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Tritium dynamics in soils and plants grown under three irrigation regimes at a tritium processing facility in Canada

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ABSTRACT

The dynamics of tritium released from nuclear facilities as tritiated water (HTO) have been studied extensively with results incorporated into regulatory assessment models. These models typically estimate organically bound tritium (OBT) for calculating public dose as OBT itself is rarely measured. Higher than expected OBT/HTO ratios in plants and soils are an emerging issue that is not well understood. To support the improvement of models, an experimental garden was set up in 2012 at a tritium processing facility in Pembroke, Ontario to characterize the circumstances under which high OBT/HTO ratios may arise. Soils and plants were sampled weekly to coincide with detailed air and stack monitoring. The design included a plot of native grass/soil, contrasted with sod and vegetables grown in barrels with commercial topsoil under natural rain and either low or high tritium irrigation water. Air monitoring indicated that the plume was present infrequently at concentrations of up to about 100 Bq/m^3 (the garden was not in a major wind sector). Mean air concentrations during the day on workdays (HTO 10.3 Bq/m^3, HT 5.8 Bq/m^3) were higher than at other times (0.7–2.6 Bq/m^3). Mean Tissue Free Water Tritium (TFWT) in plants and soils and OBT/HTO ratios were only very weakly or not at all correlated with releases on a weekly basis. TFWT was equal in soils and plants and in above and below ground parts of vegetables. OBT/HTO ratios in above ground parts of vegetables were above one when the main source of tritium was from high tritium irrigation water (1.5–1.8). Ratios were below one in below ground parts of vegetables when irrigated with high tritium water (0.4–0.6) and above one in vegetables rain-fed or irrigated with low tritium water (1.3–2.8). In contrast, OBT/HTO ratios were very high (9.0–13.5) when the source of tritium was mainly from the atmosphere. TFWT varied considerably through time as a result of SRBT's operations; OBT/HTO ratios showed no clear temporal pattern in above or below ground plant parts. Native soil after ~20 years of operations at SRBT had high initial OBT that persisted through the growing season; little OBT formed in garden plot soil during experiments. High OBT in native soil appeared to be a signature of higher past releases at SRBT. This phenomenon was confirmed in soils obtained at another processing facility in Canada with a similar history. The insights into variation in OBT/HTO ratios found here are of regulatory interest and should be incorporated in assessment models to aid in the design of relevant environmental monitoring programs for OBT.

1. Introduction

Tritium (as a by-product of the use of heavy water as a moderator) is a major element of the permitted releases of CANDU (CANada Deuterium Uranium) nuclear power reactors in Canada. Releases are regulated by the Canadian Nuclear Safety Commission (CNSC) under the Nuclear Safety and Control Act (NSCA) and represent a dose of less than 10 μSv/year to the public. Tritium is also released to the environment through other nuclear applications (mainly as tritiated water or HTO, and tritiated hydrogen gas HT). Small quantities are continuously generated in the upper atmosphere from the interaction of gases and cosmic radiation. The signature of fallout tritium from the legacy of nuclear weapons testing is also still detectable (Eyrolle-Boyer et al., 2014). The proposed use of fusion technology for power generation is also likely to release tritium (Nie et al., 2015). Hence, its fate and transport when released to the environment are of fundamental interest to

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regulatory bodies.

Tritium has been studied extensively with the results of theoretical, field and laboratory studies incorporated in regulatory assessment models such as the CSA N288.1 model used in Canada (Canadian Standards Association, 2014). Canada has made major contributions to tritium science, including the hosting of an international effort at Chalk River in 1994 to characterize the fate and transport of HT releases to the atmosphere (Davis et al., 1995). Of particular interest has been the measurement of organically bound tritium (OBT) in agricultural produce because of its contribution to public dose through consumption of local foods. The CNSC was one of the first organizations to conduct a detailed study of OBT in Canadian foods (Brown, 1995). Since then, OBT has been studied extensively in plants (Boyer et al., 2009; Kim et al., 2013a) and more recently in soils (Kim et al., 2013b). However, many questions still remain on the dynamics of OBT in the environment.

As part of the CNSC tritium studies project (Thompson et al., 2011), a major field survey was conducted in 2008/2009 on the forms of tritium (HTO, OBT) in vegetation and soils near four nuclear facilities in Ontario and Quebec releasing HTO or both HTO and HT (Thompson et al., 2015). Similar surveys are routinely conducted for compliance purposes by CNSC licensees and as special studies, but OBT is not often measured, only estimated. This has resulted in a large body of information on HTO, but far less data for OBT. These data are needed to validate assessment models (Davis et al., 2005), especially for facilities releasing HT (Kim et al., 2014). The 2008/2009 survey detected unexpectedly high OBT/HTO ratios in plants and soils in various contexts, but particularly at one processing facility releasing HT as well as HTO (SRBT, ~40 km from the 1994 HT study at Chalk River). The nature of OBT/HTO ratios near nuclear facilities and also at background locations have recently been under intense scrutiny by many researchers. However, mechanisms resulting in disequilibrium with HTO are still not well understood (Boyer et al., 2009; Eyrolle-Boyer et al., 2014, 2015; Galeriu et al., 2013; Kim et al., 2008, 2009, 2012a, 2012b, 2013a, 2013b; Kim and Korolevych, 2013; Korolevych et al., 2014; Korolevych and Kim, 2013; Vichot et al., 2008).

The environmental processes accounting for the incorporation of HTO and OBT in soils and vegetation following releases of tritium to the atmosphere from Canadian nuclear facilities were recently reviewed (CNSC, 2009) and are only briefly highlighted here. At SRBT, a key feature of its operations is the release of HT as well as HTO (Appendix). HT (a gas) is oxidized at a very slow rate in the atmosphere to HTO (water), hence local deposition is almost entirely due to dry deposition and ensuing equilibrium with soil air. Uptake in plants occurs indirectly through oxidation of HT to HTO in soil air spaces by microbes. The resulting HTO follows the same complex pathways as water in soils and plants (root uptake, air exchange, evapotranspiration, etc.). Only a few studies have quantified the rates of oxidation of HT under natural conditions (Davis et al., 1995; Kim et al., 2014). Hence, models for processes related to HT are largely based on empirical observations (CSA, 2014). In contrast, the behavior of HTO itself has been well characterized through various international efforts (IAEA, 2003, 2014), with many sophisticated models now available for predicting the dynamics of HTO and OBT in plants both above and below ground.

The goal of the present study was to follow up on Thompson et al. (2015) with a more focused experimental study at one of the tritium processing facilities (SRB Technologies, SRBT) in Pembroke, Ontario included in previous investigations. SRBT has been the subject of several scientific studies over many years of operations and is described in Thompson et al. (2015) and CNSC (2009, 2010a, b). It has also been the focus of several comparisons of observed and modelled measurements of HTO in various environmental media [CNSC 2009, 2010a]; hence the behavior of tritium in the environment near SRBT in Pembroke is very well documented.

The goal of the present study was to validate previous findings in a rigorous statistical sense, especially in terms of obtaining timely, replicated data to characterize the circumstances under which high OBT/HTO ratios may occur for diverse release conditions. To achieve this, an experimental garden with representative vegetables and sod was set up near SRBT in spring of 2012 in parallel with focused air and release monitoring for both HT and HTO. SRBT emits both forms of tritium from two adjacent stacks. Processing releases occur during working hours (daylight hours) from Monday to Friday; only fugitive releases occur at night and on weekends. A unique feature of operations is that SRBT does not process tritium during precipitation (i.e., rain or snow); but note that fugitive releases occur continuously (HTO in particular, Appendix). As the contribution of wet deposition is much less than that of dry deposition (IAEA, 2003), this unique aspect of SRBT's operations is not critical for interpretation of the results reported here. SRBT was chosen for this study as the best example of high OBT/HTO ratios from Thompson et al. (2015), but similar high ratios were observed at three other nuclear facilities that release tritium under all weather conditions.

The garden was next to a small plot of native grass that was also monitored. HTO and OBT were measured in soils and plants on a weekly basis, with twice-daily monitoring of HTO and HT in air, and with real-time monitoring of local weather and total tritium stack releases. A few supporting experimental contrasts were also included in the design to scope the relative importance of root uptake as opposed to atmospheric uptake of tritium, and to mimic irrigation as practiced at a typical home garden. This paper reports the main methods and detailed tritium results for soils and vegetation, with further papers to be prepared on tritium in air and environmental transfer model intercomparisons for these data.

2. Materials and methods

2.1. General

2.1.1. Study area and context

Tritium was measured in matched soils and vegetation, air and precipitation samples during the 2012 growing season (i.e., May to September) at Pembroke, Ontario, at a site adjacent to SRBT. SRBT is a gaseous tritium light source manufacturing facility that began operations in 1990. It emits tritium as tritiated hydrogen gas (HT) and tritiated water (HTO) from two adjacent stacks with weekly monitoring of emissions. During the experiments carried out in 2012, tritium releases averaged 575 GBq/week (range 174–1160) with an average HT/HTO ratio of 1.8 (range 0.8–4.2). Releases from the facility are not constant; pulsed releases due to bulk processing and the filling of small light sources occur during working hours (Monday - Friday). The pulsed releases are recorded through real-time stack monitoring (total tritium only).

2.1.2. Tritium analysis

Methods of analysis for both HTO and OBT were similar to those used in a previous study (Thompson et al., 2015), and are only briefly noted here. HTO was analyzed by liquid scintillation counting (LSC) of water (Perkin Elmer Quantulus 1220). Water was obtained directly (precipitation, tap and groundwater), by entrapment in a water bubbler (air), by catalytic conversion (HT in air catalytically converted to HTO and entrapped in a water bubbler), or by extraction through vacuum drying and entrapment in liquid nitrogen (water in vegetation and soils). HTO in soil and vegetation is referred to as Tissue Free Water Tritium (TFWT) here for consistency with past studies (Kim et al., 2013a).

Once water was extracted, fully dried material was analyzed for