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TOCATTA: a dynamic transfer model of ^{3}H from the atmosphere to soil–plant systems

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ABSTRACT

This paper describes a dynamic compartment model (TOCATTA) that simulates tritium transfer in agricultural plants of several categories including vegetables, pasture and annual crops, exposed to time-varying HTO concentrations of water vapour in the air and possibly in irrigation and rainwater. Consideration is also given to the transfer pathways of HTO in soil. Though the transfer of tritium is quite complex, from its release into the environment to its absorption and its incorporation within the organic material of living organisms, the TOCATTA model is relatively simple, with a limited number of compartments and input parameters appropriate to its use in an operational mode. In this paper, we took the opportunity to have data obtained on an ornamental plant – an indoor palm tree – within an industrial building where tritium was released accidentally over several weeks (or months). More specifically, the model's ability to provide hindsight on the chronology of the release scenario is discussed by comparing model predictions of TFWT and OBT activity concentrations in the plant leaves with measurements performed on three different leaves characterized by different developmental stages. The data-model comparison shows some limitations, mainly because of a lack of knowledge about the initial conditions of the accident and when it actually started and about the processes involved in the transfer of tritium. Efforts are needed in both experimental and modelling areas for future evaluation of tritium behaviour in agricultural soil and plants exposed to gaseous HTO releases and/or to irrigation with contaminated water.

1. Introduction

Tritium (^{3}H, T) is a radioactive form of hydrogen that is naturally present in the environment butsss also routinely released by almost all nuclear facilities. In future fusion power plants, tritium inventory will increase to an amount comparable to the global tritium inventory coming from actual nuclear fission power plants. This has raised concerns on how tritium could migrate in the environment under normal operating conditions or after potential accidental releases from facilities for which the accidental health impact on humans is mainly driven by tritium, such as heavy water nuclear power plants, fuel reprocessing plants, tritium defence facilities and the ITER fusion facility (Gariel et al., 2009; Guétat et al., 2008b; Gulden and Raskob, 2005; IAEA, 2012; Okada and Momoshima, 1993).

Once released into the environment, tritium has a complex behaviour. It is extremely volatile and has the ability to substitute for stable hydrogen atoms in the composition of water (liquid water or water vapour). Therefore, tritiated water released as HTO from nuclear facilities is easily incorporated into water within living organisms as tissue-free water tritium (TFWT) and may then label the organic matter as organically-bound tritium (OBT) through metabolic processes, especially photosynthesis in the case of plants. Equilibrium is thought to be quickly reached between TFWT and tritiated water in the environment; therefore this fraction of free tritium is said to be representative of instantaneous tritium levels (Boyer et al., 2009a; Vichot et al., 2008). Conversely, OBT results from non-equilibrium processes and remains in the organisms for a long time, giving insight into the chronology of retrospective events and the consequent dose estimation for humans near nuclear plants (Boyer et al., 2009a; Choi et al., 2002; IAEA, 2012; Vichot et al., 2008). Its "non-exchangeable" fraction is of primary interest since it represents the incorporation of TFWT within the organic fraction during growth of organisms and more generally throughout their lives. Therefore, non-exchangeable OBT is a good indicator of tritium contamination (Guenot, 1984). Several studies

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estimated that the contribution of plant OBT to the ingestion dose would be about 20–30% in case of chronic exposure and about 60%–80% in case of accidental exposure (Guétat et al., 2008a,b; Raskob and Barry, 1997).

In this context, a mathematical model to predict the radioecological impact of tritium on agricultural plants is a prerequisite to assessing ingestion doses for humans. However, the dynamic movement of tritium through the different pools (i.e. air, soil, plant tissue free water and organic matter) is not easy to model because hydrogen – and hence tritium – take part in the water cycle, which affects all pools of the biosphere. More specifically, tritium transfer to crops is subject to changing environmental conditions (e.g. local meteorology, relative humidity, soil characteristics) affecting the plant physiological processes and strongly depends on the magnitude and time history of the release. Moreover, very few experimental data exist on short and long term tritium exposures to support models available in the literature (ASN, 2010; Boyer, 2009; IAEA, 2012; Thompson et al., 2011). Consequently, an increased knowledge of mechanisms of tritium transfer to plants with regard to tritium exposure and metabolic processes involved is essential for the management of potential accident situations, to obtain a (moderately) conservative assessment of doses to members of the public and to prepare the conditions for the management of the emergency situations (IAEA, 2012).

A great number of dynamic modelling approaches to predict the transfer of tritium to plants, soil and animals have been developed over the last two decades, ranging from the very simple specific activity models to more complex process-oriented models. The latter type of models such as UFOTRI (Galeriu et al., 1995; Raskob, 1990, 1993; Täschner et al., 1997), ETMOD (Russell and Ogram, 1992), NORMTRI (Raskob, 1994), SOLVEG-II (Ota and Nagai, 2011) or the FDMH (Food Dose Module Tritium) model in RODOS (Galeriu et al., 2000) simulate the detailed movement of tritium through the different pools using hourly meteorological data. However, these detailed models are not practical for calculating the doses some weeks or months after the release since they require a long computation time and a large amount of input data (Keum et al., 2006) that often are not easily available. In order to deal with the uncertainties involved in modelling the behaviour of tritium in the environment after an accident, the EMRAS II (Environmental Modelling for Radiation Safety) international programme in 2008–2012 (see http://www-ns.iaea.org/projects/emras/) has formulated the requirements to obtain a harmonised approach concerning the development of a robust, transparent and relatively simple model with a limited number of input parameters to be used for a quick and straightforward assessment after an accidental tritium release.

The main part of this paper is dedicated to the description and critical analysis of a dynamic compartment model (TOCATTA) for evaluating the tritium behaviour in agricultural soil and plants exposed to spray irrigation with contaminated water and/or to gaseous HTO releases from nuclear facilities. This description is made in continuity to the previous paper regarding the modelling of ^{14}C transfer with TOCATTA (Le Dizès et al., 2012) and is considered as a first step towards future model developments. Processes considered in this paper are assigned to two major sub-models: (1) a plant sub-model that estimates the aboveground biomass and the dynamics of tritium activity concentration in various categories of agricultural plants, (2) a soil sub-model that simulates tritium dynamics in soil water exchange processes at the soil/canopy atmosphere interface. Tritium transfer to animals is not described. The related objectives are (1) to document the scientific basis, major assumptions, conceptual modelling and mathematical formulations of the tritium transfer in soil–plant systems, (2) to discuss the weaknesses and future developments of the model to improve its predictive capability. The model was applied to the case of a real accident involving tritium releases that occurred over several weeks (or months) in 2010, the specificity of which, compared to usually treated accidental scenarios, being that the chronicle of releases prior to the identification of the contamination was not known. Thus the objective is to discuss the comparison of the model outputs with measurements so as to find clues about the most likely scenario regarding the exposure of operators within the industrial facility that may have occurred in the six months period prior to the discovery of the accidental tritium contamination.

2. Model description

2.1. Main assumptions and characteristics

The model is based on a daily time step and is mainly driven by daily atmospheric tritiated water vapour concentration and agrometeorological data. The main chemical forms of tritium released from nuclear power plants (fission and fusion types) and fuel reprocessing plants are tritiated hydrogen (HT), tritiated methane (CH_3T) and tritiated water vapour (HTO) (Akata et al., 2011; Bartels et al., 1998; Belot et al., 1996; Koarashi et al., 2004; Ota et al., 2012), the latter being the only form of atmospheric emission taken into account in the model. The other forms play no role in photosynthesis and therefore may not be directly transferred to plants. Previous studies have confirmed that plant OBT is essentially formed through photosynthesis from airborne HTO and, to a negligible extent, from airborne tritiated hydrogen (HT) (Belot et al., 1996). For that species, the only pathway leading to plant contamination would be through the chemical transformation by microorganisms of a deposit of atmospheric HT on the soil into tritiated water, and the following uptake of the water by plant roots (Ota et al., 2007). The HTO in surface soil may also eventually be converted to organically bound tritium, a main contributor to doses resulting from tritium exposures (Atarashi-Andoh et al., 2002; Belot et al., 1996; Boyer et al., 2009b; Raskob and Barry, 1997; Thompson et al., 2011). Yet, all these processes and associated kinetics were not considered in our application. Consequently, as a first approximation, the model does not consider any releases of molecular tritium-HT or tritiated methane-CH_3T nor their behaviour in the environment. Only the tritium released into the air in the form of tritiated water vapour (HTO) and associated transfers into plant non-exchangeable OBT and soil HTO are taken into account, with an explicit consideration of an isotopic fractionation during OBT formation in plants. Moreover, given the 12-year radioactive half-life for tritium, the model also considers its radioactive decay since the simulations performed by the model might exceed the time scales of years. The model has been implemented in the SYMBIOSE modelling and simulation platform that aims to assess the fate and transport of a wide range of radionuclides in various environmental systems, and their impact on humans (Gonze et al., 2011). It is parameterized for various types of agricultural plants – annual crops, vegetable crops and pasture grass – according to categories defined in the platform (Calmon, 2009). The TOCATTA model computes daily activity concentrations of tritium in various types of agricultural soil, as well as plant and animal products. The subsequent calculation of tritium transfer to man and assessment of doses are performed within the SYMBIOSE platform, based on a typical terrestrial food-chain scenario. The model has been successfully used with SYMBIOSE to perform an area-wide dose assessment around a French nuclear power plant (Mourlon et al., 2011). It has the following main characteristics, most of which are similar to those used for modelling the transfer of ^{14}C (Le Dizès et al., 2012):