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Development, description and validation of a Tritium Environmental Release Model (TERM)

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ABSTRACT

Tritium is a radioisotope of hydrogen that exists naturally in the environment and may also be released through anthropogenic activities. It bonds readily with hydrogen and oxygen atoms to form tritiated water, which then cycles through the hydrosphere. This paper seeks to model the migration of tritiated species throughout the environment – including atmospheric, river and coastal systems – more comprehensively and more consistently across release scenarios than is currently in the literature. A review of the features and underlying conceptual models of some existing tritium release models was conducted, and an underlying aggregated conceptual process model defined, which is presented. The new model, dubbed 'Tritium Environmental Release Model' (TERM), was then tested against multiple validation sets from literature, including experimental data and reference tests for tritium models. TERM has been shown to be capable of providing reasonable results which are broadly comparable with atmospheric HTO release models from the literature, spanning both continuous and discrete release conditions. TERM also performed well when compared with atmospheric data. TERM is believed to be a useful tool for examining discrete and continuous atmospheric releases or combinations thereof. TERM also includes further capabilities (e.g. river and coastal release scenarios) that may be applicable to certain scenarios that atmospheric models alone may not handle well.

1. Introduction

Tritium (^{3}H, T) is an isotope of hydrogen (H) formed naturally in the stratosphere that, like H, readily forms a chemical bond with oxygen (O) and another H atom to form tritiated water (HTO, as opposed to the more common H_2O). Notwithstanding isotopic effects, T behaves essentially identically to its stable analogue (hydrogen), and thus traces a similar path through biological and chemical systems in the environment. The physico-chemical similarity of HTO to water is even greater, as the supplemental H and O elements in the molecule effectively reduce the relative significance of the two extra neutrons on the radioactive isotope.

Anthropogenic sources of tritium include nuclear power, medical and industrial applications: these can raise concentrations in the global hydrosphere above levels naturally induced by cosmic ray bombardment. Weapons use and testing have also raised ambient concentrations significantly in the past. The physical half-life of ^{3}H, which decays via beta emission, is 12.32 years, and knowledge of past peaks in ambient background concentrations can be used to date recharge and withdrawals of water in, for instance, aquifers (Gat, 2010; Michel, 2005).

Like all beta-emitters, ^{3}H also poses potential risks to human health. These risks are exacerbated because of the significant role, ready dispersion and uptake by biological systems of the two chemical forms, tritiated gas (HT) and tritiated water vapour (HTO). This is further complicated by the relative difficulty of beta particle detection at practicable distances in various environmental compartments. Thus, models capable of tracking ^{3}H concentrations in compartments of interest over short- and long-term time-scales form a valuable asset – alongside monitoring – in the regulatory, assessment and engineering asset toolset. Widely used models previously described in the literature are discussed in the next section. This paper proposes to supplement the existing literature with a novel conceptual model, dubbed 'Tritium Environmental Release Model' TERM. The underlying conceptual model, major processes and assumptions are described herein, followed by a series of validation tests and findings. Like other work in the literature, the model proposed deals with understanding transfers from source to air to receptors, considering all pathways. An integrative approach for short-term and long-term exposures in terrestrial and aquatic media is less well developed, and it is the aim here to present a new dynamic model of tritium transfer in

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both terrestrial and aquatic systems. This novel model is designed to both supplement and bridge the capabilities of existing models, with particular respect to environmental media (and hydrological systems especially) and release duration.

2. Overview of approach and methods

2.1. Methodology

A review of the features and underlying conceptual approaches of existing tritium release models was conducted. From these, gaps and needs were identified which were useful in finalizing model structure and formulation of TERM. We proceed with the review of four typical tritium models of significantly different complexity, which are currently in use. They are designated here as: DRLG/Hart (Hart, 2008), PCCREAM (Smith and Simmonds, 2009), GENII (Napier et al., 2004) and UFOTRI (Raskob, 1990, 1993).

The first two models are intended to provide regulatory guidance for long-term releases at relatively low emission levels, consistent with routine or near-routine releases, for instance. The third, GENII, has also been modified for use in cases of a discrete release to the atmosphere. The first three of these models (DRLG/Hart, PCCREAM and GENII) are primarily geared towards modelling the release of other radionuclides. In multi-isotopic release (e.g. fission reactor accident) scenarios, the tritium component is typically much less significant as a source of radiological dose (and also biochemical toxicity, where this may be considered) than other radionuclides (though this simplification may not hold in e.g. the aquatic environment where tritium can be a key radionuclide and contribute significantly to the doses received by aquatic biota and humans). Accordingly, these three models avoid use of detailed tritium-specific processes and opt instead for a simplistic Specific Activity concept that can broadly estimate the significance of the tritium release as a supplementary release and risk component. In contrast to these, UFOTRI is a model focused specifically on the radiological consequences of an accidental atmospheric tritium release. The processes modelled by UFOTRI are significantly more extensive and, as some also assume equilibrium conditions, more robust for a dynamic case. Of the four models discussed here, UFOTRI is probably the best model for use in the dynamic, discrete atmospheric release case. A range of other models not considered here also exists, but UFOTRI in general compares well, for our purposes, to alternatives due to its physical basis, its documented application for generic, as well as site-specific, assessments and the range of trials in literature it has undergone (c.f. Raskob, 2007; Galeriu et al., 1995).

There is also a wide discrepancy between the chemical forms and compartments considered in each model. This lack of harmony between model forms leads to inherent challenges in addressing novel conditions and conditions including multiple release types. None of the models reviewed are well suited for both discrete and prolonged releases of tritium or are capable of considering simultaneous release to multiple environmental systems (e.g. river and atmosphere). Specific inclusion of these features in the model, as well as other important hydrological systems, would be a significant and useful contribution to the modelling and assessment of tritium releases to the environment (see also, for instance, International Atomic Energy Agency [IAEA], 2012).

The methodology presented here began with a detailed review of each of the above models, and particularly an assessment of alternative formulations of component processes. This review led in turn to the formulation of a new aggregate model which emphasizes the aforementioned characteristics. The new model, TERM, was then tested against multiple validation sets from the literature, including experimental data and reference tests for tritium models.

2.2. State of the art

Release of tritium to the environment entails cycling of its chemical forms through several compartments, including those comprising plants and animals. Implied is the significant role the organically bound tritium (OBT, a chemical form also considered in TERM) can play in dose calculations, though the focus in this paper will remain on environmental concentrations. A summary of characteristics of the reviewed models is shown in Fig. 1.

The first three models in Fig. 1 are generalized radionuclide dispersion models and, regardless of the number of compartments tracked, make simplifying assumptions to estimate, broadly, specific activities of T in the environment. In contrast, the final model, UFOTRI, is significantly more granular, as it is tailored to tritium processes and, even more specifically, to those relevant for very short-term, discrete, releases thereof. GENII can also model discrete releases to the atmosphere, as well as continuous river releases. No model is suitable for discrete releases to rivers, lakes/reservoirs or coastal regions, though these are generally significant compartments from the hydrological perspective. Therefore, no model is well suited for combined scenarios, or comparison of scenarios, that involve discrete and continuous releases to the whole hydrosphere. This article proposes to find improvements to bridge and advance these research gaps (that may also reflect regulatory and compliance gaps).

3. Model definition

TERM is, first and foremost, a conceptual modelling framework that seeks to integrate existing state-of-the-art models for the migration of tritium in the environment. The conceptual submodels are selected for overall appropriateness to the discrete/continuous release scenarios, and for other properties that emphasize robustness, scalability (alongside transparency and moderate complexity/data requirements) across the entire hydrologic system in the region of interest. Thus, for example, many submodels are readily simplified (or even altogether ignored) through the use of constants *en lieu* of equations. One advantage this provides for is direct comparison with existing datasets and models (by selecting equivalent processes to be considered), but also insights such as which advantages further model complexity, parameterization, and/or data might offer. Some discussion of these features will be presented in the model validation section later in this article, while this section details the underlying conceptual model and major processes considered. Fig. 2 shows an outline of the model functionality and organizational structure.

Fig. 2 illustrates the structure of the model as two major branches (though the model can, in fact, also consider these together, which is a distinctive feature when compared with other models reviewed here). The first major branch is the discrete release case, which represents a relatively short-term, finite release. This is consistent with scenarios such as sudden discharges or accidental release. The discrete model routes the initial release into one of three transport pathways: two atmospheric pathways (one for each chemical form of T), and river releases (where HTO-related processes should dominate). Atmospheric releases use a simple Gaussian dispersion model from a point (though more complex cases may be modelled using multiple sources, for example), whereas river releases may be considered as planar, point, or line source (e.g. diffusers for a controlled released event). The model construction makes no significant a priori assumption on the reasonableness of any given release scenario, and leaves such decisions to the modeler.

The second major branch represents the continuous model, with assumptions more suited for long term releases. This may be