Journal of Environmental Radioactivity 129 (2014) 157–168

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

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

OBT/HTO ratio in agricultural produce subject to routine atmospheric releases of tritium

V.Y. Korolevych [a,*], S.B. Kim [a], P.A. Davis [b]

[a] Environmental Technologies Branch, Nuclear Sciences Division, Chalk River Labs, AECL, Chalk River, ON, Canada K0J 1P0
[b] Rad.Safe Inc., Canada

ARTICLE INFO

Article history:
Received 6 November 2012
Received in revised form
17 December 2013
Accepted 18 December 2013
Available online 4 February 2014

Keywords:
Tritium
HTO
OBT
Vegetation-atmosphere exchange

ABSTRACT

The mean expected value of the OBT/HTO ratio (i.e. generic ratio) is derived in this study on the joint basis of a long-term study conducted at Atomic Energy of Canada Limited (AECL)'s Chalk River Laboratories (CRL), model simulations targeted at filling gaps in a yet incomplete timeline of CRL measurements and a reference dataset comprised of numerous experiments reported in the literature. Cultivar variability and disparity in site-specific settings are covered by the reference dataset. Dynamical variability caused by meteorology has been a specific target of the long-term experimental campaign at CRL, where the former two types of variability were eliminated. The distribution of OBT/HTO ratios observed at CRL appears to be a fairly good match to the distribution of OBT/HTO ratios from the literature. This implies that dynamical variability appears important in both cases. Dynamics of atmospheric HTO at CRL is comprised of a sequence of episodes of atmospheric HTO uptake and re-emission of plant HTO. The OBT/HTO ratio appears sensitive to the proportion of the duration of these two episodes: the lesser the frequency (and duration) of plume arrivals, the higher the expected mean OBT/HTO ratio. With the plume arrival frequency defined by the typical wind rose, one would encounter a mean OBT/HTO ratio close to 2. It is important to note that this number is seen both in the reference dataset, and in the continuous timeline of HTO and OBT reconstructed from CRL observations by dynamical interpolation (modelling). Many datasets (including that of CRL) targeted at the OBT/HTO ratio are biased high compared to the suggested number. This could be explained by scarce measurements of the low OBT/HTO ratios in the short phase of uptake of atmospheric HTO by the plant.

1. Introduction

Organically bound tritium (OBT) in plants is responsible for an appreciable portion of the ingestion dose during chronic releases of tritium (Evans, 1969; Hisamatsu et al., 1989; Gulden and Raskob, 1992; Kim and Han, 1999; Kotzer and Trivedi, 2001; Peterson and Davis, 2002). The cost of frequent monitoring of OBT makes it logical to look at the plant tissue-free water tritium (HTO) as its proxy, provided the OBT/HTO ratio is known. Conversely, the ratio could be used for probabilistic quantification of plant HTO on the basis of OBT (and especially non-exchangeable OBT), which does not fluctuate as much as plant HTO does. OBT/HTO ratio also indicates the ability of plants to concentrate tritium into the organic fraction (Okada and Momoshima, 1993; Boyer et al., 2009). Quantification of the OBT/HTO ratio and interpretation of measurements, however, appears difficult (CNSC, 2011). The major difficulty is with explanation of measurements. According to prevailing considerations of the Specific Activity model presently underlying all regulatory algorithms (CSA, 1987), the OBT/HTO ratio is expected to be less than unity. However, field observations do not support this suggestion (Bogen and Welford, 1976; Hisamatsu et al., 1987, 1989, 1990, 1992; Brown, 1988, 1995; Pointurier et al., 2004; Inoue and Iwakura, 1990; Momoshima et al., 2000; Baglan et al., 2005; CNSC, 2011). Unambiguous measurements are difficult primarily due to the high cost, which prevents the analysis of large enough samples. Ambiguity inherent to small samples stems from the natural spatio-temporal variability in the ecosystem (cultivar, climatic, meteorological, hydrological, etc.) and analytical uncertainties (Diabate and Strack, 1993; Baumgärtner and Donhärl, 2004; Pointurier et al., 2004). This study targets the temporal variability caused by the meteorology and the associated scatter

* Corresponding author. Tel.: +1 613 584 8811.
E-mail addresses: korolevv@aecl.ca, vladi_kor@yahoo.com (V.Y. Korolevych).

0265-931X/$ – see front matter
http://dx.doi.org/10.1016/j.jenvrad.2013.12.014

observed in the OBT/HTO ratios (Kim and Davis, 2008; Korolevych, 2013).

Most tritium released to the atmosphere from CANDU reactors is in the form of tritiated water vapour (HTO). The tritium in the effluent exchanges readily with hydrogen in air moisture, precipitation and soil water. Some of this tritium appears in plants via diffusion from the air through the stomata and via root uptake through the transpiration stream (Belot, 1986). This tritium is distributed throughout the free water portion of the plant fresh tissue, in which form it is referred to as either tissue-free water tritium (TFWT) or HTO. The HTO designation will be used here. Some of this tritium can be incorporated into organic compounds to form organically bound tritium (OBT) (Diabate and Strack, 1993). OBT is formed primarily in the leaves and can be translocated to the edible parts of the plant.

The exchange of tritium (T) and hydrogen (H) atoms in the various free water compartments of the environment is very rapid. HTO concentrations in plants have a biological half-life of a few hours (Strack et al., 2005) and tend to track changes in air concentrations. In contrast, OBT formation is a chemical rather than an exchange process and occurs over the lifetime of the plant when the relevant metabolic processes are operative. An OBT measurement at a point in time reflects the integrated effects of the daily rates of dry matter production in the previous weeks and the HTO concentration in the plant water that took part in the production. Tritium in OBT is fixed in the plant to the same extent as carbon, with a biological half-life of about 25 days (NCRP, 1978). It does not immediately change in response to changes in the air concentration or plant HTO concentration. If the tritium source is removed, the OBT concentration in the plant decreases slowly by conversion to HTO as the dry matter breaks down through catabolic processes and by dilution with new, uncontaminated dry matter as the plant continues to grow.

The great mobility of tritium in the HTO form has led to the belief that the T/H ratio (or equivalently, the HTO concentration) is the same in all interacting water compartments of the environment, which is the basis of the specific activity (SA) model. The SA model underlies almost all environmental tritium models (Evans, 1969). It is expected that SA concepts apply to OBT as well since the OBT formed by a given plant process at a given time has a T/H ratio that reflects the ratio in the water that enters into that process. In theory, OBT concentrations are lower than HTO concentrations because the large difference in mass between hydrogen and tritium gives rise to significant isotopic effects in OBT formation. The oxygen-tritium bonds of tritiated water are split less frequently than oxygen-hydrogen bonds during photosynthesis, so less tritium than hydrogen is incorporated into organic molecules. In contrast, carbon-tritium bonds in the plant dry matter are severed more slowly than carbon-hydrogen bonds. The net result is an OBT concentration that is slightly lower than the HTO concentration. This can be interpreted as an isotopic discrimination factor (IDp) and also as the OBT/HTO ratio in the plant.

Observed values of the OBT/HTO ratio in plants show large variations that are not consistent with the value of IDp (e.g. IDp = 0.8 as per recommendations of CSA, 1987) expected from the SA model (Bogen and Welford, 1976; Hisamatsu et al., 1987, 1989, 1990, 1992; Brown, 1995; Pointurier et al., 2003, 2004; Inoue and Iwakura, 1990; Momoshima et al., 2000; Baglan et al., 2005). At first glance, observations such as these suggest that processes other than simple isotopic fractionation are at play in concentrating or diluting tritium in organic tissue (Baumgärtner and Donhärl, 2004). The existence of such processes would cast doubt on the SA model, which is the basis for all regulatory dose assessments of chronic tritium releases. In order to maintain confidence in the model, the large ratios need to be explained in terms of a process, condition or experimental procedure that violates SA assumptions of steady state, but not the principles set forth by the SA approach.

Murphy (1993) analyzed the HTO concentrations in air, plants and soil, and noticed that all these variables are in a highly dynamic state. When the atmospheric plume is present, air concentrations are high, plant HTO concentrations build up rapidly and then level off, and surface soil concentrations increase steadily. When the atmospheric plume moves off, air concentrations decrease abruptly but remain well above the undisturbed background due to re-emission of deposited tritium. Similarly, plant concentrations drop off rapidly to levels that reflect concentrations in the transpiration stream. Surface soil concentrations also decrease quickly, although concentrations at deeper levels change only slowly with time. The cycle repeats itself when the wind brings the plume back over the location in question. In general, concentrations in any one part of the system are not in equilibrium with concentrations in any other part at any time. This creates problems in interpreting tritium field measurements. For example, the HTO concentration in vegetation can be much lower than the air concentration (if the plant sample is taken just after the airborne plume reaches the site) or much higher (if the sample is taken just after the plume moves off the site). These are not examples of isotopic effects or tritium bioaccumulation, but simply analytical artefacts pertinent to transient system.

The key part of the dynamical exchange process is the succession of phases of HTO uptake and re-emission. The proportion between the average length of deposition (when the plume is present) and the average length of re-emission (when the wind is blowing the plume away from the receptor) is site-specific and is well-defined by a wind rose. The difference between instantaneous measurements of HTO and OBT in a plant is defined by a stage of the depuration process, and is quantified by a difference between the rate of HTO re-emission and the OBT depuration rate. OBT/HTO ratios measured and reported worldwide, as well as at CRL, were assembled into a database and compared with results of calculations. Comparison revealed consistency in the generic OBT/HTO ratio and showed that the uncertainty of the ratio is lower than the currently known scatter of experimental data. The comparison also showed that the OBT/HTO ratio is sensitive to uncertainties in plant-atmosphere exchange rates and the frequency of plume presence (as per wind rose).

In this study, changes in air HTO concentration are hypothesized to be the sole cause of persistently non-unit values of the OBT/HTO ratio in plants. This implies that normal vegetation sampling procedures inevitably lead to values of the plant OBT/HTO ratio significantly different from one, and in most cases greater than one. However, whether the occurrence of high values of the OBT/HTO ratio arise simply as a result of normal plant sampling procedures, or these values provide a cause to doubt the validity of the SA principles and are an indication of the bioaccumulation of tritium in plants, is not yet clear. It is therefore the objective of the present study to investigate the consequences of random sampling and the dynamics of tritium transfer processes that occur under typical meteorological conditions and subsequently result in changing HTO concentration in the air.

In Section 2, the analytical techniques used to determine HTO and OBT concentrations in plants are discussed. The SA model is described in more detail in Section 3, along with a theoretical discussion of the conditions and processes leading to extreme values for OBT/HTO ratio. Measured values of the ratio from the literature are reported in Section 4. Values significantly different from one are explained in terms of the conditions and mechanisms identified in the theoretical part of the study.