Interaction matrices and associated processes for terrestrial pathways of tritium transfer

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1. INTRODUCTION

This document presents a preliminary analysis of the important feature, events and processes (FEPs) that would be relevant to model the behaviour of terrestrial agricultural systems in response to accidental releases and time-varying environmental conditions. Accidental release involves an emission lasting from a few minutes to less than two days. It is characterized by less than 1 g of tritium released at ground level (or less than 10 g of tritium released at stack level). The focus of the analysis is an aqueous and gaseous release of tritium onto agricultural systems under various climates and agricultural practices.

FEPs are terms used to define the relevant scenarios, whereby:

- Features include the components of the site, such as soil and water bodies ;
- Events include those incidents that may occur on the system, such as climatic changes, agriculture practices…; and
- Processes include those things that are ongoing, for example irrigation of agricultural land, percolation, etc.

For completeness, all participants should be involved in this analysis in order to get a recognised generic list that takes into consideration all the potentially relevant FEPs of the system. This list needs to be audited, so that modellers might be able to more transparently explain their conceptual models for tritium. Indeed, they can compare and review which FEPs are considered in their model, and why, if applicable, certain FEPs have been disregarded.

Interaction matrices (IMs) have been developed from this analysis, forming the basis for conceptual models for the assessment of terrestrial pathways of tritium transfer.

2. METHODOLOGY

The methodology used for our analysis is based on the one defined in the BIOPROTA interim report for C14 modelling (Limer et al., 2010). The following steps are used to carry out the FEP audit and the subsequent conceptual model development:

1. Refine the FEP list from the generic list, by screening those that are relevant to the specific question that the model is supposed to address;
2. Choose a set of key conceptual model objects (CMOs), which make up the leading diagonal elements of the IM
3. Go through all the off-diagonal elements (ODEs) to identify processes that affect transfer of tritium among those CMOs. This can be done in two steps:
    a. Consider each leading diagonal element in turn and how tritium might be transferred to other leading diagonal elements.
    b. Check that all the FEPs in the refined list are somewhere in the IM, or explain why the FEP is not included in the IM.
4. This process may identify redundant leading diagonal elements or the need to create new leading diagonal elements, such that step 3 may need to be repeated.

By doing so, we get a conceptual model; a non-quantitative description of components in the environment and the processes of tritium transfer, or affecting tritium transfer between them.

The mathematical model development and the search for data to support parameter value choice then follows on from this FEPs analysis and conceptual model development. Where data gaps are highlighted, this may signal the need to go back and simplify the processes being modelled, or the need to instigate a research program.

3. INTERACTION MATRICES OF TERRESTRIAL PATHWAYS OF TRITIUM TRANSFER

3.1. ASSESSMENT CONTEXT

The site context concerns any agricultural ecosystems. Differences between temperate or tropical ecosystems should be noted. The source term could be in groundwater or in atmosphere (gaseous form). Current context assumes acute releases of tritium, which may occur in the form of HTO or HT.

3.2. BIOSPHERE SYSTEM FEATURES

Agricultural environments include living components (e.g. animals, plant materials…) and non-living components (soil water, soil and canopy atmosphere,…) of ecosystems. Climate impact is assessed for temperate and other climatic zones.

3.3. CONCEPTUAL MODEL OBJECTS

The conceptual model objects (CMOs) that were identified and located in the IMs are described in Table 1.

It is considered that soil could be broken down into two layers: an upper layer (UL) which is subject to ploughing, and a lower layer (LL) which is not normally disturbed by human activity. Relevant soil features are similar for both layers.
Soil macrobiota, soil organic matter and mycorrhizae are not considered for inclusion as a CMO in the context of accidental release. We assume they all play a negligible role in our current acute release scenario. Soil microbes are not considered explicitly either, although they are responsible for the process of HT oxidation to HTO in the soil, in case of an HT atmospheric release. Rather than including soil microbes as an explicit CMO, their presence is implicit in the inclusion of the process “Oxidation(HTtoHTO)” in the soil layer interaction matrix (see Figure 1).

Tritium is readily incorporated in the form of tissue water (HTO) in biological organisms. This fraction is particularly important in plants, whose water content is 80-95 % of fresh weight, depending on species and stage of development considered. Plant is represented by a water compartment (including HTO) and a dry matter (including organic ^{3}H) compartment in both belowground and aboveground plant material (Figure 1). A distinction should be made between foliar system and edible plant organs, intended for human or animal consumption (leaves, grains, fruit, root or tuber).

Metabolic processes in animals are also described by considering the two compartments - water and dry matter - of the animal (Figure 3 ¤ 4).

Table 1 : Conceptual model objects

Conceptual model object	Description
Source	Gas: tritiated water (HTO) and tritiated hydrogène (HT). Specific flux rates would need to be defined for a specific scenario. Water: Groundwater contaminated with HTO, used for irrigation and upwelling into soil of interest. Scenario specific flux rates would also need to be defined.
Soil water	Liquid water (HTO) in the soil pores. Agricultural soil (depth, texture, pH...)
Soil gas	Tritiated water vapour in the soil pores
Plant canopy atmosphere	Within the canopy (with or without lateral air flow)
Belowground plant material	Liquid water (HTO) and dry matter (OBT) in Roots.
Aboveground plant material	Liquid water (HTO) and dry matter (OBT) in leaves, stems fruits and grains.
Animal water	Liquid water (HTO) in the animal
Animal dry matter	Dry matter (OBT) of the animal
Sink	Anything outside the system of interest

3.4. TRITIUM INTERACTION MATRICES

The interaction matrix that has been developed for a water source or a gas source scenario is given in Figure 1. The two soil layers are considered further, in particular how they interact with each other, in Figure 2; the yellow boxes indicate the lower soil layer (LL) and the grey boxes indicate the upper soil layer (UL). The interaction matrix for animals is given in Figure 3. A draft of the general tritium interaction matrix for the terrestrial environment is given in Figure 4.

	A	B	C	D	E	F	G	H
1	SOURCE (Gas)		Wet deposition Sprinkling irrigation Interception by soil	Advection/diffusion			Wet input Interception by plant	
2		SOURCE (Water)	Irrigation (Infiltration) Upwelling Capillary rise				Interception of irrigation water	
3		Percolation	SOIL WATER	Diffusive exchange	Evaporation	Root uptake		Percolation to groundwater Surface run-off
4			Gaz sorption Diffusive exchange	SOIL ATMOSPHERE	Diffusion	Root uptake	Aerenchyma	
5					CANOPY ATMOSPHERE		FoliarUptake (HTO) Gross photosynthesis (OBT) Oxidation (HTtoHTO) (if HT gaseous release)	Free air
6				Root respiration		BELOWGROUND PLANT MATERIAL	Translocation (assuming root uptake)	Cropping loss
7					Transpiration Foliar respiration	Translocation	ABOVEGROUND PLANT MATERIAL	Cropping loss Weathering
8				Oxidation (HTtoHTO) (if HT gaseous release)				SINK

Figure 1 : Gas or water source interaction matrix for H3

	A	B	C	D	E	F
1	SOURCE (gaz)				Wet deposition Sprinkling irrigation Interception by soil	Advection/diffusion
2		SOURCE (Water)	Upwelling		Infiltration (Irrigation)	
3		Percolation	SOIL WATER	Diffusive exchange	Capillary rise (HTO)	
4			Gaz sorption	SOIL ATMOSPHERE		Diffusion/advection
5	Evaporation		Percolation		SOIL WATER	Diffusive exchange
6				Diffusion	Gaz sorption	SOIL ATMOSPHERE

Figure 2 : Soil layer interaction matrix (water source). The yellow boxes indicate the lower soil layer (LL) and the grey boxes indicated the upper soil layer (UL).

	A	B	C	D	E	F	G
1	ATMOSPHERE				Inhalation		
2		SOIL			Ingestion	Ingestion	
3			PLANT MATERIAL		Ingestion	Ingestion	
4		Excretion Death and decomposition	Excretion	ANIMAL	Translocation H metabolism		
5	Exhalation	Inhalation (burrowing animals)			WATER		Excretion
6						DRY MATTER	Excretion Death and decomposition
7							SINK

Figure 3 : Tritium interaction matrix for animals. Animals are described by considering the two compartments - water (HTO) and dry matter (OBT).

ATMOSPHERE	**Deposition**		**Deposition and interception** **Gross photosynthesis**	**Gross photosynthesis**		**Inhalation**		**Dry deposition** **Precipitation** **Interception**		
Evaporation **Droplet production**	WATER BODIES		**Root uptake** **Irrigation**			**Ingestion**		**Irrigation** **Recharge by surface waters**	Release from solution	Recharge by surface waters
		VEGETATION (ABOVE - BELOWGROUND)				**Ingestion**	**Ingestion**			
Respiration **Transpiration**	Senescence and death		WATER						Root respiration	Biological weathering
Respiration Leaf fall Release of other organic compounds				DRY MATTER					Root respiration	Litterfall Senescence and death Biological weathering
	Excretion Death	Excretion			ANIMALS	Translocation H metabolism	Translocation			
Exhalation						WATER	OBT formation	Excretion	Inhalation (burrowing animals)	Excretion
							DRY MATTER			Excretion Death and decomposition
Evaporation	Groundwater recharge		**Root uptake**		Ingestion			SOIL WATER	**Diffusive exchange**	Surface run-off
Diffusion			**Root uptake** **Transport in aerenchyma**					**Diffusive exchange**	SOIL ATMOSPHERE	Diffusive exchange
Resuspension Diffusion	Desorption	External contamination Irrigation			Ingestion Bioturbation			**Diffusion** Advection Colloid transport	**Diffusive exchange**	Interface with geosphere

Figure 4 : The general tritium interaction matrix for the terrestrial environment, with the processes of potential importance for H3 highlighted in bold.

3.5. DESCRIPTIONS OF EVENTS AND PROCESSES

In this section, descriptions of the events (underlined text) and processes (bold text) within the interaction matrices are given, as well as how they relate to tritium behaviour in the biosphere.

Aerenchyma (transport in…)
Aerenchyma are inter-connected gas-filled pathways found in some plants, e.g. rice, which grows on water logged soils. They are a potential route of transport for water vapour from soil atmosphere to plant tissues.

Bioturbation
One of the agents of organic weathering*, bioturbation is the disturbance of the soil or sediment by living organisms. It may include displacing soil by plant roots, digging by burrowing animals (such as ants or rodents), pushing sediment aside (such as in animal tracks), or eating and excreting sediment, as earthworms do. Bioturbation aids the penetration of air and water and loosens sediment to promote winnowing or washing (transportation).

Capillary rise
Capillary rise is the upward movement of water through soil layers above the water table. This process arises as a result of capillary forces relating to evaporation and transpiration. Capillary rise is important in the overall water and nutrient dynamics in soil-crop systems and is a potential transport route of HTO in groundwater to the soil rooting zone.

Cropping loss (plants & animals)
Potentially, cropping provides an important removal process, at least for the higher values of root uptake. Some models may conservatively ignore this process based on the assumption that radionuclides taken up into crops would eventually be returned to the same soil through a variety of processes (including plant senescence and degradation or animal excretion*).

Death and Decomposition
The death of animals or plants (e.g. plant roots) leads to the release of radionuclides to the immediate environment during decomposition. During plant senescence and decomposition, changes in the location and chemical form of tritium may be identified (e.g. transfer from above ground to below ground storage organs during senescence or incorporation within detritivorous animals or decomposing micro-organisms).

Deposition (wet and dry)
HTO atmospheric deposition on vegetation or deposition on soil are determined similarly in the models, using a deposition velocity, an exchange coefficient or a net deposition modelled by constant transfer coefficients.
Concerning dry deposition:
HT velocity to the soil surface is about 4.10^{-5}- 4.10^{-3} $m.s^{-1}$
HTO dry deposition velocity to the soil surface is about 10^{-3}- 10^{-2} $m.s^{-1}$
It depends on soil composition, soil humidity, landcover...
Exchange velocity follows Fick law;

Concerning wet deposition during rain:
HT solubility is very weak. Deposition is negligible
HTO solubility is important. HTO is exchanged with H2O in rain drops. To estimate HTO wet deposition, we have to calculate the specific activity of rain water. Use of a washout rate or a washout coefficient

Diffusion or Diffusive Exchange
Diffusion is a physical process whereby material moves under the influence of a concentration gradient, resulting in a net flux from high to low concentration regions.
Water-air diffusive exchange can be a significant environmental transport pathway, notably in the soil-solution to soil-atmosphere phase.
See also foliar uptake*.

Discharge from below (upwelling)
In assessing the discharge of HTO in groundwater from below, consideration would be required as to chemical processes associated with changes in redox conditions as groundwater migrates from sub-soil to surface soil.

Environmental change
In the long term, climatic and associated environmental changes may alter plant uptake dynamics, through both physical changes (e.g. water regimes) and biological changes (e.g. vegetation species present). This may be important for long-term modelling of soil-plant tritium dynamics.

Erosion
Erosion is a process of transporting solids (sediment, soil, rock and other particles) in the natural environment and deposits them elsewhere. Not a tritium specific issue. It should be included in models if the site context suggests wind or surface run-off*. Erosion is distinguished from weathering*, which is the process of chemical or physical breakdown of the minerals in the rocks, although the two processes may occur concurrently.
In general, given similar vegetation and ecosystems, areas with high-intensity precipitation, more frequent rainfall, more wind, or more storms are expected to have more erosion. Sediment with high sand or silt contents and areas with steep slopes erode more easily, as do areas with highly fractured or weathered rock. Porosity and permeability of the sediment or rock affect the speed with which the water can percolate into the ground. If the water moves underground, less runoff is generated, reducing the amount of surface erosion. Sediments containing more clay tend to erode less than those with sand or silt.

Evaporation
Transfer of water from the ground directly to the atmosphere, which includes the transfer of tritium in the form of HTO. In some models, evaporation is treated in the equation describing deposition. When it is treated in a separate module, models include either constant transfer coefficients, or a rate of reemission (i.e. the ratio between the amount of HTO re-emitted over time by the total quantity of HTO deposited, generally expressed in % per unit of time). Other models that rely on complex calculations of energy balances to estimate the flux of tritiated water vapour from soil require the determination of a large number of micro-meteorological parameters, including field measurements difficult to achieve.

Excretion
Process by which waste products of metabolism and other non-useful materials are eliminated from an organism. Models simulating tritium behavior in animals may differ in the value of the elimination rate of tritium from the animal.

Exhalation (or **expiration**)
Loss of tritium in the animal gaseous phase to the external environment during breathing. Exhalation has a complementary relationship to inhalation*; the cycling between these two efforts defines respiration.

Foliar uptake
Foliar uptake is an important pathway to consider for atmospheric tritiated water uptake in the soil-plant system through leaves absorption. Tritium as tritiated hydrogen (HT) is not absorbed by vegetation because of its very low solubility in water.
The absorption of atmospheric tritiated water by leaves is based on the diffusion of vapor through the stomata. This diffusive process is controlled by climatic conditions (temperature, humidity, light) and physiological characteristics of the plant (size and density of stomates, hormonal factors). Stomatal resistance plays an important role as the plant is subjected to conditions of darkness or drought, leading to closure of the ostiole. Because of the different degrees of stomatal closure, an acute presence of ^{3}H in the plume when the latter passes above the plant during the day is likely to have a different radioecological impact on plants than if the plume passed during the night with the same ^{3}H concentration. Consequently, the determination of stomatal resistance is essential to estimate the incorporation of atmospheric HTO by leaves in response to an intermittent presence of the contaminated plume.

Gas sorption
Tritium in soil gas may dissolve in soil water.

Gross photosynthesis and growth
A fraction of tritium is incorporated into plant organic matter (OBT) following transport across stomata through the process of photosynthesis. Absorption of HTO in irrigation water across stomata and subsequent incorporation into plant material by photosynthesis (OBT) cannot be precluded.
Net photosynthesis results implicitly in the balance between synthesis of biomass (photosynthesis or gross primary production) and degradation of biomass (foliar respiration).
In models, the process of formation and elimination of organic tritium in plants are described by using transfer coefficients (constant or variable with environmental conditions) or by the use of plant growth curves. However, the values of parameters necessary for accurate prediction of transfers are rare and mostly sparse, particularly for acute releases. The highly variable behaviour of tritium as a function of exposure duration, environmental parameters (humidity, plant water status, day / night duration of the release ...) means that each test result only gives the values of parameters specific to the conditions under which the experiment took place, difficult to generalize.

H metabolisation
Process metabolizing organic hydrogen of food within living organisms.

Infiltration
Infiltration is the process by which HTO presents in contaminated rain or irrigation water enters soil. Infiltration rate of water in soil depends on several characteristics such as water supply (by rain or irrigation*), type of vegetation covering the soil and soil depth. Its minimum value is given by the hydraulic conductivity of saturated soil.
The balance of infiltration, wash-off and evaporation* is determined by a number of factors including soil type, topography, climate and rate of input. For most controlled systems, wash-off is likely to be negligible.

Ingestion
Incorporation of tritium into animals, micro-organisms or soil macrofauna.
For tritium in animals, ingestion pathways may include ingestion of water and organic matter derived from plants in the field (pasture grass) and consumer products intended for animal feed (corn silage, hay, etc..). Another source of contamination may also be drinking water.

Inhalation
Incorporation of tritium water vapour as a result of breathing. See exhalation*.

In the case of atmospheric releases of short duration, most models consider the ingestion of food (mainly grass) and absorption by inhalation as the main routes of tritium incorporation in animals.

Interception
The interception by vegetation can be defined as the fraction of the wet deposit retained by plants covering the ground. Indeed, radionuclides dissolved in rain or in groundwater applied to plants via spray irrigation* may be intercepted by plant aerial parts, thus preventing direct transport of water (and HTO) to the soil. Interception is a process that is largely considered in current models. It is particularly important for direct contamination of crops such as green vegetables whose edible parts can directly intercept contamination. The interception efficiency depends more on the nature of the deposit, the species, the development stage and the biomass density of vegetation than on the radionuclide considered (Vandecasteele et al., 2001).
HTO in intercepted water may bind to plant material leading to surface contamination or be taken up through stomata and be incorporated into plant material. Alternatively, intercepted water and HTO may subsequently be deposited in soil as a result of plant run-off*.

<u>Irrigation</u>
Use of abstracted water (containing HTO) to supplement natural supplies to agricultural crops. Irrigation may involve spraying of water directly onto plants or by application to soils (surface soil or flood irrigation).

Micro-organism metabolism and assimilation
Micro-organisms play an important role in the environmental fate of many elements, with a multiplicity of physico-chemical and biological mechanisms affecting changes in mobility and speciation. Physico-chemical mechanisms of removal include adsorption and ion exchange.
Microbial activity may be particularly important for the gas scenario, whereby H3 enters the soil in the form of HTO, HT or CH_3T. The tritiated hydrogen (HT) undergoes microbial oxidation when it enters the ground, and then behaves as tritiated water (HTO) being rapidly re-emitted in the atmosphere or migrate into the deeper soil layers. We do not know about the fate of the CH_3T form and still less of any other organic forms. Some degree of assimilation of tritium into microbes may occur as a result of the CH_3T metabolic process.

Microbial activity is dependent on a number of factors including nutrient availability. Thus nutrient deficient soils may have a slower rate of microbially-induced speciation than nutrient-rich soils.

OBT formation
This process covers the formation of OBT from tritiated water present in the animal. This formation rate may have very different values in models. More generally, models may differ in the main description of mechanisms for organic tritium: OBT intake and metabolic processes within the animal.

Oxidation (HTtoHTO)
In most cases, contamination of plants by tritium gas (HT) involves prior oxidation of the chemical form in tritiated water (HTO) in the aerial parts of plants or soil. In normal climatic conditions in temperate regions, the rate of conversion of tritiated hydrogen to tritiated water in soil or on leaf surfaces is very high. Values of the conversion rate between 0.104 and 1.64 Bq per min and per gram fresh weight were measured for seedlings of lettuce. Due to the rapid oxidation of HT, the mechanism of transfer of tritium from the environment to plants can be roughly equated to that of tritiated water absorption.

Percolation
This is the process by which tritium in soil water moves downwards into deeper horizons. The losses by vertical migration may be described by using constant transfer coefficients or by classical equations of migration or by a combination of both approaches.

Ploughing
This farming practice is used for initial cultivation of soil in preparation for sowing seed or planting. Its primary purpose is to turn over the upper layer of the soil, bringing fresh nutrients to the surface, while burying weeds and the remains of previous crops, allowing them to break down. It also aerates the soil, and allows it to hold moisture better.

Precipitation
The rate of precipitation drives water flow from the soil surface to depth and acts to dilute HTO in groundwater. Precipitation of water may inhibit release of HTO in the gas phase.

Resuspension
A renewed suspension of insoluble particles after they have been precipitated.

Root uptake
Water absorption by roots of plants involves the transfer of tritium content in the soil. Tritiated water generally borrows the same biological pathways that water within plants. Tritiated water in the soil is absorbed and transported in the xylem by a movement of "mass flux" due to an energy gradient associated with the evaporation of water from the leaves. This route of exposure to tritium is highly dependent on the concentration and distribution of tritiated water in soil and soil characteristics (structure, texture, water content ...). The transfer is also likely to be modulated by the plant species considered, its stage of development and condition of its root system parameters. In models, root uptake of HTO is generally assumed to be equal to foliar transpiration, or taken into account by exchange rates determined from differences in humidity, or described using constant transfer coefficients. The root uptake of OBT is negligible because of the small pool of soil organic tritium. In general, in cultivated soils characterized by a low humus content (1-5 %), organically bound tritium is mostly negligible. However, in soils containing high humus content (organic matter

content above 10%), the degradation of organic molecules may be a secondary source of tritium for plants, which remains however low. According to the activity concentration of tritium in atmospheric water vapour, this contribution from soil would be variable but small. All available data indicate that soil is not a compartment where organic tritium accumulates, unlike what is observed for most other radionuclides. Photosynthesis is assumed to be the main process responsible for the incorporation of OBT in plants following a sprinkling irrigation of contaminated water.

Root respiration
Respiration in root tissues may lead to the release of tritium in the form of HTO into soil atmosphere.

Soil additives
The addition of additives (fertilization, lime addition,…) to agricultural soils may affect the behaviour of tritium.

Surface run-off
Surface runoff is the water flow that occurs when soil is infiltrated* to full capacity and excess water from rain, meltwater, or other sources flows over the land. This is a major component of the hydrologic cycle. It more commonly occurs in arid and semi-arid regions, where rainfall intensities are high and the soil infiltration capacity is reduced because of surface sealing, or in paved areas. Surface runoff causes erosion* of the soil surface.

Translocation
In plants, translocation involves the transfer of tritium in the form of HTO from one part of a plant to another. It is a process largely considered in current models.
In animals, translocation stands for the allocation process of radioactive contamination in the water or dry matter pools of the different organs, once ingested by the animal.

Transpiration
Transpiration is a process similar to evaporation* and occurs through stomata. It is the loss of water vapor from parts of plants, especially in leaves but also in stems, flowers and roots. The rate of transpiration is directly related to the degree of stomatal opening, and to the evaporative demand of the atmosphere surrounding the leaf. The amount of water lost by a plant depends on its size, along with surrounding light intensity, temperature, humidity, and wind speed (all of which influence evaporative demand). Soil water supply and soil temperature can influence stomatal opening, and thus transpiration rate.

Weathering
Weathering is the breaking down of rocks, soils and minerals through direct contact with the atmosphere. Weathering occurs *in situ*, or "with no movement", and thus should not be confused with erosion*, which involves the movement of rocks and minerals by agents such as water, ice, wind, and gravity. Weathering involves the loss of tritium from the system. It can involve loss of superficial tritium from leaf surfaces or physical loss of tritium associated with surface soils as a result of atmospheric processes.

4. CONCLUSIONS AND WAY FORWARD

The interaction matrices presented here provide a list of FEPs against which models can be audited. They help to support information used in the wider case, and provide the modeller with an opportunity to clearly demonstrate which processes have been included in each model, and how ; as well as the justification for the exclusion of any processes from the model. Interaction matrices also provide a basis for comparison between the models. Indeed, FEPs have been applied here compared with our own understanding and modelling approaches.

5. REFERENCE

Limer, L.M.C., Smith, G., 2010. C-14 Long-Term Dose Assessment: Data Review, Scenario Development, and Model Comparison, FEP ANALYSIS, BIOPROTA Interim report version 1.0., 19 July 2010.