Journal of Environmental Radioactivity 178-179 (2017) 212–231

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Role of soil-to-leaf tritium transfer in controlling leaf tritium dynamics: Comparison of experimental garden and tritium-transfer model results

Masakazu Ota [a, *], Nana-Owusua A. Kwamena [b], Steve Mihok [b], Volodymyr Korolevych [c]

[a] Research Group for Environmental Science, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195 Japan
[b] Canadian Nuclear Safety Commission, 280 Slater Street, PO Box 1046, Station B, Ottawa, Ontario K1P5S9, Canada
[c] Environmental, Radiological and Chemical Sciences Division, STN51A, Canadian Nuclear Laboratories, Chalk River, Ontario, Canada

ARTICLE INFO

Article history:
Received 7 April 2017
Received in revised form
24 August 2017
Accepted 1 September 2017

Keywords:
Organically-bound tritium
Tissue-free water tritium
Root uptake
Foliar deposition
Land surface model

ABSTRACT

Environmental transfer models assume that organically-bound tritium (OBT) is formed directly from tissue-free water tritium (TFWT) in environmental compartments. Nevertheless, studies in the literature have shown that measured OBT/HTO ratios in environmental samples are variable and generally higher than expected. The importance of soil-to-leaf HTO transfer pathway in controlling the leaf tritium dynamics is not well understood. A model inter-comparison of two tritium transfer models (CTEM-CLASS-TT and SOLVEG-II) was carried out with measured environmental samples from an experimental garden plot set up next to a tritium-processing facility. The garden plot received one of three different irrigation treatments – no external irrigation, irrigation with low tritium water and irrigation with high tritium water. The contrast between the results obtained with the different irrigation treatments provided insights into the impact of soil-to-leaf HTO transfer on the leaf tritium dynamics. Concentrations of TFWT and OBT in the garden plots that were not irrigated or irrigated with low tritium water were variable, responding to the arrival of the HTO-plume from the tritium-processing facility. In contrast, for the plants irrigated with high tritium water, the TFWT concentration remained elevated during the entire experimental period due to a continuous source of high HTO in the soil. Calculated concentrations of OBT in the leaves showed an initial increase followed by quasi-equilibration with the TFWT concentration. In this quasi-equilibrium state, concentrations of OBT remained elevated and unchanged despite the arrivals of the plume. These results from the model inter-comparison demonstrate that soil-to-leaf HTO transfer significantly affects tritium dynamics in leaves and thereby OBT/HTO ratio in the leaf regardless of the atmospheric HTO concentration, only if there is elevated HTO concentrations in the soil. The results of this work indicate that assessment models should be refined to consider the importance of soil-to-leaf HTO transfer to ensure that dose estimates are accurate and conservative.

1. Introduction

Radiation dose via ingestion of organically-bound tritium (OBT) contained in food products comprises an important portion of the public dose due to tritium releases from nuclear facilities (Bartels, 1991; Gulden and Raskob, 1991; Diabaté and Strack, 1993; Boyer et al., 2009; Le Goff et al., 2014; Melintescu and Galeriu, 2017). Continuous releases of tritium from tritium-releasing facilities such as heavy water reactors and tritium processing facilities can result in elevated concentration of tritium in the surrounding environment (Davis et al., 2002; Kim et al., 2008, 2011). Therefore, concentrations of tritium, particularly OBT in plants, in impacted terrestrial areas must be understood to reliably assess doses from tritium to members of the public living near nuclear facilities (Melintescu and Galeriu, 2017).

OBT concentrations are often estimated due to the complexity of measuring OBT (Baglan et al., 2011; CNSC, 2013; Korolevych et al., 2014; Thompson et al., 2015; Melintescu and Galeriu, 2017). The ratio of the concentration of OBT (Bq kg-water^{-1}; here 'water' is the water equivalent of the organic matter analyzed) to that of tissue-free water tritium (TFWT) (hereafter referred to as the OBT/HTO

* Corresponding author.
E-mail address: ohta.masakazu@jaea.go.jp (M. Ota).

http://dx.doi.org/10.1016/j.jenvrad.2017.09.002

ratio) is used to estimate the OBT concentration in plants from measured concentrations of TFWT. OBT/HTO ratios around 0.7–0.8 are recommended for regulatory assessment purposes and environmental transfer models (Davis et al., 2002; Kim et al., 2011; Korolevych et al., 2014; Thompson et al., 2015). The value is based on fractionation of tritium during formation of OBT from TFWT by photosynthesis (Guenot and Belot, 1984; Diabaté and Strack, 1993). The use of an OBT/HTO ratio of 0.7–0.8, therefore, implies that there is equilibrium between OBT and TFWT in plants; i.e., the concentration of tritium in the plant dry matter is equal to that in the plant water, apart from the fractionation that occurs during photosynthesis (Pointurier et al., 2003; Kim et al., 2011).

However, studies in the literature have shown that OBT/HTO ratios in plants are variable, ranging from 0.1 to 10 or greater (Bogen and Welford, 1976; Belot et al., 1986; Hisamatsu et al., 1991; Davis et al., 2002; Pointurier et al., 2003, 2004; Choi et al., 2005; Jean-Baptiste et al., 2007; Kim et al., 2008, 2011, 2012a; CNSC, 2010, 2013; Baglan et al., 2011; Korolevych et al., 2014; Thompson et al., 2015; Mihok et al., 2016). Generally, observations of OBT/HTO ratios tend to be greater than the equilibrium ratio (Diabaté and Strack, 1993; Kim et al., 2011, 2013; Le Goff et al., 2014). Jean-Baptiste et al. (2007) reported that OBT/HTO ratios of grass plants and food items sampled from the southeast region in France were in general greater than unity, with a mean and standard deviation of 1.9 ± 0.9. A comprehensive survey by Pointurier et al. (2003, 2004) showed that OBT concentrations of tree-leaf samples at two background sites (not affected by any facility's release of tritium) were higher than the regional ambient HTO concentration averaged over the growth period of the leaves, with mean OBT/HTO ratios of 1.6 ± 0.5 and 3.0 ± 1.5. These elevated OBT/HTO ratios likely represent the enhanced accumulation of OBT during past growing periods and the dynamic nature of TFWT in plants and HTO in the atmosphere at the time of the sampling (Fujita et al., 2007; Kim et al., 2011, 2013; Kim and Korolevych, 2013; Korolevych et al., 2014; Thompson et al., 2015; Melintescu and Galeriu, 2017).

OBT/HTO ratios close to the equilibrium ratio have nevertheless been observed for plants growing in specific environments: for example, plants growing at sites where the soil has elevated levels of HTO (Davis et al., 2002; Kim et al., 2008, 2011; Korolevych et al., 2014; Mihok et al., 2016). The soil at Duke Swamp, located in Chalk River, Canada, has elevated levels of HTO due to groundwater discharges of tritium from the adjacent waste-management area. OBT/HTO ratios in leaves of various plants (grass, balsam fir, pine, and alder) growing at the site appear to be at semi-equilibrium during their growth period (0.7 ± 0.4) (Kim et al., 2008, 2011). Mihok et al. (2016) have also shown that OBT/HTO ratios in leaves of grass growing in HTO-impacted soil remained systematically low (1.4 ± 0.8), compared to substantially higher ratios (15.1 ± 9.1) in grass growing in soil with low tritium concentrations. It is expected that soil-related processes, particularly transfer of HTO from impacted soil to leaves, may dominate when low OBT/HTO ratios are observed (Kim et al., 2008; Melintescu and Galeriu, 2017). Clarifying the role of soil-to-leaf HTO transfer in controlling the dynamics of tritium in the plants growing in such specific environments is important to obtain a well harmonized value of the OBT/HTO ratio and thus accurate estimates of tritium doses (Kim et al., 2011; CNSC, 2013; Korolevych et al., 2014; Mihok et al., 2016). However, direct, experimental measurements of soil-to-leaf HTO transfer and the corresponding dynamics of TFWT and OBT in leaves are difficult.

Alternatively, applying model simulations to environmental data from an actual field site is an effective approach to investigating the importance of each process in controlling site-specific tritium transfer. So far, many land surface tritium models have been developed (Raskob, 1992; Yamazawa, 2001; Korolevych and Kim, 2011; Ota and Nagai, 2011a, 2011b; Ota et al., 2012; Aulagnier et al., 2012; Le Dizès et al., 2013, 2015, 2017; Korolevych et al., 2014). UFOTRI (Raskob, 1992) calculates the transfer of HTO from soil to plant compartments by using the transpiration stream based on the Penman-Monteith's evapotranspiration model. Recent models, such as CTEM-CLASS-TT (Korolevych and Kim, 2011; Korolevych et al., 2014) and TOCATTA-χ (Aulagnier et al., 2012; Le Dizès et al., 2013, 2015, 2017), include more sophisticated approaches to calculating soil-to-leaf HTO transfer by using the Farquhar's photochemistry as well as the Ball-Berry and Leuning's (Ball et al., 1987; Leuning, 1995) photosynthesis-stomata conductance interdependent model (CTEM-CLASS-TT) or Huntingford's (Huntingford et al., 2015) photosynthesis-stomata conductance model (TOCATTA-χ). The complex model, SOLVEG-II (Yamazawa, 2001; Ota and Nagai, 2011a, 2011b; Ota et al., 2012), further considers vertical distribution of water-absorbing roots and HTO within many soil layers, allowing for a more precise prediction of root uptake of HTO. It is expected that application of such sophisticated models to the tritium transfer to experimental data will clarify the impact of soil-to-leaf HTO transfer on the dynamics of tritium in leaves.

The aim of the present study is to investigate the role of soil-to-leaf HTO transfer via root-water uptake in controlling the dynamics of TFWT and OBT transfer, and therefore OBT/HTO ratios, in leaves. We hypothesize that the HTO stream from soil with elevated concentrations of HTO via continuous root-water uptake increases the concentration of TFWT in plants, and therefore quasi-equilibrium is achieved between the TFWT and OBT in the leaves, with OBT/HTO ratios approaching the equilibrium ratio. To test this hypothesis, simulations of two sophisticated tritium models, CTEM-CLASS-TT and SOLVEG-II, were compared to measured tritium concentrations of environmental samples obtained at experimental garden plots located near a tritium-processing facility where the rooting zone of the soil was maintained at different HTO concentrations through different water irrigation treatments (Mihok et al., 2016).

2. Models, experiment, and simulation settings

2.1. CTEM-CLASS-TT

CTEM-CLASS-TT is a one-dimensional model with single canopy, litter layer, and layered root and soil compartments (Korolevych and Kim, 2011; Korolevych et al., 2014). A schematic of the tritium transfer processes in the CTEM-CLASS-TT is illustrated in the literature (Fig. A1 in Korolevych et al. (2014)) and key equations of the model are presented in Appendix A. The framework of the model is the multi-layer Canadian LAnd Surface Scheme (CLASS 2.7) and phenomenological refinement of the Canadian Terrestrial Ecosystem Model (CTEM) of Environment Canada (Arora, 2003). CTEM-CLASS-TT is run by inputs of meteorological data at a reference height. Energy, water, and carbon cycles are explicitly modelled and extensively validated (Arora, 2003). The coupled dynamic photosynthesis model uses the Farquhar's photochemistry (Farquhar et al., 1980) within the Ball-Berry and Leuning's stomata-photosynthesis model (Ball et al., 1987; Leuning, 1995), which makes photosynthesis, stomatal conductance and energy balance interdependent. Plant growth (leaf area index (LAI), and mass of leaf, stem, and root) is modelled by CTEM for nine plant phenotypes.

In modelling tritium transfer (Appendix A), inputs of HTO to the soil compartment are considered by gaseous exchange of HTO, wet deposition of HTO, and dry deposition of HT (i.e., oxidation of HT to HTO). The added HTO is transported in the layered soil following forecasted advection caused by Darcy's law, and the soil HTO is taken-up by plant roots through the transpiration stream. In the