

This is a model developed in Karlsruhe Institute of Technology for assessing the radiological consequences of an accidental atmospheric tritium release [35]. Some process rates are calculated assuming equilibrium conditions, while and others are calculated from established equations (e.g. Monteith's bulk resistance formula for evaporation rate [35]). The rates specified in the model can be used for the dynamic case of an accidental release. Atmospheric release is modelled using a segmented Gaussian plume model, and re-release from the ground is modelled using straight line Gaussian plume models with an initial broadening of the plume to model an area source.

Table 2: Model Comparison

	Hart	PC CREAM 08	GENIIv2	UFOTRI
HT release	•		•	•
HTO release	•	•	•	•
HT to HTO conversion	•		•	•
Outside and inside air concentrations modelled separately			•	
Continuous release	•	•	•	
Accidental one-off release			•	•
Atmospheric release	•	•	•	•
River release	•	•	•	
Coastal release	•	•	•	
Suspended sediment		•		
Soil pore water	•			•
Plant water	•	•	•	•
Plant organic content	•		•	•
Animal water	•	•	•	•
Animal organic content	•		•	•
Well water	•			
Pond water	•			
Irrigation	•			
Honey	•			
Aquatic animals	•	•		
Aquatic plants	•	•		
Dose from inhalation HTO	•	•	•	•
Dose due to skin absorption included in inhalation dose	•	•	•	
Dose due to skin absorption modelled independently				•
Dose from HT inhalation	•		•	
Dose from drinking water	•	•	•	
Dose from plant HTO	•	•	•	•
Dose from plant OBT	•		•	•
Dose from animal HTO	•	•	•	•
Dose from animal OBT	•		•	•
Dose from honey	•			
Dose from liquid water immersion	•		•	
Dose from aquatic plant HTO	•	•	•	
Dose from aquatic plant OBT	•		•	
Dose from aquatic animal HTO	•	•	•	
Dose from aquatic animal OBT	•		•	

5 TERM

The main objective for the new Tritium Environmental Release Model (TERM) was to incorporate all of the important processes of previous models (see Table 2) and model surface runoff to reservoirs, an additional processes not included in existing models. A simple outline of the model is shown in Figure 2. This model offers varying complexity with the following features:

- Different options for the environmental compartment to which the radioactivity is released (atmosphere, river, coastal)
- The option to model the release as continuous or discrete. The discrete release models give the unsteady evolution of concentration values.
- The option of 3 different source types for the river models and an area or point source for the discrete atmospheric release

- The option to include or exclude different terrestrial components depending on the area under consideration
- The choice of varying the number of image sources used
- The option of approximating some equationally modelled values by constants

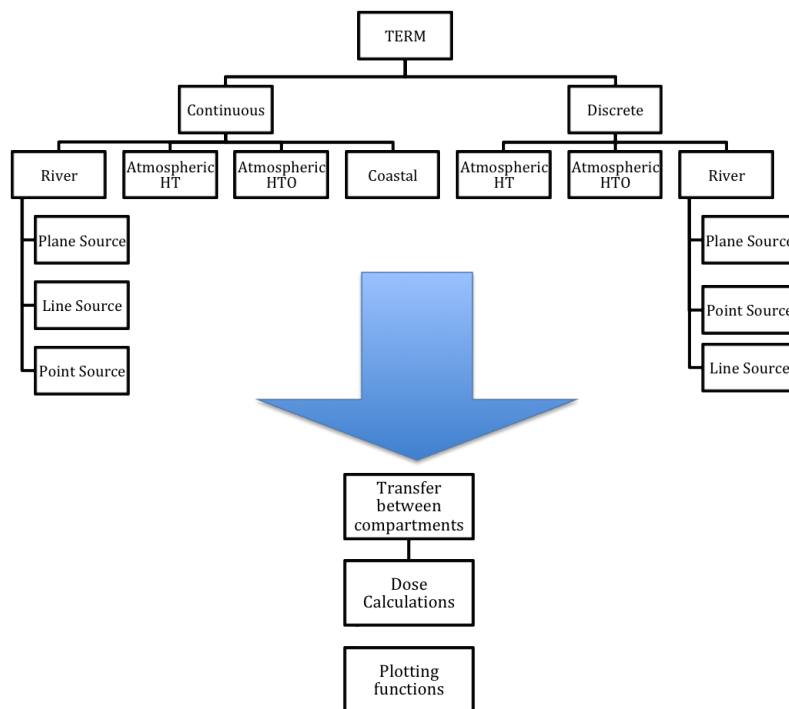


Figure 2: Outline of the model

It is assumed that neglecting the transfer to sediment is reasonable, although PC CREAM 08 models it in surface waters via the use of a partition coefficient (K_d). Transfer to sediment was not included in the new model because there is a wide range of values for (K_d) quoted in the literature [1, 38] (from between 0.03 and $1\text{m}^3\text{t}^{-1}$) and its value is heavily dependent on conditions. $1\text{m}^3\text{t}^{-1}$ is orders of magnitude less than the values for all other radionuclides and all the models investigated, apart from PC-CREAM 08, assume that the $K_d = 0$ for tritium.

The details of the various components of the model are given in the sections below. An important equation throughout many of the models is that of the Gaussian Puff and the Gaussian Plume. These are explained first.

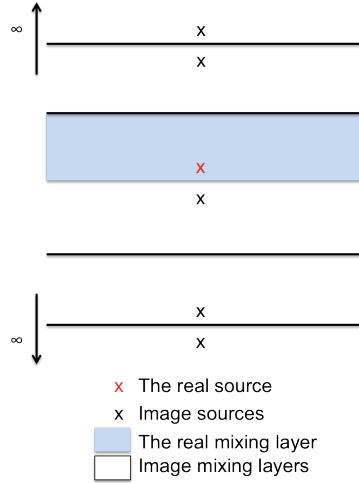


Figure 3: Image Sources

5.1 Gaussian Plume and Gaussian Puff

The advection and diffusion of an instantaneous release of mass M of pollutant from a point source in an infinite domain is modelled as a Gaussian puff:

$$C(x, y, z, t) = \frac{M}{(4\pi t)^{3/2} \sqrt{D_{xt} D_{yt} D_{zt}}} \exp \left(-\frac{(x - x_o - U_x t)^2}{4D_{xt} t} - \frac{(y - y_o - U_y t)^2}{4D_{yt} t} - \frac{(z - z_o)^2}{4D_{zt} t} \right) \quad (5.1)$$

Where x_o , y_o and z_o specify the point at which the release occurs. For continuous releases this can be integrated to give the Gaussian plume equation:

$$C(x, y, z) = \frac{Q}{4\pi (x - x_o) \sqrt{D_{yt} D_{zt}}} \exp \left[-\frac{(y - y_o)^2}{4D_{yt} \frac{(x - x_o)}{U}} - \frac{(z - z_o)^2}{4D_{zt} \frac{(x - x_o)}{U}} \right] \quad (5.2)$$

Equivalent equations can be found for line and plane sources by integrating over the z and then the y directions [22]. In atmospheric models, the diffusion coefficients are replaced by dispersion coefficients: $\sigma = \sqrt{2D_t}$. The equations given are for the infinite domain. In the finite domain, an infinite number of image sources gives the correct representation. For atmospheric models, the plume is constrained by the ground plane and the mixing layer, the correct representation is therefore an infinite number of image sources in the z direction. For river models, the plume is constrained by the river bed, free surface and the river banks, therefore an infinite number of sources in both the y and the z directions represent the system. This idea is illustrated in Figure 3.

5.2 Continuous atmospheric release

The continuous atmospheric release can model HT or HTO releases, both are detailed below.

5.2.1 HTO release

The annual average concentrations in a polar coordinate domain due to a continuous release are calculated using a sector averaged Gaussian plume equation. This equation is the same as the Gaussian plume discussed in subsection 5.1 but averaged over a sector of the compass:

$$C_{air_outdoor} = \frac{Q}{x\Delta\theta\sqrt{2\pi}} \sum_{ij} \left[\frac{F_{ijk}}{u_k\sigma_{zi}} (\text{exponential term}) \right] \quad (5.3)$$

The exponential term chosen depends the number of image sources included in the model. The number of image sources used in TERM can be varied depending on the accuracy required.

The newly developed surface runoff model calculates concentrations in water reservoirs. The cells over which the catchment area of a reservoir is spread are specified so that the air concentrations in these cells can be averaged. The average air concentration is then divided by the absolute humidity (L/m^3) to obtain an average value for the concentration in the rain. The total amount of tritium (in Bq) entering the reservoir per year is calculated by multiplying the annual rainfall with the runoff coefficient, catchment area and average concentration of rainfall. The number of turnover times per year is calculated from the lake volume and outflow information. The total amount of tritium into a reservoir in a year is then divided by this number and the lake volume to obtain the average concentration in the reservoir. The concentration in domestic water is set equal to that calculated for the reservoir.

Transfers from the atmosphere to various compartments are calculated via the following set of equations, which are the same as those in Hart [18].

$$\begin{aligned}
\text{Atmosphere to soil pore water:} \quad C_{spw} &= \frac{C_{air_outdoor} RF_{sw}}{H_{av_snow_free}} \\
\text{plant water:} \quad C_{plantHTO} &= \frac{C_{air_outdoor} RF_p (1-DW_p)}{H_{av_growing}} \\
\text{plant organic matter:} \quad C_{plantOBT} &= \frac{C_{air_outdoor} RF_p DW_p ID_p WE_p}{H_{av_growing}} \\
\text{animal water:} \quad C_{animalHTO} &= \frac{C_{air_outdoor} (1-DW_a) f_{wa}}{H_{av_annual}} \\
\text{animal organic matter:} \quad C_{animalOBT} &= \frac{C_{air_outdoor} f_{wa} DW_a ID_a WE_a}{H_{av_annual}} \\
\text{pond water:} \quad C_{pond} &= C_{air_outdoor} Bucket_ratio
\end{aligned}$$

The transfers between compartments in the terrestrial environment are mainly based on those in Hart [18]; however the transfer from plant water and plant organic matter to animal water and animal organic matter are split into 4 separate processes (in contrast to Hart [18]).

$$\begin{aligned}
\text{Soil pore water to well concentrations:} \quad C_{well} &= C_{spw} \exp \left[\frac{-\lambda Z_{tos} (porosity)}{q_{infil}} \right] \\
\text{Well water to animal water:} \quad C_{animalHTO} &= C_{well} k_{ww} f_{ww} (1 - DW_a) \\
\text{Well water to animal organic content:} \quad C_{animalOBT} &= C_{well} k_{ww} f_{ww} DW_a ID_a WE_a \\
\text{Pond water to animal water:} \quad C_{animalHTO} &= C_{pond} k_{pw} f_{ww} (1 - DW_a) \\
\text{Pond water to animal organic content:} \quad C_{animalOBT} &= C_{pond} k_{pw} f_{ww} DW_a ID_a WE_a \\
\text{Plant water to animal water:} \quad C_{animalHTO} &= C_{plantHTO} k_{fp} f_{wpw} \frac{(1-DW_a)}{(1-DW_p)} \\
\text{Plant organic matter to animal water:} \quad C_{animalHTO} &= C_{plantOBT} k_{fp} f_{wdw} \frac{(1-DW_a)}{(DW_p WE_p)} \\
\text{Plant water to animal organic matter:} \quad C_{animalOBT} &= \frac{C_{plantHTO} k_{fp} f_{wpw} ID_a DW_a WE_a}{(1-DW_p)} \\
\text{Plant organic to animal organic matter:} \quad C_{animalOBT} &= \frac{C_{plantOBT} k_{fp} f_{wdw} DW_a ID_a WE_a}{DW_p WE_p}
\end{aligned}$$

Some tritium in domestic water evaporates to increase the concentration of tritium indoors. The evaporated tritium is added to the outdoor air concentration to give the indoor

air concentration as below:

$$C_{air_indoor} = C_{air_outdoor} + (Andelman) C_{domestic} \quad (5.4)$$

Andelman is the Andelman volatilisation factor for radionuclide pollutants. The suggested value for ^{222}Rn is $0.1L/m^3$ and this is said to be reasonable for tritium (Napier et al. [13]).

The doses due to various intakes are calculated by multiplying the annual intake of a substance by its specific activity and a dose factor. The dose factor for inhalation of HTO is 1.5 times the dose factor for ingestion of HTO to take account of skin absorption. The dose factor for the ingestion of OBT is higher than for the ingestion of HTO. The dose factor for the ingestion of HT is less than for HTO and OBT because of reasons discussed in subsection 3.2.

5.2.2 HT release

The same methodology is applied for the atmospheric dispersion of a continuous HT release as in subsubsection 5.2.1. Different input parameters are required because some of the transfers differ from the HTO release case.

Conversion of HT to HTO in the atmosphere is via oxidation in the soil. This transfer process (deposition onto soil, oxidation and re-emission) is summarised into one transfer equation:

$$C_{airHTO_outdoor} = C_{airHT} R_{HT} f_{oxid} H_{av_annual} \quad (5.5)$$

Transfer of HT to plant is modelled as:

$$C_{plantHTO} = C_{airHT} C F_{HT} (1 - DW_p) \quad (5.6)$$

$$C_{plantOBT} = C_{airHT} C F_{HT} DW_p ID_p W E_p \quad (5.7)$$

The only tritium that remains in the soil is that due to the oxidised HTO, therefore the remaining transfers are the same as specified in subsubsection 5.2.1.

5.3 Discrete atmospheric release

For the instantaneous release, cartesian coordinates are used rather than polar coordinates.

5.3.1 HTO release

The release is assumed to occur instantaneously and is modelled as a Gaussian puff (see subsection 5.1). The dispersion coefficients are calculated using the Pasquill-Gifford (NRC) diffusion formulae [13] and depend on stability class and distance from the source. This is discussed further in subsubsection 6.3.2. The stability class also specifies the mixing height.

Although deposition to and re-emission from the terrestrial environment is modelled, the effect of this on the air concentrations is not taken into account. Consequently concentrations in the vicinity of the primary plume are over-estimated, while air concentrations are under-estimated once the plume has passed.

Once the atmospheric concentrations are calculated at each node for each point in time the terrestrial transfers are calculated. Various parameters are input to the model, and there are also parameters derived within the model. For example the vapour pressure, saturated vapour pressure and gradient of the vapour pressure curve are calculated from the temperature and relative humidity values.

Friction velocity is calculated using the Equation 5.8, an empirical expression valid for the flat plate boundary layer [26]. It is used as an approximation here:

$$u^* = \sqrt{0.0286} U_o \left(\frac{x}{\nu} \right)^{-0.1} \quad (5.8)$$

In the code, x is replaced by “distance travelled” given that U_o changes magnitude and direction with time. The Stanton number is as follows [10], assuming Pr is of order 1:

$$St_k = 0.8 Re^{*-0.2} Pr^{-0.44} \quad (5.9)$$

Aerodynamic resistance, boundary layer resistance and atmospheric resistance are calculated as in UFOTRI [35] and combined with the input values for soil and canopy resistance to give the total resistance of transfer to soil and grass and plants respectively. v_{HTO} can be defined as a constant in the parameter input file, or calculated as
$$v_{HTO} = \frac{1}{r_{aerodynamic} + r_{boundary.layer} + r_{soil}} .$$

The increase in soil concentrations of the first layer in a given time step is given by:

$$\Delta(C_{spw1}) = \frac{v_{dHTO} C_{air.outdoor}}{1000\theta_1 h_{s1}} \Delta t \quad (5.10)$$

The change in soil concentrations due to washout from the plume is modelled using:

$$\Delta(C_{spw1}) = \frac{(amount\ in\ plume\ above\ ground)(1 - \exp[-\Lambda\Delta t])}{1000\theta_1 h_{s1} \Delta x \Delta y} \quad (5.11)$$

The washout coefficient is defined as [35]:

$$\Lambda = (9 \times 10^{-5}) (\text{precipitation})^{0.6} \quad (5.12)$$

The amount in the plume above the ground is calculated by integrating over one Gaussian plume (not all images) in the z direction:

$$\text{vertically integrated} = \frac{M}{(2\pi) \sigma_x \sigma_y} \exp \left[-\frac{(x - x_o - U_x t)^2}{2\sigma_x^2} - \frac{(y - y_o - U_y t)^2}{2\sigma_y^2} \right] \quad (5.13)$$

This is the amount of activity above one point, this is then multiplied by the area of the cell to give an approximation for the amount of activity in the plume above that given cell:

$$\text{amount in plume above ground} = (\text{vertically integrated}) \Delta x \Delta y \quad (5.14)$$

UFOTRI uses the following equation for the concentration of tritium in plant water:

$$C_{\text{plantHTO}} = C_{\text{air}} \frac{\alpha}{H_{\text{sat}}} (1 - e^{-kt}) \quad (5.15)$$

Where $k = \frac{H_{\text{sat}}}{\alpha \mu r_{\text{plant}}}$. This equation was modified so that air concentration can change with time. The derivation and lines of code used are given in Appendix A. The same equation is used for transfer from the atmosphere to grass with the appropriate parameters.

Transfer to ponds is modelled using the same equation as the continuous release is used (subsubsection 5.2.1), and the pond assumed to come into equilibrium with the atmospheric air concentration instantaneously.

The transfer from the water content of a plant to the organic part is calculated via a photosynthesis model:

$$\Delta C_{\text{plantOBT}} = T_{\text{uniformHTOtoOBT}} (\text{Day Factor}) (\text{Development Factor}) C_{\text{plantHTO}} \Delta t \quad (5.16)$$

$$\Delta C_{\text{plantHTO}} = -T_{\text{uniformHTOtoOBT}} (\text{Day Factor}) (\text{Development Factor}) C_{\text{plantHTO}} \Delta t \quad (5.17)$$

$T_{\text{uniformHTOtoOBT}}$ is the uniform transfer rate from plant water to plant organic matter. The derivation of this is given in Appendix B. The “Development Factor” is a factor for the rate into OBT depending on the development stage of the plant [35]. If data is not available it can be set to 1. The “Day Factor” takes into account the increased amount of photosynthesis during the day compared to at night [35]. The same method is applied for grass.

Tritium is re-emitted from the soil. The change in soil concentration due to this process

is given by:

$$\Delta C_{spw1} = \frac{-E_{soil} C_{spw1} \Delta t}{1000 h_{s1} \theta_1} \quad (5.18)$$

Where E_{soil} is given by Monteith's bulk resistance formula for actual evapotranspiration as defined in Raskob [35]. θ for the three soil layers can be defined as constants if the time modelled is short, or can be input as a vector of size equal to the number of time steps with the value changing with time.

Three soil layers measuring 0.05m, 0.1m and 0.15m are modelled and cover the root zone in the soil. The top layer is 0.05m thick, the second 0.1m and the third is 0.15m. Transfer from layer i to j is given by:

$$V_{i,j} = k_{i,j} \left[1 - \frac{2(S_i - S_j)}{(h_i + h_j)} \right] \quad (5.19)$$

Where the suction tension is given by: $S_i = (1.5 \times 10^5) P^{(a+bP+cP^n)}$ and P is given by: $P = \frac{(\theta_{sat} - \theta_i)}{(\theta_{sat} - \theta_{wilt})}$. k is the hydraulic conductivity of the soil and given by $k_i = \frac{\alpha_q}{S^m + \beta}$. $V_{i,j}$ is the volumetric flux of water and is positive in the downwards direction. Equation 5.19 is the discretised version of Darcy's law [42]:

$$V = k \left[\left(\frac{dS}{dZ} \right) - 1 \right] \quad (5.20)$$

Where V is the volumetric flux of water (positive upwards), k is the hydraulic conductivity of the soil and S is the soil suction (expressed as a positive value), Z is the height above a predefined datum and 1 is the unit hydraulic gradient due to gravity [42]. This expression is discretised and multiplied by -1 to give the volumetric flux as positive in the downwards direction. The UFOTRI documentation [35] uses the same expression but without multiplying by -1, which gives $V_{i,j}$ as positive upwards. $V_{i,j}$ is the percolation velocity in mm/day and so this is converted to m/s in the TERM model.

Vegetation and grass take up water from the various soil layers at a rate that is determined by the transpiration from the canopy. The model used here is the same as that specified in the UFOTRI documentation [35] which specifies the percentage of water transpired taken up from each of the soil layers. Transpiration is calculated using Monteith's bulk resistance formula [35].

The modelling of cows includes transfers from grass and air to cow water, cow organic matter, inorganic contents of milk and the organic contents of milk, and transfers from the cow to the top soil layer. This is done in the same way as specified in the UFOTRI documentation [34] and is rather involved since all compartments depend on each other. Some of the transfer rates are given by observed data (eg. the half life of hydrogen in cow organic matter), others are determined assuming equilibrium conditions and equating

transfer quantities. Transfer from the air to cow via breathing, for example, can increase to levels much higher than other transfer processes if the air becomes humid with a high concentration of tritium. In these cases, the equilibrium balance can result in the transfer rates from grass to cow to become negative. In this case the transfer rate from grass to cow is set to zero, and the concentration in the cow will rise.

The concentration in well water is conservatively set to be the same as that in the third soil layer. Assuming all domestic water comes from wells, indoor atmospheric concentrations are calculated in the same way as in the continuous release scenario (subsection 5.2).

5.3.2 HT Release

This is very similar to the HTO release (subsubsection 5.3.1). The primary air concentrations due to the initial plume are calculated in the same way. Deposition velocity can be set to a constant value, or calculated using the equation given in the UFOTRI documentation [35], which depends on the diffusion of HT in air, temperature, tortuosity factor and water content of the soil. Washout of HT is assumed negligible. However, given that HT, once deposited on the soil, is oxidised to HTO, which is more dangerous if inhaled, modelling re-emission to the atmosphere is more important in this case.

The quantity re-emitted in each time step, above each cell, is calculated by summing the amount of evapotranspiration from soil, grass and plants and modelling this as a Gaussian puff release at ground level. To take account of the fact that the re-emission is from an area source and not a point source, the Gaussian is given initial values for the x and y dispersion parameters: $\sigma_{initial_x} = \frac{\Delta x}{4.3}$ and $\sigma_{initial_y} = \frac{\Delta y}{4.3}$. The factor 4.3 results from the geometry of the Gaussian plume, and is the value used in UFOTRI [34]. The actual σ_x or σ_y at any point is then calculated by combining $\sigma_{initial}$ with the values defined by Equation 6.2 in quadrature.

5.4 Continuous river release

There are three options of increasing complexity for modelling the concentration in a river due to a fluvial release. The simplest is the 1D model, followed by the 2D and then 3D models. The difference between the models is that the source is modelled as a plane, line and point source respectively.

5.4.1 Plane Source

The model for a continuous release from a plane source in a river is equivalent to assuming instantaneous mixing. The equation used is: $C_{river} = \frac{Q}{F}$.

5.4.2 Line Source

A continuous release from a line source in a river (ie. concentrations are constant with respect to vertical position). The concentrations are calculated using a Gaussian plume equation for a line source. A user-specified number of image sources in the transverse direction are taken into account because the plume is constrained by the river banks.

5.4.3 Point Source

The continuous release from a point source in a river is calculated using the Gaussian plume equation for a point source. A user-specified number of image sources are required in the transverse and vertical directions to take into account the fact that the plume is constrained by the river banks, bed and free surface.

5.5 Discrete river release

5.5.1 Plane Source

An instantaneous release from a plane source is modelled via a convection dispersion model, ie. the Gaussian Puff equation for a planar source is solved at various points along the stream-wise direction.

5.5.2 Line source

An instantaneous release via a convection dispersion model assuming a line source is modelled using the Gaussian Puff equation for a line source (the source is along the z -axis at position y_o in the y direction, specified by the user). Because the diffusion of the slug is not unconstrained (it is constrained by the banks of the river), a user-specified number of image sources are used.

5.5.3 Point Source

An instantaneous release via a convection dispersion model assuming a point source is modelled via a Gaussian Puff equation for a point source. A user-specified number of image sources in the y and z direction are needed since the plume is being constrained by the banks of the river, river bed, and the river's top surface.

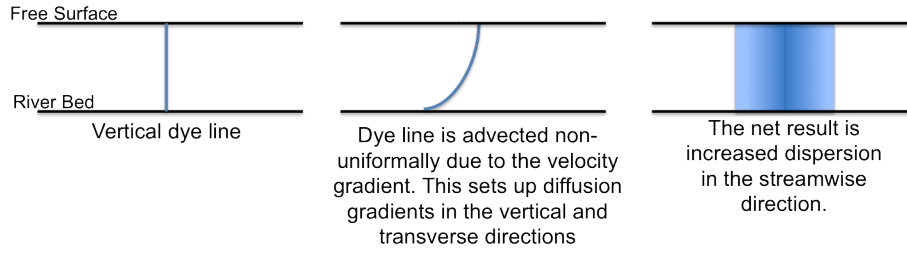


Figure 4: Concentrations in a river due to a continuous line source (Bq/L)

5.6 Determining the values of the diffusion coefficients (D_{xt} , D_{yt} and D_{zt})

A Manning coefficient suitable for the river under consideration is specified and the friction coefficient calculated using [8]:

$$u^* = \sqrt{\frac{9.81n^2U^2}{R_h^{1/3}}} \quad (5.21)$$

The friction velocity can then be used to calculate the turbulent diffusion coefficients as [8]:

$$D_{xt} = D_{yt} = 0.15hu^* \quad (5.22)$$

$$D_{zt} = 0.067hu^* \quad (5.23)$$

Dispersion is enhanced in the stream wise direction as a result of the velocity gradient present due to the no slip condition at the river bed (see Figure 4). The effective dispersion coefficient in the x direction is therefore equal to the sum of the turbulent diffusion coefficient defined in Equation 5.23 and the component due to the dispersion caused by the velocity gradient which is calculated as [8]:

$$D_L = 5.86hu^* \quad (5.24)$$

5.7 Continuous coastal release

The equation for calculating the concentration at a point due to a continuous release to coastal waters is taken from Hart's model [18].

$$C_{coastal_water} = \frac{Q\alpha_{coast}\beta_{coast}}{D_F Q_v} \exp\left(-\lambda \frac{x}{U_c}\right) \quad (5.25)$$

The dilution factor for the coastal model:

$$D_F = \left[\left(\frac{1000d\beta_{coast}\sqrt{\pi\kappa}}{Q_v U_c^{0.17}} \right)^{\frac{1}{1.17}} + D_o^{\frac{1}{1.17}} \right]^{1.17} \quad (5.26)$$

For $\beta_{coast}=0$ the dilution factor is simply D_o

The translation of these equations to an executable model that calculates concentrations over a cartesian domain can be seen in the DVD of supporting material (MATLAB script continuous-sc.m).

5.8 The contribution of the river and coastal models to dose

The fraction of the year spent swimming in the various water bodies by residents can be defined, and then doses due to skin absorption and accidental ingestion can be calculated. The concentration of tritium in aquatic plants is assumed to come into equilibrium with the surrounding water concentration, therefore the activity of the water content and organic matter of the aquatic plants are determined as follows [18]:

$$C_{aq.pHTO} = C_{water.body} (1 - DW_{aq.p}) \quad (5.27)$$

$$C_{aq.pOBT} = C_{water.body} DW_{aq.p} ID_{aq.p} WE_{aq.p} \quad (5.28)$$

The same equations are used for aquatic animals.

6 Model Validation

The atmospheric components of the model have been validated. The continuous HTO release has been compared to other models and measured data for a real release. The discrete HTO release has been compared to UFOTRI for a hypothetical release.

6.1 Continuous Atmospheric HTO Release

To validate this component of the model, the annual release of tritium from the SRB Technologies site in 2006 was considered. SRB Technologies Ltd. is a tritium light manufacturer in Pembroke, Ontario, Canada [11]. Modelling and monitoring investigations of the site were undertaken by EcoMetrix in 2008. Air measurements were taken using a passive sampling system and the modelling investigation was completed using the IMPACT code which is also a steady-state, sector-averaged Gaussian Plume model.

The IMPACT code used 2006 annual emissions and meteorological data [11]. The annual emission was available from EcoMetrix as 74,000 GBq/year [11]. Hourly wind direction and speed data in Ottawa (the location of the nearest weather station) for 2006 was obtained from Canada's National Climate Data and Information Archive [7] to input to the model. A MATLAB script was written to complete the following tasks:

- Assign each hourly wind speed into one of the six available wind speed classes defined in the TERM continuous atmospheric release model (subsection 5.2)
- Calculate the relative frequency of the wind speed classes in each of 16 sectors of the compass
- Multiply the joint wind speed and direction probabilities by estimated probabilities for stability classes. This gives the triple joint probability of a given stability class occurring with a given speed in a given direction (F_{ink})

The probabilities of stability classes were inferred from Table 3, to be of the order of the numbers given in Table 4. This highlights the difficulty of quantifying atmospheric stability for atmospheric models. The stability categories of Pasquill are useful, but it is often difficult to exclusively determine the stability class of a given set of conditions.

The effective release height was set to zero, given that there is no information available regarding the height of the release or whether the release would be susceptible to plume rise before being advected by the wind.

Table 3: Stability Categories of Pasquill [27]

Wind speed at 10m (m/s)	Day Solar Intensity			Night	
	Strong	Moderate	Slight	Overcast	Clear
0-2	A	A-B	B	-	-
2-3	A-B	B	C	E	F
3-5	B	B-C	C	D	E
5-6	C	C-D	D	D	D
>6	C	D	D	D	D

Table 4: Estimated probabilities for each stability class in each wind speed class.

Wind Speed Class (m/s)	A	B	C	D	E	F
1.5	0.5	0.5	0	0	0	0
2to3	0.0833	0.25	0.1667	0	0.25	0.25
3to5	0	0.25	0.25	0.25	0.25	0
3to5	0	0.25	0.25	0.25	0.25	0
5to6	0	0	0.25	0.75	0	0
morethan6	0	0	0.1667	0.8333	0	0

A graph showing the log of air concentrations as a function of sector and radial position is given in Figure 5. Similar graphs were found for the concentration in soil pore water,

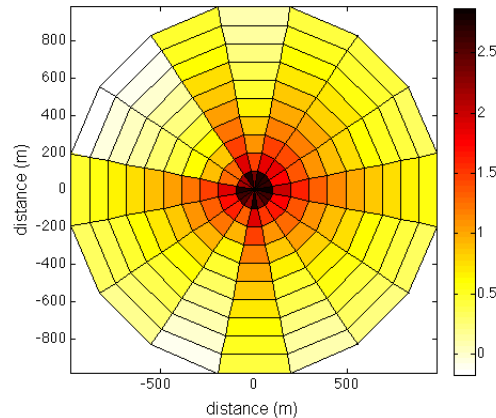


Figure 5: Air concentration assuming zero release height, 2006 meteorological data, and stability probabilities given in Table 4. Colour bar gives the log of air activity concentration (HTO) ($\log_{10}(Bq/m^3)$)

plant water, plant organic matter, animal water, animal organic matter, ponds, well water and domestic water concentration, indoor air concentrations and total annual dose.

Figure 6 compares the measured value of air activity at various distances from the source with those predicted by the TERM and IMPACT models. The TERM model predictions are 8% less than measured concentrations, on average, compared to 83% greater than measured concentrations, on average for the IMPACT model [11]. Approximately 89% of TERM model predictions are within a factor of 2 of the measured concentrations, compared to 60% of model predictions in the case of the IMPACT model [11].

The main source of uncertainty of the input data is in the stability classes. No information is given in the literature regarding the stability class input to the IMPACT model. The TERM model was run again assuming the stability to be class D throughout the year and then the same for class F to bracket the worst case scenarios. The results normalised by measured values are given in Figure 7.

The continuous HTO atmospheric release component of the TERM model is used for the Wylfa case study (subsection 7.1). However, the wind rose available for the site has 12 sectors rather than 16. The above analysis was repeated by assigning all wind directions to 12 sectors rather than 16. The results normalised by the measured values are shown in Figure 8.

The IMPACT and HART models (equivalent to TERMb) were compared with various versions of the TERM model, the difference between each TERM model being the number of image sources used.

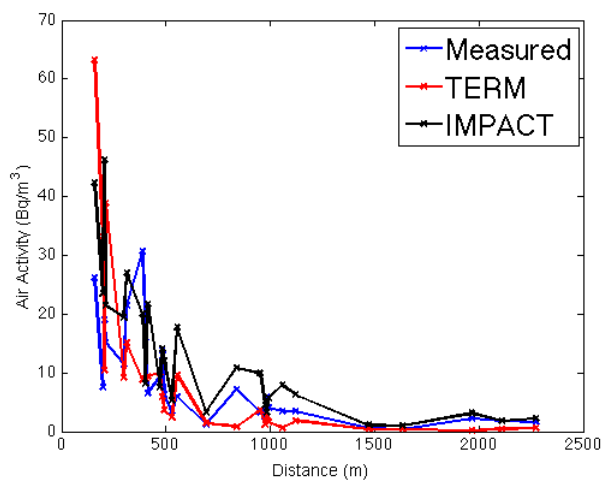


Figure 6: Comparison of TERM using estimated stability class probabilities with measured values and IMPACT model results.

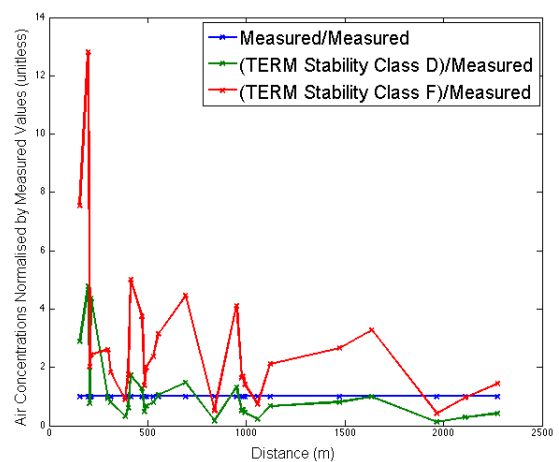


Figure 7: Comparison of TERM assuming stability class D and stability class F with measured values.

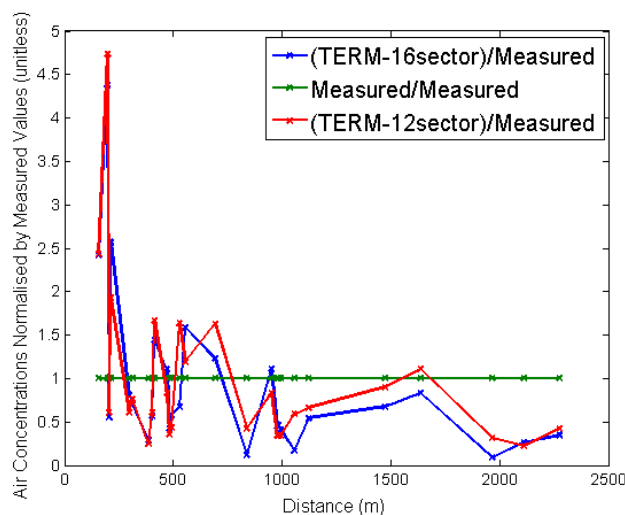


Figure 8: 12 and 16 sector model comparison. (with estimated stability classes from Table 4)

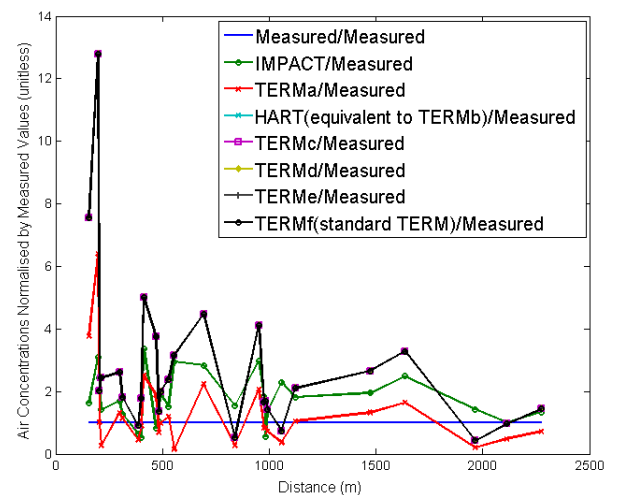


Figure 9: Model Comparison. HART and all TERM models run assuming stability class F. TERMa=no image sources, TERMb=1 image source, TERMc=3 image sources and then in increments of 2 up to TERMf (standard TERM) which has 9 image sources.

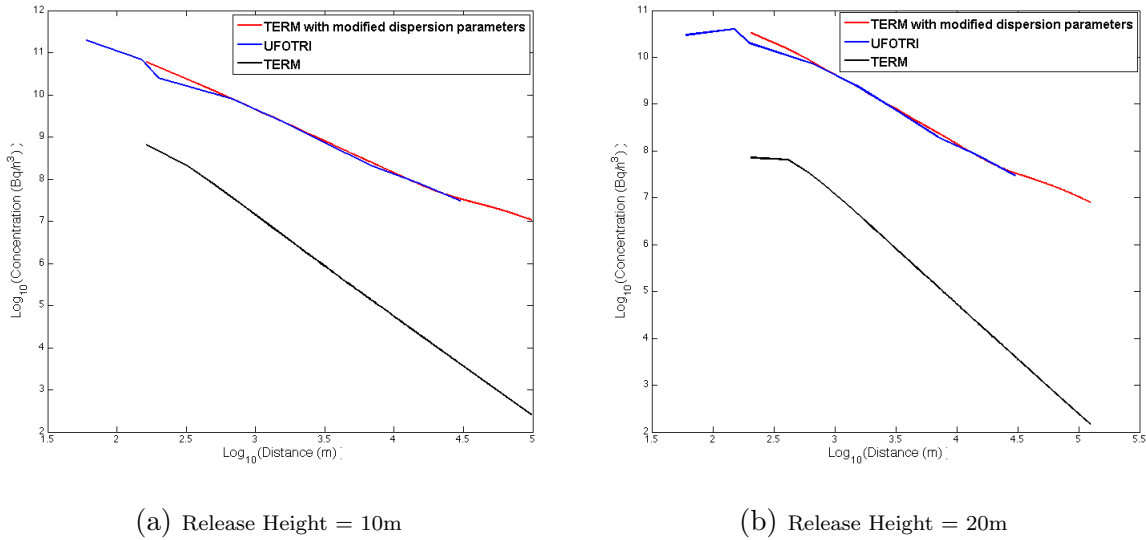


Figure 10: Comparison of TERM and UFOTRI results for a hypothetical release.

6.2 Discrete Atmospheric HTO Release

To validate this model, the atmospheric concentrations predicted by TERM were compared to those predicted by UFOTRI for a hypothetical release [34]. Details of the release can be found in Raskob [34]. The documentation for the hypothetical release states that the release quantity is 100g of HTO. The activity of the 100g is calculated to be $2.072 \times 10^{13} Bq$ given the assumptions of Appendix C. This quantity of release and the same atmospheric conditions as the hypothetical release were input to TERM. The results given by TERM are compared to those given by UFOTRI for release heights of 10m and 20m in Figure 10. The results for the same release amount with the dispersion coefficients of TERM set to develop in the same way as for UFOTRI are shown as the red line in Figure 10.

6.3 Discussion of model validation

6.3.1 Continuous Atmospheric Release of HTO

IMPACT is also a sector averaged gaussian plume model, and therefore should give similar results to the continuous atmospheric release component of TERM. It is expected that all models will overestimate measured values of concentrations in the near field due to the assumption that the release height is at ground level. TERM's atmospheric results showed the same general trend as measured values and the IMPACT model Figure 6. As can be seen in Figure 6, TERM's results significantly overestimate the air concentrations near the release ($< 250m$), as expected, but underestimate further downstream. Estimates

in radioactive release models need to be conservative, and this model does not meet this requirement with these inputs. The fact that 89% of TERM model predictions are within a factor of 2 of the measured concentrations compared to 60% of model predictions suggests that the TERM model is more accurate. However, concentration predictions were not conservative, given that the predictions are on average 8% less than measured. By contrast, IMPACT predicts concentrations at 83% higher than measured. Discrepancies between the IMPACT model and TERM could be because of the different assumptions made regarding the stability classes.

It was noted in subsection 6.1 that there is a high level of uncertainty in the stability classes, and that the true stability classes throughout the year could be quite different from those input to the model. Assuming a constant annual stability of class D and the same for class F gives two sets of concentrations that should bracket the worst case scenarios. As can be seen from Figure 7 the two cases straddle the measured values. This is evidence for a reliable model. Assuming stability class F throughout gives the most conservative estimate of activity.

The results predicted using a 12 sector and 16 sector wind rose agree reasonably well, with the 12 sector model performing slightly better than the 16 sector model at large distances. This suggests that at this level of discretisation, small variations in the number of sectors do not make very much difference in predicted concentrations. The model should be appropriate for use with the annual average Wylfa release, for which the wind data is only available in the form of a 12 sector wind rose (subsection 7.1).

Figure 9 shows the results for many different models normalised by the measured values. All versions of the TERM models and the HART model (which is equivalent to TERMb) were run assuming a constant stability class throughout the year of F for conservative estimates. TERMa assumes no image sources, TERMb assumes reflection in the ground plane only, TERMc has 3 image sources, TERMd has 5 image sources and so on until TERMf which has 9 image sources. The results for TERMb (HART) through to TERMf (the standard TERM model) are the same; the effect of the image Gaussians on the ground air concentrations are negligible due to their large distance from the ground plane. One image source is sufficient for this particular case.

6.3.2 Discrete Atmospheric Release of HTO

The concentrations predicted by UFOTRI are much higher than those of TERM. The reason behind this is that the method for obtaining the dispersion coefficients in TERM is different from that in UFOTRI. The two schemes are summarised below (both select various coefficients depending on the stability class. The values for the constants written below are for stability class D: the stability class used in this test case).

TERM Diffusion Coefficients

The dispersion parameters are calculated using the Pasquill-Gifford (NRC) diffusion formulae [13]. σ_y and σ_z are calculated by:

$$\sigma_y = 0.1471x^{0.9031} \quad (6.1)$$

$$\sigma_z = A_z x^{B_y} + C_z \quad (6.2)$$

Where A_z , B_y and C_z are values that depend on distance in the following manner:

Table 5: Pasquill-Gifford (NRC) diffusion formulae coefficients

Coefficient	Range of distance	Value
A_z	$x < 100m$	0.079
	$100m < x < 1000m$	0.222
	$x > 1000m$	1.26
B_z	$x < 100m$	0.881
	$100m < x < 1000m$	0.725
	$x > 1000m$	0.516
C_z	$x < 100m$	0.0
	$100m < x < 1000m$	-1.7
	$x > 1000m$	-13

Because σ_x and σ_y are in the same plane, they are assumed equal throughout.

UFOTRI Dispersion Coefficients

σ_y and σ_z are calculated as follows [35]:

$$\sigma_y = 0.418x^{0.796} \quad (6.3)$$

$$\sigma_z = 0.52x^{0.711} \quad (6.4)$$

The following constant value is then calculated:

$$C = \frac{2}{3} \tan(6.9) \quad (6.5)$$

When $\sigma_y \leq C$, then $\sigma_x = \sigma_y$. When $\sigma_y \geq C$, then $\sigma_x = C$.

Setting σ_x to a constant after $\sigma_y = C$ limits the diffusion. For stability class D, this occurs at a distance of 0.1266m in the wind direction. This explains the discrepancy in the results shown in Figure 10 because σ_x is set to a constant in UFOTRI after a distance of 0.1266m (before any values are calculated for Figure 10), whereas in TERM, σ_x continues to grow in the same way as σ_y . Because of this, the downwards slope in concentration with distance will be much bigger for TERM than UFOTRI since the overall dispersion is larger. This effect dominates the plume depletion modelled by UFOTRI.

7 Case Studies

7.1 Wylfa

Wind data is obtained from Valley, an airbase on the Isle of Anglesey. The distance from Valley to Wylfa is about 15km, and there are no large topographical structures in between, therefore the weather at this station should be representative of that at the power station. Triple joint frequencies were found by estimating the probabilities of various wind classes in particular directions from the wind rose shown in Figure 11, then multiplying these by crude probabilities of stability classes associated with certain wind speeds (see Table 4).

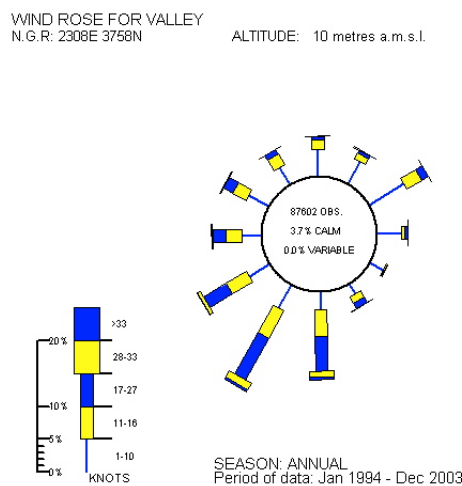


Figure 11: Wind Rose for Valley [32]

A rough estimate for humidity data was obtained by using the average value for Manchester [33] which is 86%. The atmospheric temperature is required for conversion to absolute humidity. The median of the yearly average high and low temperatures is 9°C, and a table for converting between relative and absolute humidity [37] gives $0.008L/m^3$. This is the value used for all three humidity values required for the continuous atmospheric model.

Water on the island is provided by surface water reservoirs. There are no operational wells on Anglesey, therefore values for parameters such as length to top of screen, porosity and infiltration rate are not needed. Other input parameters and their sources are specified in Appendix D. Parameters which specify the fraction of various intakes taken from the contaminated source were all set to 1 for a conservative estimate.

Water from the island's Alaw and Cefni reservoirs supply water in conjunction with some water from the main land, which can be a "blend of water from Cwellyn, Ffynnon Llugwy and Machlyn Bach reservoirs" (see supporting DVD for an email from the Water Resources Manager of Welsh water confirming this). A conservative estimate would be to assume that all domestic water supply is from the reservoir nearest to the station (Llyn Alaw) which is approximately 8km from the power station at an angle of 150° from the northerly direction at the power station. Catchment data is, however, only known for the Cefni reservoir, which is 19.25km away, in the same sector, and will be subject to more diluted rainfall over its catchment.

Assuming 100% of the water on the island is provided by the Cefni Reservoir. The code gives an activity value of $7.07 \times 10^{-2} Bq/L$ for drinking water in 1996. This result is much lower than current (2012) concentrations of tritium in drinking water on the island. Concentrations of the order of 7Bq/L are measured for the water provided to the south of the island and 8Bq/L for the water provided to the North of the island [6]. Very crude estimates for Llyn Alaw must therefore be made to obtain more realistic results.

The area of Llyn Alaw is $3.6 km^2$ and the maximum depth is 5.2m. If an average depth of 2.5m is assumed, the volume of the lake is $9 \times 10^6 m^3$. If the catchment area and turnover time is assumed to be the same as that for Llyn Cefni (very crude approximation) then the average surface runoff coefficient is calculated to be 0.77 (assuming a value of 0.35 for the soil [40] and 1 for the reservoir itself). The model gives an average concentration of $4.90 \times 10^{-1} Bq/L$. This is also lower than the amounts found in drinking water on the island today.

Crude approximations for the stability classes have been used, and this was seen to produce errors in subsection 6.1; in particular, it underestimated concentrations further downwind of the release. Therefore to estimate an upper bound on concentrations the model was run assuming a constant stability class of F throughout the year. This gives a concentration in the lake of $1.82 Bq/L$. Assuming all rainfall is from the point closest to Wylfa on a radius enclosing the area equal to the assumed catchment area, the result of the model is $3.11 Bq/L$. However, the turnover time is assumed the same as that of the Cefni reservoir which is 51 days (calculated from the data available at the Natural Environment Research Council [14]). The value for Llyn Alaw may be much larger. Assuming a turnover time of one year gives $4.48 Bq/L$ as the concentration in the reservoir. Accounting for a background radiation level of about $1.7 Bq/L$ (assuming the background activity of tritium in water in the Northern Hemisphere is relatively constant and using Canadian data [11]), the concentration in the water will be $6.18 Bq/L$ this is within 23% of the concentration in north Anglesey's drinking water and 12% of the concentration in south Anglesey's drinking water.

The concentrations in various compartments due to the Wylfa release, (assuming stability F and water concentrations of $4.48 Bq/L$), can be seen in Figure 12. During the year of 1996, the largest dose is estimated to be of the order of $4.2 \times 10^{-4} mSv/year$.

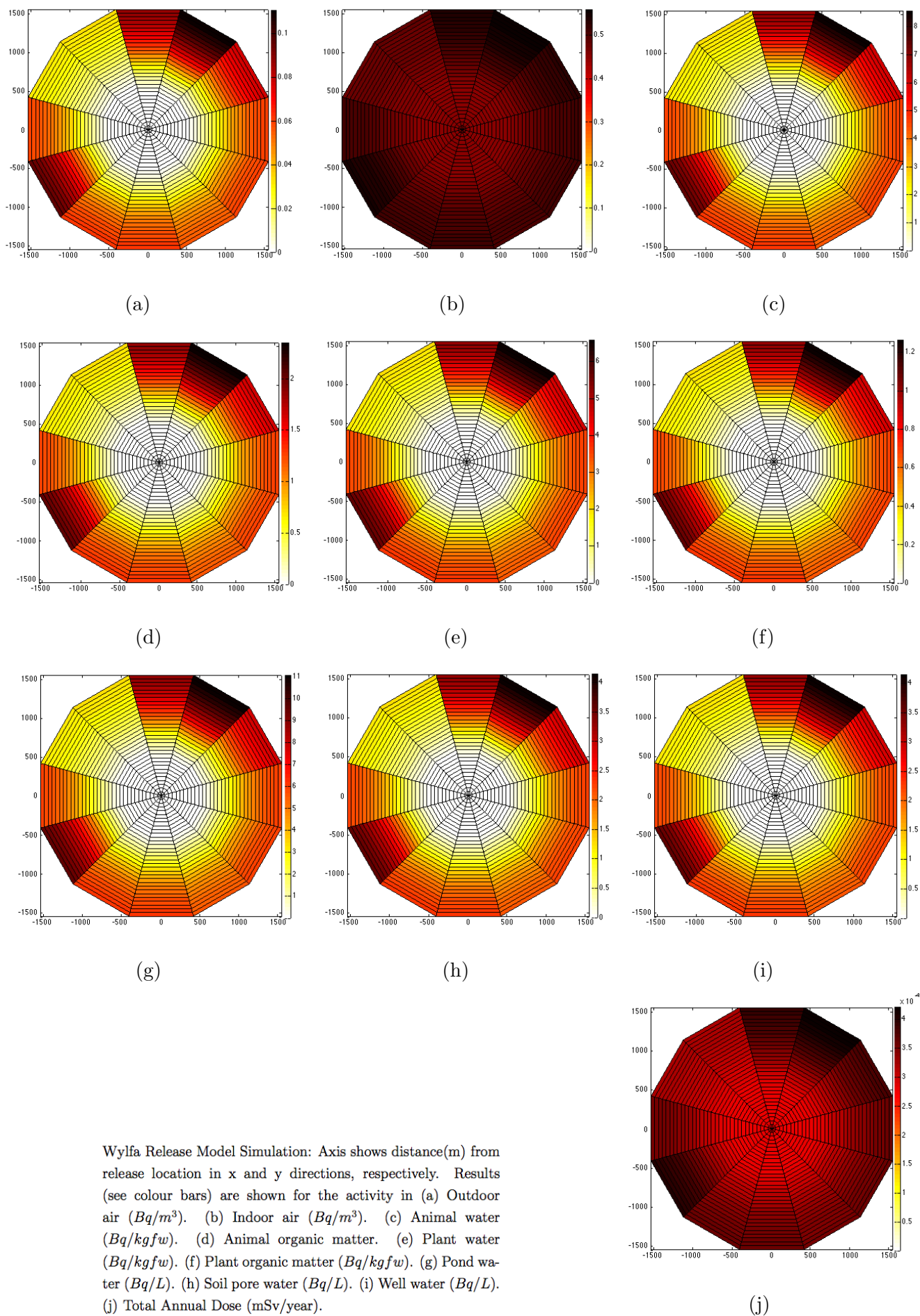


Figure 12

Table 6: Ottawa Valley Release Statistics [12]

Quantity released (kg)	Quantity released (Bq)	Fate	Model to be used
4.5	4.5×10^{12}	Vented almost immediately into the atmosphere as steam (HTO)	Discrete atmospheric release of HTO
28.5	28.5×10^{12}	Pumped to an on-site treatment facility for radioactive liquids, and subsequently all released to the Ottawa River in the form of “controlled releases” between December 2008 and February 2009 [4]	As it is unclear as to how this was released, the two extreme scenarios will be analysed: a discrete release to a river all on one day, and a steady state model assuming a release rate of 3.6651×10^{-6} Bq
14	14×10^{12}	Recovered and stored in sealed drums	Not modelled because not released

7.2 Chalk River Laboratories 2008 release

A discrete release from the National Research Universal (NRU) reactor at Chalk River laboratories occurred on December the 5th 2008. 47kg of heavy water leaked from the core of the NRU reactor, the fate of which is noted in Table 6. The event was considered to be “without safety significance” [5].

7.2.1 River Release

The width of the river at the point of release was estimated from maps to be 375m. The water level was taken from the Ottawa River Regulation Planning Board website [3], as the mean monthly average level at Pembroke (the closest river level monitoring station to the Chalk River site), at approximately 110m. The streamflow data is $1890m^3/s$ [3].

Britannia treatment plant is near Ottawa, about 200km downstream of Chalk River Laboratories. The maximum concentration of tritium measured at the plant during 2009 was 16.6Bq/L [31].

As discussed in section 5, 6 possible models for a river release are available. Given the large distance between Chalk River and the treatment plant, the plane source models are most appropriate. They are also the least conservative (compared to the line and point source models) and so are a lower bound. This is because instead of modelling the source as a point source, the pollutant is assumed instantaneously fully mixed in the cross-section, and therefore more highly diluted at the offset.

Discrete Release assuming a plane source.

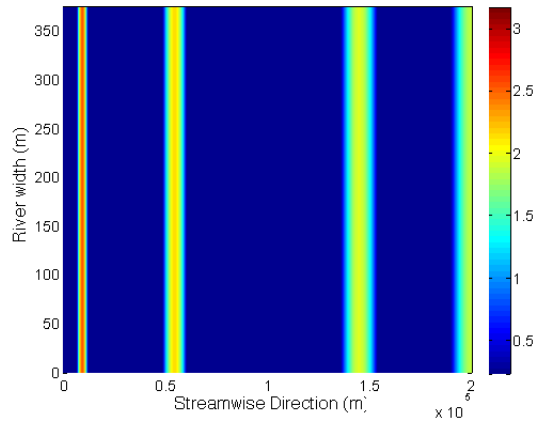


Figure 13: Graphical representation of the model assuming a discrete release from a plane source to a river. Each vertical line represents the slug at different stream wise distances. The increased amount of spreading that has occurred at larger stream wise distances is clear by the increase in the width of the slug. Colour bar gives concentration in $\log_{10}(Bq/L)$

The model results are illustrated in Figure 13, by showing the concentration distribution in the river at 4 different times.

The centre of the slug reaches Ottawa (the location of the Britannia treatment plant) at about 2km downstream of the release. The blue line shows the variation of plume-centre concentration with downstream distance from the source. The maximum concentration of tritium measured at the plant during 2009 is plotted on the graph for comparison. The green line on the graph shows the results from the same model, but assuming an effective width of 1790m instead of 375m.

Assuming a river width of 1790m, a discrete release of $3 \times 10^{10} Bq$, gives the results in Figure 15.

Continuous Release assuming instantaneous mixing.

Assuming steady state is achieved with a constant release rate of $3.88 \times 10^6 Bq/s$, the concentration of tritium in the river will be $3.75 Bq/L$.

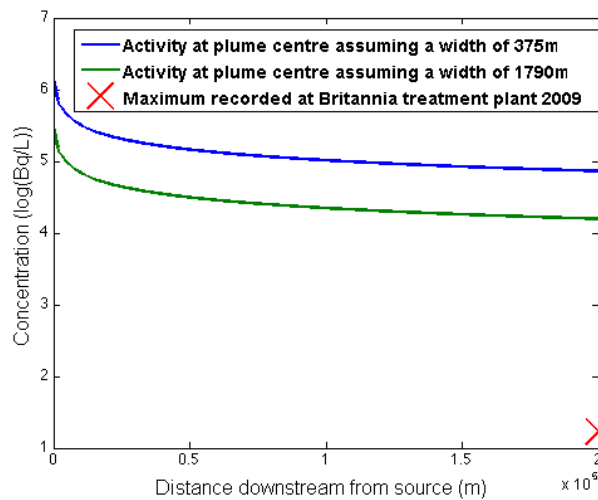


Figure 14: Planar discrete release predicted slug center concentrations

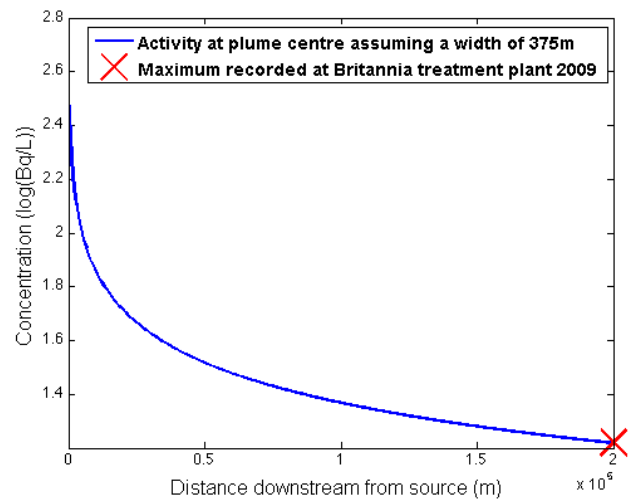


Figure 15: Results assuming a river width of 1790m and a release of $3 \times 10^{10} Bq$.

7.2.2 Atmospheric Release

Many parameters are required to model the discrete atmospheric release. These, and the sources for each, are summarised in Appendix E.

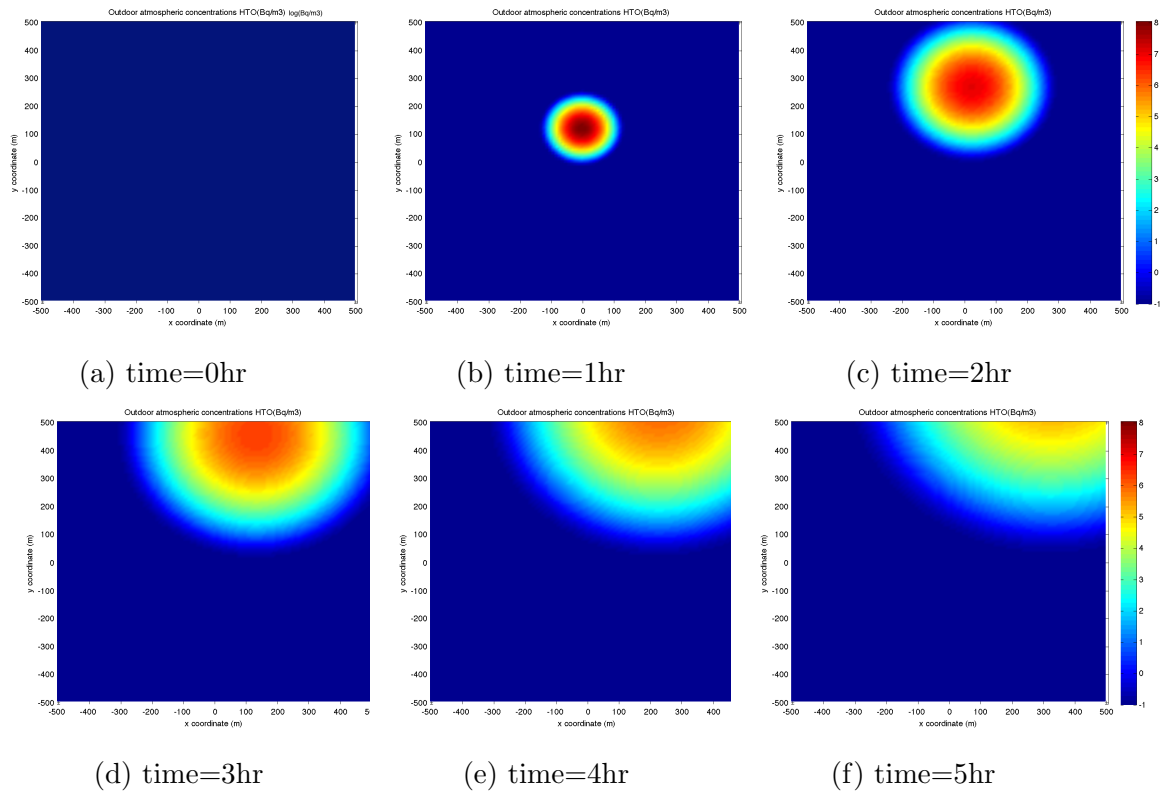


Figure 16: The air concentrations predicted by TERM for a discrete release of 4.5TBq. Colourbar in $\log_{10}(Bq/m^3)$

Illustrations of the results are given in Figure 16, Figure 17, Figure 18 and Figure 19. Results for all the compartments accounted for in the discrete release model (subsubsection 5.3.1) were obtained, but space restrictions mean they cannot be shown here.

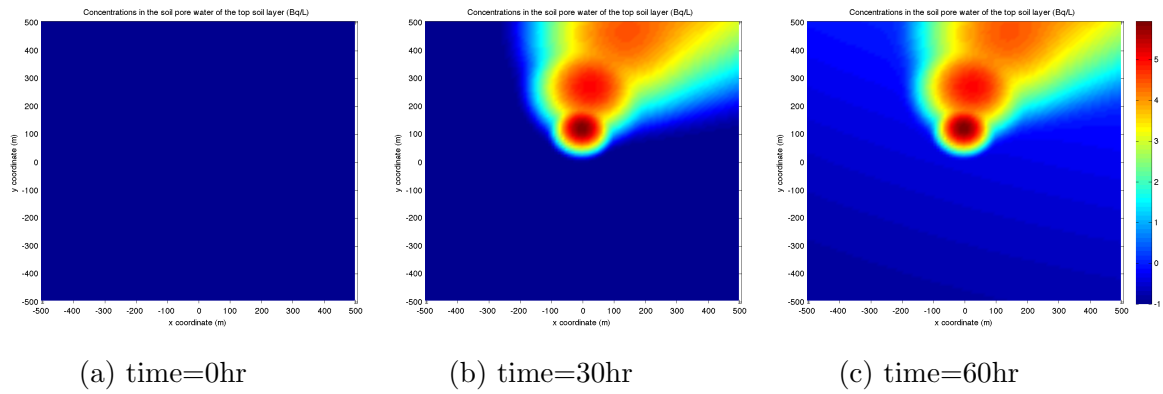


Figure 17: The soil concentrations in layer 1 predicted by TERM for a discrete release of 4.5TBq (Colourbar in $\log_{10}(Bq/L)$)

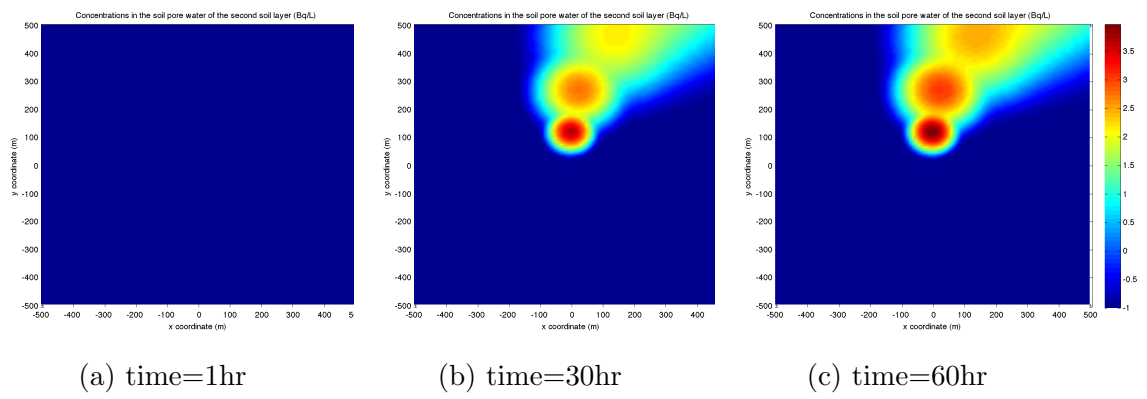


Figure 18: The soil concentrations in layer 2 predicted by TERM for a discrete release of 4.5TBq (Colourbar in $\log_{10}(Bq/L)$)

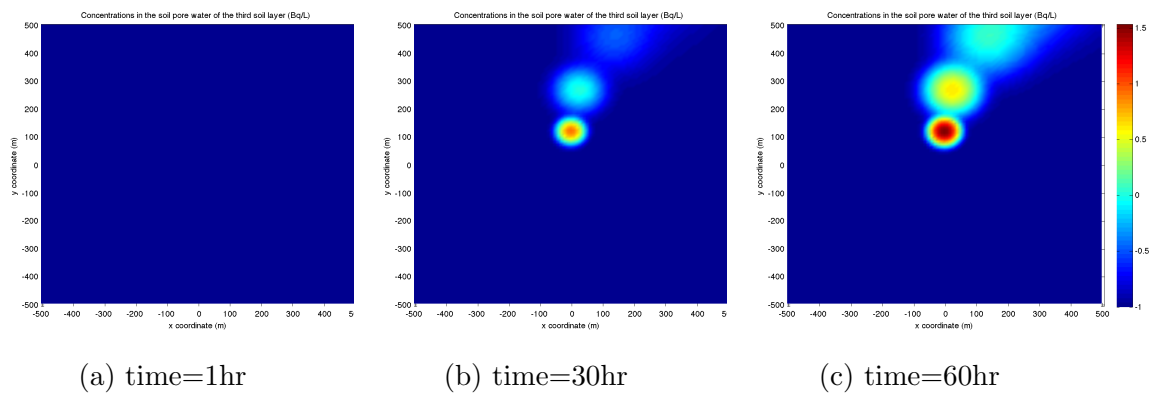


Figure 19: The soil concentrations in layer 3 predicted by TERM for a discrete release of 4.5TBq (Colourbar in $\log_{10}(Bq/L)$)

7.3 Case Study Discussion

7.3.1 Wylfa

The large discrepancy in results between the measured activity in drinking water and the model prediction for reservoir activity could be due to many reasons discussed below:

Errors in weather input data: The weather data obtained from the wind rose of Figure 11 is likely to have errors, as the values for wind speed were obtained by measuring by hand from the graph. If absolute quantities of wind speed and direction were available as in subsection 6.1, then the results might be more accurate. As previously discussed, the uncertainty in stability classes may also affect the results.

Errors in lake and catchment area data: Crude estimations of average reservoir depth, catchment area and reservoir turnover time were required.

Unknown background radiation: This is not known, but is estimated to be of the order of that in Canada, because both places are in the northern hemisphere. A more accurate value would, however, be beneficial in determining whether the difference between modelled and measured values is due to background radiation alone, uncertainty in input data or the model structure itself.

Groundwater contribution: Groundwater may have different concentrations to precipitation. Groundwater is likely to feed the reservoir as well as direct runoff. Anglesey was affected by nuclear fallout as a result of the Chernobyl disaster in 1986. Even though it is 26 years since the disaster, and therefore just over 2 tritium half-lives, if significant amounts were deposited and percolated to groundwater then the contribution from groundwater feeding the reservoir could be significant. Further work would be required to validate this claim.

Releases in 1996, but drinking water concentrations taken in 2012: The difference in time between measurements may make a big difference. However, releases of tritium from Wylfa have reduced since 1996 [23].

Possible effects of other releases: Other releases, for example, those from Trawsfynydd power station could be affecting the concentration of the drinking water. Some of the water reservoirs used to supply Anglesey from the mainland were in areas severely effected by the Chernobyl disaster.

The maximum predicted total dose of $4.2 \times 10^{-4} mSv$ is orders of magnitude below the limit of 1mSv per year for members of the public. The model input data was, however, quite uncertain and a more accurate figure could be obtained with better weather and catchment area data.

7.3.2 Chalk River Release to the Ottawa River

The model used for the blue line in Figure 14 is overestimating the value of the concentration at the treatment plant by several orders of magnitude. A possible reason for this is that the river is not 375m wide throughout (as modelled), and wider sections will allow more dispersion. The river meanders and this may cause increased dispersion at localised spots due to the secondary flows induced.

A limitation of the model is that the width of the river cannot be varied along its path length, which results in problems when the river stretches over large distances over which the river characteristics change significantly (as in this case). The widest part of the Ottawa river: Lac Deschênes lies between Chalk River and Ottawa at a width of 3.2km.

The results assuming a larger constant width of 1790m are also orders of magnitude larger than measured values (Figure 14).

The value predicted by the continuous release is much lower than the maximum measured at the Britannia treatment plant. However, the correct answer is straddled by the two approximations. This shows that TERM can be used to provide upper and lower bounds for river concentrations even when a site is a long way from the source, with a complex river system in between and uncertainty in release details.

A discrete release of $3 \times 10Bq$ gives reasonable agreement with the measured value at the treatment plant, as can be seen from Figure 15. This suggests that this amount is of the order of the maximum “controlled amount” released to the river. This implies 950 discrete “controlled” releases over the three months giving an average number of releases per day as 11, which is rather a lot. The value used for width is probably still too conservative, and a larger value may give more reasonable results.

7.3.3 Chalk River Release to the atmosphere

Figure 16 illustrates the plume diffusing and being advected by the wind. This shows that the model is useful as a tool for mapping the unsteady evolution of concentrations. Figure 17, Figure 18 and Figure 19 show the concentrations in the 3 soil layers, and the delay time in the infiltration of tritium to the third soil layer.

8 Conclusions and Further Work

The TERM model was built so that it can model HTO and HT releases to the atmosphere as continuous or discrete. The discrete release can be modelled as a point or area source

by specifying initial values for the dispersion parameters in the Gaussian puff equation. Discrete and continuous releases of HTO to rivers can be modelled as releases from three different source types (point, line and plane source). To model the continuous release of HTO to coastal waters, Hart's techniques were translated to an executable model. A novel runoff model was added to the continuous HTO release component of TERM.

The continuous and discrete atmospheric HTO release components of the model were validated by comparing results to measured data, and the results of other models. The reasons for differences between the TERM and IMPACT results for the continuous release case are suspected to be because of difference in the stability classes input to the models. TERM gave conservative estimates of concentration when the stability class was assumed as F throughout. For better results, more accurate information regarding atmospheric stability would be required. The difference between the TERM and UFOTRI results for the discrete release case are explained by the difference in the method for determining the dispersion parameters.

Once validated TERM was applied to two case studies: the steady state atmospheric release of HTO from Wylfa power station and an accidental release from Chalk River laboratories. The Wylfa release showed the ability of the model to calculate concentrations in various different terrestrial compartments due to a continuous release of HTO. The annual average doses received at various locations were found, and the maximum average dose found to be orders of magnitude less than the annual limit of 1mSv to members of the public. The Chalk River release showed the advantage of being able to model both discrete and continuous releases to a river, an option not available in the existing models reviewed. Given that only vague details of the release were available, using both the discrete and continuous models gave upper and lower bounds for concentrations that straddled the true amount measured at the treatment plant. The advantage of being able to model releases to more than one environmental compartment is also shown in the Chalk River case study, since the one accident (heavy water leak) resulted in both a release to the atmosphere and the river.

The model structure is flexible. Some values (for example deposition velocity of HTO and HT to soil) can be set to a constant, or calculated by equations specified in the code which depend on the surrounding conditions. Values of the water content of the three soil layers can be set to a constant for short releases, or can be input as a vector of values, where each element represents the soil moisture at a particular time, if the data is available.

There are two areas for possible further work. The efficiency of the code could be improved so that larger domains can be accommodated with more practical execution times. Further validation of the coastal release and the addition of a discrete release model to coastal regions would also be a valuable addition to TERM.

References

- [1] United States Environmental Protection Agency. Understanding variation in partition coefficient, k_d , values. Volume 2. (EPA 402-R-99-004B), August 1999.
- [2] Gauthiers D. Camus H. Caput C Belot, Y. Prediction of the flux of tritiated water from air to plant leaves. *Health Physics*, 37:575–583, 1979.
- [3] Ottawa River Regulation Planning Board. Historical water level and streamflow summary. <http://www.ottawariver.ca/emain.htm>.
- [4] Mike Buckthought. *Tritium on Tap: Keep radioactive tritium out of our drinking water*. Sierra Club Canada, 2009.
- [5] Canadian Nuclear Safety Commission (CNSC). Minutes of the canadian nuclear safety commission (cns) meeting held thursday february 19, 2009. <http://nuclearsafety.gc.ca/eng/commission/pdf/2009-02-19-Minutes-final-e-ED0CS-3373570.pdf>.
- [6] Dŵr Cymru. Drinking water quality. http://www.dwrcymru.com/public_dwr/zonedetailsinfo.asp.
- [7] National Climate Data and Information Archive. Hourly data report. http://climate.weatheroffice.gc.ca/climateData/menu_e.html?Prov=QC&StationID=4003&Year=2012&Month=5&Day=12&timeframe=1.
- [8] Cambridge University Engineering Department. Module 3d5 data card.
- [9] Marcello Donatelli, Gianni Bellocchi, Roberto Confalonieri, Ephrem Habyarimana, Ephrem Habyarimana, Marcello Donatelli, and Laura Carlini. Evapotranspiration software website. <http://agsys.cra-cin.it/tools/evapotranspiration/help/>.
- [10] Dr.N.Swaminathan. 3a6: Heat and mass transfer. forced convection. *Cambridge University Lecture Notes*, 2012.
- [11] ECOMETRIX and RWDI Air Inc. Investigation of the environmental fate of tritium in the atmosphere. *Canadian Nuclear Safety Commission*, (INFO-0792), 2009.
- [12] G. Edwards. Background: What we know about tritium in the ottawa valley. Technical report, 2009.
- [13] Napier et al. GENII Version 2 software design document. (PNNL-14584), November 2004.
- [14] Centre for Ecology and Natural Environment Research Council Hydrology. 102001-cefni at bodffordd. http://www.ceh.ac.uk/data/nrfa/data/time_series.html?102001.

- [15] Office for Nuclear Regulation and Environment Agency. Generic design assessment progress report. reporting period 1 april 2011 - 30 june 2011. (Quarterly Report 2 2011), July 2011.
- [16] J.R. Gat. *Isotope Hydrology, A Study of the Water Cycle*, volume 6 of *Series on Environmental Science and Management*. Imperial College Press, 2010.
- [17] Joachim Gruber. High-level radioactive waste from fusion reactors: The problem of alloy constituents and impurities.
- [18] D. Hart. Derived release limits guidance. (COG-06-3090-R2-I), November 2008.
- [19] P. Harteck. The relative abundance of HT and HTO in the atmosphere. *The Journal of Chemical Physics*, 22(10):1746–1751, 1954.
- [20] ITER. Tritium breeding. <http://www.iter.org/mach/tritiumbreeding>, 2011.
- [21] D.J. Jacobs. Investigation of the environmental fate of tritium in the atmosphere. (USAEC Report No. TID 24635), 1968.
- [22] Rebecca Jeffers. Tritium in the environment. October 2011.
- [23] Rebecca Jeffers. *Modelling of tritium cycling in the water environment under uncertainty*. Cambridge University Engineering Department, January 2012.
- [24] ANL (Argonne National Laboratory). Tritium (hydrogen-3). evs human health fact sheet. <http://www.evs.anl.gov/pub/doc/tritium.pdf>, 2005.
- [25] UK Government Legislation. The ionising radiations regulations 1999. http://www.legislation.gov.uk/ukxi/1999/3232/pdfs/ukxi_19993232_en.pdf, 1999.
- [26] Prof. E. Mastorakos. 4a12: Turbulence and vortex dynamics. part ii: Turbulence. *Cambridge University Lecture Notes*, 2012.
- [27] Prof. E. Mastorakos. 4a8: Environmental fluid mechanics, dispersion of pollution in the atmospheric environment. *Cambridge University Lecture Notes*, page 41, 2012.
- [28] R.L. Michel. Tritium in the hydrologic cycle. In Pradeep K. Aggarwal, Joel R. Gat, and Klaus F.O. Froehlich, editors, *Isotopes in the Water Cycle*, pages 53–66. Springer Netherlands, 2005.
- [29] J.K. Miettinen. Transfer and uptake mechanism of tritium in soil. In *Behaviour of Tritium in the Environment*, San Francisco, 16-18 October 1978.
- [30] Department of Energy and Climate Change. National policy statement for nuclear power generation. (EN-6), July 2011.

- [31] City of Ottawa Website. Tritium in drinking water. http://ottawa.ca/en/env_water/water_sewer/water_wells/quality/facts/tritium/index.html.
- [32] UK MET Office. Wales: Climate, wind. <http://www.metoffice.gov.uk/climate/uk/wl/>.
- [33] The Washington Post. Historical weather data. http://www.washingtonpost.com/wp-srv/weather/longterm/historical/data/manchester_united_kingdom.htm.
- [34] W. Raskob. UFOTRI: Program for assessing the off-site consequences from accidental tritium releases. (KfK 4605), April 1990.
- [35] W. Raskob. Description of the new version 4.0 of the tritium model UFOTRI including user guide. (KfK 5194), 1991.
- [36] Ulrich Schotter, Frank Oldfield, and Klaus Frohlich. Global network for isotopes in precipitation, 1996.
- [37] Transport Information Service. Climate/humidity table. http://www.tis-gdv.de/tis_e/misc/klima.htm.
- [38] J.G. Smith and J.R. Simmonds. The methodology for assessing the radiological consequences of routine releases of radionuclides to the environment used in PC-CREAM 08. (HPA-RPD-058), November 2009.
- [39] North Carolina Health Physics Society. Nuclide safety data sheet. hydrogen-3 [tritium]. <http://hpschapters.org/northcarolina/NSDS/3HPDF.pdf>.
- [40] American Society of Civil Engineers Urban Water Resources Research Council. *Design and Construction of Urban Stormwater Management Systems, Issue 77*. ASCE Manuals and Reports of Engineering Practice No.77, 1992.
- [41] U.S. Department of Energy. *DRAFT Health Physics Manual of Good Practices for Tritium Facilities*, December 1991. MLM-3719.
- [42] W.J. Walley and D.E.D.A. Hussein. Development and testing of a general purpose soil-moisture-plant model. *Hydrological Sciences Journal*, 27(1):1–17, 1982.

A Discrete Atmospheric Model Plant Transfer Term

For a constant atmospheric concentration the equation for the concentration in the plant is given by Equation 5.15. This is the step response of the plant to an air concentration equal to C_{air} . The pulse response (assuming the concentration remains at C_{air} for one

time step) is given by the following expressions:

$$C_{plantHTO} = \frac{C_{air}\alpha}{H_{sat}} (1 - e^{-kt}) \quad \text{for } 0 < t < \Delta t \quad (\text{A.1})$$

$$\begin{aligned} C_{plantHTO} &= \frac{C_{air}\alpha}{H_{sat}} (1 - e^{-kt}) - \frac{C_{air}\alpha}{H_{sat}} (1 - e^{-k(t-\Delta t)}) \\ &= \frac{C_{air}\alpha}{H_{sat}} (e^{-k(t-\Delta t)} - e^{-kt}) \\ &= \frac{C_{air}\alpha}{H_{sat}} e^{-kt} (e^{k\Delta t} - 1) \quad \text{for } t > \Delta t \end{aligned} \quad (\text{A.2})$$

Therefore at the end of time step n the concentration can be written as:

$$C_{plantHTO} = \frac{\alpha}{H_{sat}} (e^{k\Delta t} - 1) [C_{air1}e^{-kt} + C_{air2}e^{-k(t-\Delta t)} + C_{air3}e^{-k(t-2\Delta t)} + \dots + C_{airn}e^{-k(t-n\Delta t)}] \quad (\text{A.3})$$

Where $(t - n\Delta t) = \Delta t$. If n is the time-step number, then the following sequence of equations can be written as part of a loop:

$$\text{pre-factor} = \frac{\alpha}{H_{sat}} (e^{k\Delta t} - 1) \quad (\text{A.4})$$

$$\text{concentration.term}(n) = [\text{concentration.term}(n-1) + C_{air}(n)] e^{-k\Delta t} \quad (\text{A.5})$$

$$C_{plant}(n) = (\text{pre-factor}) (\text{concentration.term}(n)) \quad (\text{A.6})$$

B Uniform transfer rate from plant water to plant organic matter.

As is clear from Figure 20, the transfer of hydrogen exchange to and from the organic content of plants occurs with the plant's water only. Therefore, under equilibrium conditions, the transfer of hydrogen to and from the compartment must be equal:

$$\begin{aligned} t_{lossOBT} M_{gOBT} &= t_{gHTOgOBT} M_{gHTO} \\ t_{gHTOgOBT} &= \frac{t_{lossOBT} M_{gOBT}}{M_{gHTO}} \end{aligned} \quad (\text{B.1})$$

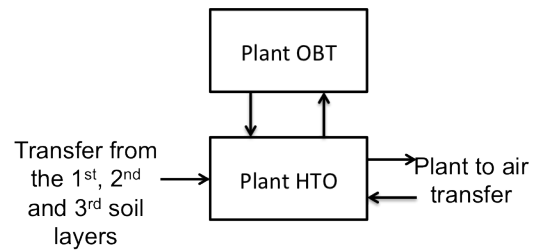


Figure 20: Plant compartment of the discrete release model [34]

The half life of tritium in plant organic matter is 10 days [34] and so $t_{gOBT} = \frac{\ln 2}{10 \times 24 \times 60 \times 60}$. The mass of hydrogen in the organic compartment of the plant is $\frac{W_p DW_p}{13}$ and the mass of hydrogen in the plant water is $\frac{W_p(1-DW_p)}{9}$.

C Activity of pure, undiluted HTO

The activity of pure tritium is 9800Ci/g [24]. From this, the activity of pure, undiluted tritiated water can be calculated as follows:

$$\frac{(9800Ci/g \times 3gT/molHTO) + (0Ci/g \times 17g/molHTO)}{20g/molHTO} = 1470Ci/gHTO \quad (C.1)$$

This roughly agrees with the value of 1400Ci/g found from [17].

This gives $5.18 \times 10^{13} Bq/gHTO$.

Assuming 40g of the 100g release is pure HTO the initial activity, the activity of the release is calculated to be:

$$Activity = \frac{40 (5.18 \times 10^{13}) + (100 - 40) (0)}{100} \quad (C.2)$$

$$= \frac{40 (5.18 \times 10^{13})}{100} \quad (C.3)$$

$$= 2.072 \times 10^{13} Bq \quad (C.4)$$

D Input Parameters for Wylfa's Routine Atmospheric Release.

Parameter	Value and Units	Explanation
Ratio of HTO concentration in soil water to that in air moisture	0.3	This is the recommended value in Hart [18] who obtained the information from the IAEA
Air humidity	$0.008L/m^3$	This has been obtained as explained in subsection 7.1. The code asks for three different values of air humidity, one averaged over the total year, another over the snow-free period and finally one averaged over the growing period of vegetation. A single value is used for all here.
Ratio for HTO concentration in plant water to that in air moisture	0.68	This is the arithmetic mean of current observed data [18]
Dry weight fraction of plants	0.3	Common assumption for living organisms [34]
Isotopic discrimination factor for plant metabolism	0.8	This is the arithmetic mean of current available data [18]
Water equivalent of plant dry matter	0.56	Hart model's default value [18]
Fraction of animal water intake derived from direct inhalation of air and absorption through skin	0.019	The value for beef in Table 1.14 Hart [18]
Dry weight fraction of animals	0.3	Common assumption for living organisms [34]
Isotopic discrimination factor for animal metabolism	0.8	The value for beef in Table 1.14 Hart [18]

The ratio of the precipitation concentration of HTO (Bq/L) and the air concentration of HTO (Bq/m^3) measured having been collected in open buckets onsite	$100m^3/L$	This is the value recommended for COG sites [18] as there is no data available for Wylfa, this value will be used as a nearest approximation
Fraction of animal water derived from ingestion of water	0.495	The value for beef in Table 1.14 Hart [18]
Fraction of animal water intake derived from metabolic water from oxidative metabolism of plant dry matter	0.074	The value for beef in Table 1.14 Hart [18]
Andelman's volatilisation factor	$0.1L/m^3$	The value recommended for tritium, although better data is required [13]
Breathing rate of humans	$19710m^3/year$	Calculated assuming a volume intake of $2.5dm^3$ per breath and 15 breaths per minute.
Fraction of year spent outdoors	0.2	Assuming an individual spends a third of their waking hours outdoors (this may be higher for Anglesey because of the number of farmers)
Total plant produce intake rate	300kg	Typical diet
Total animal produce intake rate	70kg	Typical diet
Total water intake rate	$730L/year$	Assuming 2L of water consumed daily
Dose factor for the inhalation of HTO	$3 \times 10^{-11} \frac{Sv}{Bq}$	Value recommended in Hart [18]
Dose factor for the ingestion of HTO	$2 \times 10^{-11} \frac{Sv}{Bq}$	Value recommended in Hart [18]
Dose factor for the ingestion of HTO	$4.6 \times 10^{-11} \frac{Sv}{Bq}$	Value recommended in Hart [18]
Fraction of the year spent swimming in pools	0.018	This is assuming 3 hours of swimming per week
Intake rate of water whilst swimming	$20L/year$	Estimate
Skin surface area	2.19	The value for the average adult [18]
Diffusion rate of water-wetted skin	$105L/m^2/year$	The recommended value in Hart [18]
Fraction of the year spent showering	0.0104	This is assuming a daily 15min shower
Intake rate of water whilst showering	$20L/year$	Estimate

E Input Parameters for Chalk River Laboratory Accidental Release

Parameter	Value	Source
Effective release height (h_{eff})	0	Given that the leak was in the form of water and steam, I will assume that the effective release height is zero. This is a conservative assumption for near field concentrations
Wind Data		It is not known what time the release first occurred therefore I am assuming the release takes place at noon. The hourly weather data is provided by the National Climate Data and Information Archive of Canada [7].
Stability Data		The stability classes are chosen based on the wind speed and time of day via the guidance given in the Cambridge University Engineering Environmental Fluid Mechanics course [27]
Dry deposition velocity of HTO (V_{dHTO})	$1.8 \times 10^{-2}m/s$	This is the value used in the UFOTRI model [34]

The quotient of isotopic ratios T/H in liquid and vapour due to isotopic effects (α)	1.1	Belot et. al. [2]
Precipitation Data		Daily values of rainfall in mm were obtained from the National Climate Data and Information Archive of Canada [7] and divided by 24 to obtain an approximate value for the hourly rainfall in mm/hr.
Dry weight fraction of grass (DWg)	0.2	This value is taken from suggested values for grass in the UFOTRI documentation [35]
Dry weight fraction of vegetation (DWp)	0.1	This value is taken from suggested values for leafy vegetables in the UFOTRI documentation [35]
The type of soil	Sandy loam	Raskob [34]
Relevant soil constants required for calculations in the model:	a=2.10 b=13.50 c=9.40 n=6.2 $\alpha=3.5 \times 10^4$ m=1.60 $\beta=3.5 \times 10^3$ $\theta_{sat}=0.336$ $\theta_{wilting}=0.075$	Wiley and Hussein[42]
Soil moisture volume fraction of the three soil layers θ_1 , θ_2 and θ_3	0.25	Assumptions made in the UFOTRI model [34]
Leaf area index for plants and grass (LAI_p and LAI_g respectively)	$3(m^2/m^2)$	[34]
Temperature		The hourly data is provided by the National Climate Data and Information Archive of Canada [7].
Relative Humidity		The hourly data is provided by the National Climate Data and Information Archive of Canada [7].
Weight of plants in each cell	$85kg/m^2$	Based on values given per unit area in Raskob [35]
Weight of grass in each cell	$160kg/m^2$	Based on values given per unit area in Raskob [35]
Plant canopy resistance	30s/m during the day	Values suggested by the Evapotranspiration program[9]
	200s/m during the night	
Grass canopy resistance 50 during the day and 200 during the night	Values suggested by the Evapotranspiration program[9]	
Soil resistance	150s/m	Value used in UFOTRI [35]
Weighting factor into OBT plant	1	Assumed constant throughout the year in the absence of better data
Weighting factor into OBT grass	1	Assumed constant throughout the year in the absence of better data
Bucket Ratio	100	
Radiation Balance		
Psychrometer constant	$66J/m^3K$	
The latent heat of evaporation of water	$2.27 \times 10^6 J/kg$	
Andelman's volatilisation factor	$0.1L/m^3$	The value recommended for tritium, although better data is required [13]
Breathing rate of humans	$19710m^3/year$	Calculated assuming a volume intake of $2.5dm^3$ per breath and 15 breaths per minute.
Fraction of year spent outdoors	0.2	Assuming an individual spends a third of their waking hours outdoors (this may be higher for Anglesey because of the number of farmers)

Total plant produce intake rate	300kg	Typical diet
Total animal produce intake rate	70kg	Typical diet
Total water intake rate	730L/year	Assuming 2L of water consumed daily
Dose factor for the inhalation of HTO	$3 \times 10^{-11} Sv/Bq$	Value recommended in Hart [18]
Dose factor for the ingestion of HTO	$2 \times 10^{-11} \frac{Sv}{Bq}$	Value recommended in Hart [18]
Dose factor for the ingestion of HTO	$4.6 \times 10^{-11} \frac{Sv}{Bq}$	Value recommended in Hart [18]
Fraction of the year spent swimming in pools	0.018	This is assuming 3 hours of swimming per week
Intake rate of water whilst swimming	20L/year	Estimate
Skin surface area	2.19	The value for the average adult [18]
Diffusion rate of water-wetted skin	$105L/m^2 year$	The recommended value in Hart [18]
Fraction of the year spent showering	0.0104	This is assuming a daily 15min shower
Intake rate of water whilst showering	20L/year	Estimate

F Risk Assessment Retrospective

The risk assessment submitted to the Safety Office at the start of the Michaelmas Term identified the risks of the project as limited to those associated with the use of computing equipment. This is an exact reflection of the hazards actually encountered during the course of the project. If starting the project again I would assess the risks in the same way.