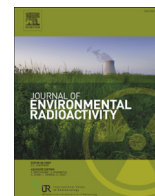


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Levels of tritium in soils and vegetation near Canadian nuclear facilities releasing tritium to the atmosphere: implications for environmental models

P.A. Thompson ^{a,*}, N.-O.A. Kwamena ^a, M. Ilin ^a, M. Wilk ^b, I.D. Clark ^b^a Canadian Nuclear Safety Commission, 280 Slater Street, P.O. Box 1046, Station B, Ottawa, Ontario, K1P 5S9, Canada^b Department of Earth Science, 140 Louis-Pasteur, University of Ottawa, Ottawa, Ontario, K1N 6N5, Canada

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ABSTRACT

Concentrations of organically bound tritium (OBT) and tritiated water (HTO) were measured over two growing seasons in vegetation and soil samples obtained in the vicinity of four nuclear facilities and two background locations in Canada. At the background locations, with few exceptions, OBT concentrations were higher than HTO concentrations: OBT/HTO ratios in vegetation varied between 0.3 and 20 and values in soil varied between 2.7 and 15. In the vicinity of the four nuclear facilities OBT/HTO ratios in vegetation and soils deviated from the expected mean value of 0.7, which is used as a default value in environmental transfer models. Ratios of the OBT activity concentration in plants ($[OBT]_{plant}$) to the OBT activity concentration in soils ($[OBT]_{soil}$) appear to be a good indicator of the long-term behaviour of tritium in soil and vegetation. In general, OBT activity concentrations in soils were nearly equal to OBT activity concentrations in plants in the vicinity of the two nuclear power plants. $[OBT]_{plant}/[OBT]_{soil}$ ratios considerably below unity observed at one nuclear processing facility represents historically higher levels of tritium in the environment. The results of our study reflect the dynamic nature of HTO retention and OBT formation in vegetation and soil during the growing season. Our data support the mounting evidence suggesting that some parameters used in environmental transfer models approved for regulatory assessments should be revisited to better account for the behavior of HTO and OBT in the environment and to ensure that modelled estimates (e.g., plant OBT) are appropriately conservative.

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1. Introduction

Tritium is formed naturally in the upper atmosphere from the interaction of gases and cosmic radiation. Human activities also contribute to tritium levels in the environment; a significant amount of tritium was released into the atmosphere as a result of thermonuclear bomb testing conducted primarily between 1954 and 1963. Tritium is also produced as a by-product of the operation of nuclear reactors such as the CANDU (CANada Deuterium Uranium) reactors. In Canada, a portion of the tritium produced by CANDU reactors is recovered and used by processing facilities to produce non-electrical self-luminescent lights and paints.

Tritium is mainly released in the form of tritiated water (HTO) vapour from the operation of CANDU reactors, as tritium gas (HT)

from tritium removal facilities and as both HTO and HT by tritium processing facilities. Once released into the atmosphere, HTO can be incorporated into plants through photosynthesis or deposited to soil and incorporated into soil moisture. It is now well accepted that HT deposited to soil is oxidized to HTO by soil micro-organisms before it can be incorporated into plants (McFarlane et al., 1979; Ichimasa et al., 1988; Yokoyama et al., 2004; Ota et al., 2007). The HTO that is formed will become part of the soil pore water or be re-emitted to the atmosphere as vapour. During metabolic activities, a portion of the HTO will become incorporated into organic molecules, including plant structural material or soil organic matter. This organically-bound tritium (OBT) has a significantly longer residence time in biological matrices than HTO.

Due to the relative complexity of measuring OBT, OBT/HTO ratios are generally used to predict OBT activities in the human food chain for regulatory assessment purposes. Current environmental transfer models assume that OBT forms directly from HTO and that under equilibrium conditions the OBT/HTO ratio is expected to be

* Corresponding author.

E-mail address: patsy.thompson@cnsccsn.gc.ca (P.A. Thompson).

in the range of 0.64–1.3, with a mean value of 0.7 (e.g., CSA, 2014). However, recent studies have shown that the OBT/HTO ratios in both plants and soils can be considerably higher and quite variable (Thompson et al., 2011; Kim et al., 2012a, b, 2013; Korolevych et al., 2014). Jean-Baptiste et al. (2011) reported that 77% of OBT/HTO ratios measured in plants and food items are greater than unity, with a mean value of 1.9.

The current study was undertaken to further investigate the relationship between HTO and OBT in soils and plants around Canadian nuclear facilities releasing either HTO or both HTO and HT. This was done by obtaining measurements of OBT and HTO in soil and vegetation samples near four nuclear facilities in Canada. This additional knowledge will aid in better understanding the fate of tritium in the terrestrial environment and improve how the transfer of tritium (HTO and HT) and the formation of OBT is modelled in environmental transfer models.

2. Materials and methods

2.1. Field site description

Tritium was measured in soil and vegetation over two field seasons (2008 and 2009). Samples were obtained near four Canadian nuclear facilities that have been in operation for approximately two decades. Table 1 summarizes the tritium (HT and HTO) emissions from the four nuclear facilities in 2008 and 2009. Samples were also obtained from two background locations.

The Darlington nuclear site (Darlington), located in Clarington, Ontario, includes four CANDU reactors and one tritium removal facility (TRF). The TRF began operating in 1990 while the four CANDU reactors started operating in the early 1990's. The facility releases HTO from the four CANDU reactors and HT from the TRF.

The Gentilly-2 nuclear generating station (Gentilly-2) was in operation from 1987 to 2012. The station consisted of one CANDU reactor which released HTO into the atmosphere. The Gentilly-2 nuclear generating station was included in the study since it is not in the vicinity of the other nuclear facilities in southeastern Ontario and only releases tritium in the form of HTO.

Shield Source Inc. (SSI) and SRB Technologies (SRBT) are tritium processing facilities located in Peterborough and Pembroke, Ontario, respectively. SSI operated from 1986 to 2013 while SRBT has been operating since 1990. SSI and SRBT both release HTO and HT but in differing proportions (Table 1).

Two background locations were chosen for the study: one background site was located in Russell, Ontario, more than 180 km away from SRBT, the closest source of tritium. The other background sites were located near the towns of Warman and Langenburg, Saskatchewan. There are no industrial sources of tritium in Saskatchewan; the only significant source other than natural background was from weapons testing in the 1950's and 1960's.

2.2. Environmental sampling

Soil and vegetation samples were obtained near four Canadian nuclear facilities and two background locations at the end of the

2008 and 2009 growing seasons. Vegetation samples included garden fruits and vegetables for human consumption as well as grass and corn for animal feed.

2.2.1. Vegetation samples

At each of the four nuclear facilities, a network of ten gardens and/or farms was selected to cover different distances from the facility. Sampling locations around each nuclear facility were selected based on the prevailing wind conditions, if possible (Vichot et al., 2008). Samples from home gardens and local farms were obtained at multiple distances between 0.4 and 11 km from the two tritium processing facilities. Samples obtained in the vicinity of the two nuclear generating stations were obtained outside of exclusion zones and at distances varying between 2.6 and 20 km. One or two gardens were also sampled at the background locations.

Fruits and vegetables were grown in gardens by local residents living near the nuclear facilities and in the background locations. Both above- and below-ground vegetables and fruits were obtained as summarized in the Table 2. Two or more fruits and/or vegetables were collected at each garden. Samples of grass and corn were also obtained from local farms. All samples were collected by the researchers. The samples were made free of adhering dirt but were not washed. The amount of sample collected varied from a few grams to a few hundreds of grams. Field samples were placed and sealed in freezer zip-lock bags and temporarily stored in a thermally insulated box. The samples were kept frozen at $-15\text{ }^{\circ}\text{C}$ until analysis.

The selection of vegetation obtained depended on what was grown by the local residents and farmers. Similar types of vegetation were obtained at locations near nuclear facilities and the background locations.

2.2.2. Soil samples

Composite soil samples were obtained at each garden using a stainless steel Oakley soil corer (2 cm i.d., 30 cm length). Fifteen soil cores (approximately 900 g total) were taken from each garden to a depth of 20 cm. The cores were mixed together before being split into three-300 g aliquots in zip-lock freezer bags. The samples were kept frozen at $-15\text{ }^{\circ}\text{C}$ until analysis.

2.3. Analytical methods

Analytical methods for determining the HTO and OBT concentrations in environmental samples are described in detail in Clark et al. (2010) and are briefly summarized below.

2.3.1. Extraction of HTO in soil and vegetation samples

The HTO measured in soil and vegetation samples is referred to as "tissue-free-water-tritium" (TFWT). The original weight of the sample, fresh weight, was determined before storage in the freezer. Losses due to sublimation were negligible. The difference between the weight before freezing and the weight after retrieval from frozen storage was less than 1%.

TFWT was extracted from soil and vegetation samples using a vacuum extraction line. Prior to attaching the samples to the

Table 1
Tritium emissions (in TBq) from the four nuclear facilities in 2008 and 2009.

Nuclear facility	2008		2009	
	HT	HTO	HT	HTO
SRB technology	33.7	6.4	28.3	14.3
Shield source inc.	98.9	28.5	83.5	6.2
Darlington nuclear generating station	83	160	54	170
Gentilly-2 nuclear generating station	n/a	170	n/a	190

Table 2
Type of vegetation obtained from local gardens and farms near four Canadian nuclear facilities and background locations.

Type of vegetation		
Vegetables	Above-ground	Beans, cucumbers, tomatoes, zucchini, squash
	Below-ground	Carrots, potatoes, beets
Fruit	Apples, pears	
Animal feed	Corn, grass	

extraction line, vegetation samples were thawed and ground using a kitchen rotary grinder to increase the surface area-to-volume ratio. The vegetation samples were weighed after being ground. Soil samples were used as received without further processing. Soil samples were weighed as is once the sample was thawed. TFWT was collected from the vacuum extraction line using a liquid nitrogen trap.

Once the TFWT was extracted from the sample, the remaining sample was dried in an oven at 60 °C to a constant weight. The final dried organic material was then re-saturated once or twice with low-tritium water, as needed, to reach background levels and to remove any remaining “exchangeable” tritium. The exchangeable tritium includes the organically-bound tritium that is bound to sulphur, nitrogen and oxygen atoms (Diabeté and Strack, 1993). This fraction behaves in a similar fashion to TFWT (Boyer et al., 2009). The rinse water was analyzed by liquid scintillation counting to ensure that the exchangeable tritium had been removed and to confirm that the tritium content of the rinse water was at background levels. If the tritium concentration in the rinse water was not at background levels the sample was rinsed again with low-tritium water. This process was repeated until the tritium in the rinse water was at background levels. It was determined that the exchangeable tritium represented approximately 5% of the measured non-exchangeable OBT.

Samples of the extracted TFWT were analyzed by liquid scintillation counting (detailed below). Measured tritium activities (Bq/L) were corrected to account for the difference between the date of collection and the date of sample analysis. The HTO measurements in this work correspond to the sum of the activities of TFWT and exchangeable tritium.

2.3.2. Combustion method for determining OBT in vegetation samples

The OBT activity was measured in the dry matter remaining after the TFWT and the exchangeable tritium had been extracted from the sample. The measured tritium activity, therefore, represents “non-exchangeable” tritium.

The OBT activity in vegetation samples was determined using the Parr Bomb combustion method and subsequent liquid scintillation counting of the collected combustion water. For each combustion process, up to 10 g of dried organic matter was weighed and placed in a 1000 cm³ Parr autoclave combustion cell (Model 1121 large capacity bomb with a T316SS two-valve pressure gauge) along with a loop of fuse wire and sealed. The system was flushed with oxygen and brought to an oxygen pressure of 300–350 psi. After ignition, the cell was cooled and the water in the chamber was collected and weighed. The combustion water from the sample was counted using liquid scintillation counting.

2.3.3. Combustion method for determining OBT in soil samples

The low organic content in most soils precluded OBT analysis by conventional Parr bomb combustion. Two identical Lindberg clamshell tube ovens were placed in series to provide two zones of independent heating capability. Two 25-mm quartz furnace tubes were placed inside the tube ovens. The first zone contained the soil sample in a quartz combustion tube. The second zone was occupied by a copper oxide catalyst. A carrier gas of dry compressed air was introduced to the furnace tube to oxidize the organic components of the soil. The combustion products were then swept inline over the hot copper oxide catalyst where a second gas feed of pure oxygen was added to complete combustion to CO₂ and H₂O. The combustion products were carried to cold traps kept at –90 °C, where only water vapour was retained. The captured water was weighed for yield and then diluted with tritium-free water and counted for tritium activity using a liquid scintillation counter.

2.3.4. Liquid scintillation counting

Samples were analyzed for tritium activity by liquid scintillation counting. Ten grams of water from the sample was mixed with 10 g of Ultima Gold scintillation cocktail. The samples were counted using a Perkin Elmer Quantulus 1220 low-background liquid scintillation counter. The average of three separate counts was taken. A suite of standards was used for calibration. The tritium activity concentrations were calculated by comparing the activity of the sample to an external standards calibration curve.

In this work, the HTO activity was reported as Bq/L and the OBT activity in the samples was reported in units of Bq/L of combustion water. The activity measurements from liquid scintillation counting (Bq/L) were used to compute the measured tritium activity concentrations for the environmental samples.

2.3.5. Quality assurance and limits of detection and quantification

The limit of detection (LOD) and the limit of quantification (LOQ) for tritium measurements made by liquid scintillation counting were based on the repeat measurement of the laboratory blank (melted mid-Holocene glacier water from the Agassiz ice cap), which gave a standard deviation of 0.39 Bq/L. The LOD is taken as three times the standard deviation of these replicate runs (1.2 Bq/L), while the LOQ is five times this value (2.0 Bq/L).

Intra- and inter-laboratory replicates were analyzed to assure quality assurance and quality control.

3. Results and discussion

3.1. HTO and OBT measurements in soils and vegetation at background locations

Soil and vegetation samples from two background locations were obtained and analyzed. In total, 43 samples were analyzed for HTO and a subset of 19 samples was also analyzed for OBT.

The soil and vegetation samples taken from background locations in this study had HTO activity concentrations between 0.7 and 4.6 Bq/L (Tables A1 and A2 of Appendix A). The HTO activity concentrations measured in the samples obtained from Russell, Ontario (1.2–4.6 Bq/L) were similar to the HTO concentrations which have been measured in vegetation samples from other background locations in Ontario (OPG, 2010; BP, 2010). The measured activity concentrations in the samples obtained from the two background locations in Saskatchewan (0.8–2.6 Bq/L) were lower than the HTO activity concentrations obtained for the samples from Russell, Ontario, and HTO activity concentrations in the literature. The two background locations in Saskatchewan are removed from industrial sources of tritium. Therefore, HTO concentrations were expected to be lower than samples obtained near Russell, Ontario, which is located closer to sources of tritium.

The range in OBT activity concentrations in samples from the background locations was between 1.0 and 24 Bq/L. The highest OBT concentrations were observed in two samples (soil and tomato) from the background locations in Saskatchewan. Similarly high OBT concentrations have previously been measured by others at background locations. For example, OPG (2003) reported values of 19.1 Bq/L in tomatoes from Lakefield, Ontario, and Kim and Davis (2008) reported activity concentrations as high as 36.1 and 65 Bq/L in apples from Bancroft, Ontario, and bean samples from Kingston, Ontario, respectively.

OBT activity concentrations were higher than HTO activity concentrations in most vegetation samples (13 out of 16 samples). OBT/HTO ratios in vegetation varied between 0.3 and 20 (Tables A1 and A2 in Appendix A). A limited number of soil samples (3) were collected. Soil OBT/HTO values were between 2.7 and 15. In general, OBT/HTO ratios were highest in the soil and vegetation samples

from Saskatchewan where HTO activity concentrations were the lowest. The OBT/HTO ratios measured in samples from background locations in this work are within the range of values found in the literature (Korolevych et al., 2014).

3.2. HTO and OBT measurements in soils and vegetation in the vicinity of four nuclear facilities

In total, 236 samples were analyzed for HTO and a subset of 84 samples was also analyzed for OBT. Tables A3 to A6 of Appendix A present the HTO and OBT activity concentrations in soil and vegetation samples along with their respective OBT/HTO ratios for each of the four nuclear facilities.

In general, HTO activity concentrations were found to decrease with increasing distance from a nuclear facility (data not shown), which is consistent with similar measurements in the literature (Ilin et al., 2005). This correlation was most pronounced in samples obtained near the two nuclear processing facilities where samples could be obtained at closer distances from the facility and over a wider distance range. No clear relationship between OBT activity concentration and distance from the facility could be extracted from the data for any of the nuclear facilities investigated (data not shown).

Tables 3 and 4 summarize the mean OBT/HTO ratios at the four nuclear facilities and the two background locations for soil and vegetation, respectively. The arithmetic and geometric means are reported along with the equivalent of one standard deviation. In the following text, reference is only made to the geometric mean and geometric standard deviation.

Environmental transfer models approved for regulatory assessments estimate plant OBT from measured plant HTO activity based on an OBT/HTO ratio of 0.7 (CSA, 2014). The ratio is expected to be less than unity due to isotopic discrimination in chemical and biological processes, such as photosynthesis, due to tritium having a greater mass than hydrogen (Boyer et al., 2009). In this work, OBT/HTO ratios for soil and vegetation samples obtained at background locations and around the four nuclear facilities were generally found to be above the expected value of 0.7. Other studies in the literature have also reported deviations from the expected ratio of 0.7 (OPG, 2003; Kim and Davies, 2008; Kim et al., 2013).

Fifteen of the 22 soil samples obtained near nuclear facilities had OBT/HTO ratios greater than 0.7. Year-to-year fluctuations were observed between the soil samples obtained in 2008 and 2009. Kim et al. (2014) also observed year-to-year variations in the OBT/HTO ratios for soil samples obtained around the Darlington NGS. In our work, the highest OBT/HTO ratios for soil were observed in samples obtained near SRBT (Table A3), with a mean OBT/HTO ratio of 5.6 ± 1.6 . The enrichment of OBT over HTO is likely a result of a high

Table 3
OBT/HTO ratios for soil samples from all sampling locations.

Location	Sample size (n)	Range	Arithmetic mean \pm SD	Geometric mean \pm Geometric SD
Background locations				
Russell, Ontario	2	2.7–3.5	3.1 ± 0.6	3.1 ± 1.2
Langenburg and Warman, Saskatchewan	1	15	15	n/d
Nuclear facilities				
SRBT	4	3.5–9.9	6.5 ± 3.3	5.6 ± 1.6
SSI	6	0.4–3.5	1.4 ± 1.3	1.0 ± 2.2
Darlington	6	0.2–1.9	1.2 ± 0.7	0.9 ± 1.7
Gentilly-2	6	0.3–4.2	1.6 ± 1.4	1.1 ± 2.1

n/d = not determined.

Table 4
OBT/HTO ratios for vegetation samples from all sampling locations.

Location	Sample size (n)	Range	Arithmetic mean \pm SD	Geometric mean \pm Geometric SD
Background locations				
Russell, Ontario	7	0.3–5.0	1.7 ± 1.7	1.2 ± 2.3
Langenburg and Warman, Saskatchewan	9	3.1–20.1	9.0 ± 6.8	6.9 ± 1.9
Nuclear facilities				
SRBT	13	0.9–15.4	3.3 ± 3.9	2.2 ± 2.6
SSI	24	0.4–4.8	1.8 ± 1.3	1.4 ± 1.9
Darlington	13	0.4–2.6	1.2 ± 0.6	1.0 ± 1.7
Gentilly-2	12	0.5–14.3	2.4 ± 3.8	1.4 ± 3.1

inventory of OBT that exists in the region due to historical emissions. The mean OBT/HTO ratios for soil samples collected near the remaining three facilities were similar, ranging from 0.9 ± 1.7 to 1.1 ± 2.1 .

Most vegetation samples had OBT/HTO ratios greater than 0.7 (55 of 62 samples). The mean OBT/HTO ratios for vegetation samples collected near the four nuclear facilities ranged from 1.0 ± 1.7 to 2.2 ± 2.6 and are comparable to values obtained elsewhere (Korolevych et al., 2014).

OBT/HTO ratios were considered high if they were above 10. In our work, two OBT/HTO ratios were observed to be greater than 10: a ratio of 15.4 for a cucumber sample and one of 14.3 for a corn sample at SRBT and Gentilly-2, respectively. If the high OBT/HTO ratios are removed, the mean of the OBT/HTO ratios for vegetation samples obtained near Gentilly-2 would be 1.1 ± 1.7 , which is similar to those ratios obtained for SSI and the Darlington nuclear generating station. Similarly, if the one high OBT/HTO ratio was removed for SRBT, the mean of the OBT/HTO ratios would be 1.9 ± 1.8 . The slightly higher OBT/HTO ratio for SRBT is again likely due to the higher inventory of OBT near the nuclear processing facility. Nonetheless, even if these values were removed from the dataset, the conclusion remains the same: the OBT/HTO ratios are greater than the expected value of 0.7.

In the natural environment HTO retention and rates of OBT formation are dynamic and affected by many environmental factors (Kim and Korolevych, 2013; Boyer et al., 2009). The activity concentrations of HTO in plants and soil are influenced by the HTO in the air and soil pore water at the time of sampling (Kim and Korolevych, 2013). Consequently, HTO activity concentrations reflect the HTO concentrations in the hours prior to sampling. In contrast, OBT activity concentrations in plants and soil do not respond as quickly due to OBT's longer residence time. Current models, do not take into account the variations in HTO and OBT over the growing period (Strack et al., 1995; Boyer et al., 2009; Baglan et al., 2011).

Since the vegetation samples were obtained at the end of the growing season, the OBT/HTO ratios above 0.7 measured in our study likely represent the accumulation of plant organic matter in the form of OBT during the growing season and the dynamic nature of HTO incorporation from the atmosphere at the time of sampling.

3.3. Partitioning of HTO in soils and vegetation

The specific activity model is included in environmental transfer models that are used for regulatory assessments (CSA, 2014). Based on the specific activity model, the HTO specific activity in plants is assumed to be similar to the HTO specific activity in air with a slight reduction to account for the contribution of HTO from soil water (equation (1)).

$$[\text{HTO}]_{\text{plant}} = 0.68[\text{HTO}]_{\text{air}} \quad (1)$$

where $[\text{HTO}]_{\text{plant}}$ is the HTO specific activity in plants and $[\text{HTO}]_{\text{air}}$ is the HTO specific activity in air.

Similarly, soil water is assumed to have the same specific activity as air, but must be corrected to account for dilution by precipitation (equation (2)).

$$[\text{HTO}]_{\text{soil}} = 0.3[\text{HTO}]_{\text{air}} \quad (2)$$

where $[\text{HTO}]_{\text{soil}}$ is the HTO specific activity in soil.

The default empirical value for the ratio of HTO in soil to that in air moisture is 0.3. Based on equations (1) and (2), the ratio of $[\text{HTO}]_{\text{plant}}/[\text{HTO}]_{\text{soil}}$ is expected to have a value of 2.3.

Table 5 summarizes the ratio $[\text{HTO}]_{\text{plant}}/[\text{HTO}]_{\text{soil}}$ for all samples obtained at the background locations and near nuclear facilities. The arithmetic and geometric means are reported along with the equivalent of one standard deviation. The ratios obtained from samples at background locations were found to vary between 0.8 and 1.8 (mean 1.2 ± 1.4). Similarly, the ratios of $[\text{HTO}]_{\text{plant}}/[\text{HTO}]_{\text{soil}}$ for samples collected around the four nuclear facilities were between 0.5 and 1.8 (mean 0.8 ± 1.7). All of these ratios were lower than the expected value of 2.3 which is used in environmental transfer models. The values reported in this study were within the range of values reported by Kim et al. (2012a).

The expected value of the $[\text{HTO}]_{\text{plants}}/[\text{HTO}]_{\text{soil}}$ ratio is 2.3, which is an average since HTO in plants responds more quickly to changes in air concentrations than soil pore water (Kim et al., 2012). Mean values of close to unity for the ratio suggests similar partitioning of HTO in vegetation and soils at the time of sampling. Similar values at background locations and near nuclear facilities suggest a similar mechanism for the partitioning of HTO in soil and vegetation.

3.4. Partitioning of OBT in soils and vegetation

Photosynthesis is the most important biological process for the conversion of HTO to OBT (Diabeté and Strack, 1993; Vichot et al., 2008). These different forms of tritium (OBT and HTO) have different residence times in soil and vegetation. HTO in plants and soil pore water reflects current steady-state conditions and the HTO in the atmosphere in the hours prior to sampling. Conversely, OBT activity concentrations in plants and soil are an integration of tritium releases during the growing season and over the long-term, respectively. As such, OBT measurements provide a more accurate representation of tritium in the environment because of OBT's longer retention time (Kim and Roche, 2012).

In this work, the amount of OBT produced in the plant over the growing season ($[\text{OBT}]_{\text{plant}}$) was compared to the amount of OBT in

the soil ($[\text{OBT}]_{\text{soil}}$) (Table 6). The amount of OBT that may be formed in the plant depends on the type of vegetation and other environmental parameters (Kim and Korolevych, 2013; Kim et al., 2012b). Much of the OBT in soil represents tritium that had been incorporated into soil organic matter. The amount of soil organic matter is controlled by two factors: input by net primary production and the decomposition rate of organic matter. Therefore, since the soil reservoir acts as a repository for decaying organic matter, the OBT concentration in soil represents a long-term tritium reservoir of historical tritium releases.

The ratio of $[\text{OBT}]_{\text{plant}}/[\text{OBT}]_{\text{soil}}$ was calculated to compare the amount of OBT formed in the plant during the growing season to the amount of OBT in the longer-term reservoir (i.e., soil) (Table 6). Low values of this ratio would indicate higher soil OBT activity concentrations due to higher historical releases. From our limited data set of co-occurring soil and plant samples, it was observed that the $[\text{OBT}]_{\text{plant}}/[\text{OBT}]_{\text{soil}}$ ratio varied between 0.07 and 3.5 (mean 0.6 ± 2.2) for samples obtained near the four nuclear facilities. The lowest $[\text{OBT}]_{\text{plant}}/[\text{OBT}]_{\text{soil}}$ ratio was observed near SRBT and the highest ratio was observed in a sample obtained near Gently-2 (Table 6). The low ratio obtained near SRBT likely represents soil organic matter bearing the signature of the historically high releases. In the vicinity of other nuclear facilities and the background locations, OBT in soils appear to be more similar to OBT in plants; this is likely representative of fairly constant tritium in air concentrations from year to year.

Given that organic matter can reside in soils for decades (Lutzow et al., 2006), a fraction of the OBT measured in soils at background locations may represent tritium incorporated into plants when tritium concentrations were elevated in the atmosphere due to bomb testing. In the northern hemisphere, bomb testing raised tritium concentrations in precipitation to levels around 470 Bq/L. Although the first phase of decomposition of natural organic matter in soils shows a turnover rate of about 1–2 years (Lutzow et al., 2006), the second phase which consists of slow decomposition, accounts for a total loss of about 90% of the organic matter in soils and can last decades (Lutzow et al., 2006).

Our samples were collected in cultivated systems; in such systems, the mean residence time of organic matter in soils has been observed to range from 26 to 400 years (Six and Jastrow, 2002). The low ratio observed in our work in a sample obtained from near SRBT can be explained by the release of greater quantities of HT and HTO from the facility in the period from 2000 to 2007 before sampling took place. Given the mean residence time of organic matter in soil, the OBT concentration in soil samples obtained near SRBT still reflects the previous high emissions from the facility. Kim et al. (2013) have also measured high OBT activity concentrations in soil organic matter many years after the source of tritium had been removed.

Table 5

Partitioning of HTO in vegetation and soil ($[\text{HTO}]_{\text{plant}}/[\text{HTO}]_{\text{soil}}$) in samples obtained near background locations and nuclear facilities.

Location	Description	Sample size (n)	Arithmetic mean \pm SD	Geometric mean \pm Geometric SD
Russell, Ontario	Background	2	1.2 ± 0.5	1.2 ± 1.4
Warman and Langenburg, Saskatchewan		1		
SRBT	Nuclear	5	1.0 ± 0.5	1.0 ± 1.5
SSI	processing facility	8	0.5 ± 0.2	0.5 ± 1.4
Darlington	Nuclear power plant	5	1.0 ± 0.3	1.0 ± 1.4
Gently-2	All nuclear facilities	20	1.8 ± 0.0	1.7 ± 1.0
			0.9 ± 0.5	0.8 ± 1.7

Table 6

Partitioning of OBT in vegetation and soil ($[\text{OBT}]_{\text{plant}}/[\text{OBT}]_{\text{soil}}$) in samples obtained near background locations and nuclear facilities.

Location	Description	Sample size (n)	Arithmetic mean \pm SD	Geometric mean \pm Geometric SD
Russell, Ontario	Background	2	0.8 ± 0.5	0.7 ± 1.7
Warman and Langenburg, Saskatchewan		1		
SRBT	Nuclear	5	0.2 ± 0.2	0.2 ± 1.8
SSI	processing facility	8	0.9 ± 0.7	0.7 ± 2.0
Darlington	Nuclear power plant	5	1.0 ± 0.6	0.9 ± 1.7
Gently-2	All nuclear facilities	20	2.3 ± 1.7	1.9 ± 2.0
			0.9 ± 0.9	0.6 ± 2.2

4. Conclusion

The current study conducted over two growing seasons measured HTO and OBT in vegetation and soil samples obtained in the vicinity of four nuclear facilities and two background locations. Our data on OBT activity concentrations in vegetation samples confirm results from many previous studies (Jean-Baptiste et al., 2011) that OBT/HTO ratios in vegetation can be considerably higher than the value of 0.64–1.3 (mean value 0.7) used in environmental transfer models to estimate OBT activity concentrations from measured HTO activity concentrations (e.g., CSA, 2014). Taking into consideration the dynamic nature of HTO retention and OBT formation and the accumulation of organic compounds through the growing season, these results are not surprising.

The HTO activity concentrations in plants relative to activity concentrations in soil obtained in this study also deviate from expected values used in environmental transfer models (e.g., CSA, 2014). These findings are also consistent with measurements obtained by Kim et al. (2012a).

Overall, the results of the current study add to the mounting evidence that simple environmental transfer models may not adequately assess the behavior of HTO and OBT in different environmental compartments. The accumulated data also suggest that some of the parameters used in environmental transfer models approved for regulatory assessments need to be re-visited to better account for data obtained both at background locations and near nuclear facilities and to ensure that modelled estimates are appropriately conservative.

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Appendix A

Table A1

HTO and OBT measurements in soil and vegetation samples taken from the background location in Russell, Ontario.

Sample ID (year-location)	Type of sample	Water content	HTO		OBT			OBT/HTO
			TFWT (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/L)	
Rus-08-01	Soil 2008	0.21	3.0	0.7	10.4	0.8	0.3	3.5
Rus-09-01	Soil 2009	0.25	3.2	0.8	8.5	0.5	0.4	2.7
Rus-08-01	Carrot 2008	0.88	3.6	0.7	7.5	2.6	1.7	2.1
Rus-09-01	Carrot 2009	0.90	2.4	0.8	12.1	3.5	0.1	5.0
Rus-09-01	Grass 2009	0.79	3.5	0.8	4.7	1.7	0.0	1.3
Rus-08-02	Corn leaves 2008	0.77	2.6	0.8	2.2	0.8	0.0	0.8
Rus-09-02	Corn leaves 2009	0.77	4.6	0.8	5.4	2.2	1.7	1.2
Rus-08-02	Corn cob + leaves 2008	0.82	3.3	0.8	1.0	0.5	1.6	0.3
Rus-09-02	Corn cob + leaves 2009	0.75	3.8	0.8	2.6	1.3	1.0	0.7

Table A2

HTO and OBT measurements in soil and vegetation samples taken from the background locations in Langenburg and Warman, Saskatchewan.

Sample ID (year-location)	Type of sample	Water content	HTO		OBT			OBT/HTO
			TFWT (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/L)	
Langenburg, Saskatchewan								
2009-Langenburg	Soil 2009	0.17	1.3	0.9	19.9	0.6	0.0	15.0
2008-Langenburg#1	Tomato 2008	0.92	1.2	0.7	23.7	5.0	1.5	20.1
2009-Langenburg#1	Tomato 2009	0.91	2.6	1.0	8.2	3.7	1.7	3.1
2008-Langenburg#3	Cucumber 2008	0.97	1.0	0.7	18.5	4.3	2.7	17.8
2009-Langenburg	Squash 2009	0.90	2.4	0.9	7.8	3.6	1.5	3.2
Warman, Saskatchewan								
2008-Warman#4	Tomato 2008	0.93	1.5	0.8	18.9	6.2	2.2	13.0
2008-Warman Park#5	Crabapple 2008	0.82	0.7	0.7	8.7	3.6	1.3	12.0
2009-Warman	Apple 2009	0.85	1.6	0.9	6.4	3.2	1.3	4.1
2009-Warman	Apple duplicate 2009	0.86	1.1	0.9	5.1	2.4	1.2	4.8
2009-Warman	Berries 2009	0.80	2.2	0.9	7.1	3.3	1.5	3.2

Table A3

HTO and OBT measurements in soil and vegetation samples in the vicinity of SRB Technologies, a tritium processing facility.

Sample ID (year-location)	Type of sample	Water content	Distance from facility (km)	HTO		OBT			OBT/HTO ratio
				Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	
Soils									
Pem-08-01	Soil 2008-1	0.19	0.4	102.0	2.3	1010.0	53.1	1.2	9.9
Pem-09-01	Soil 2009-1	0.22	0.4	122.0	2.5	493.0	8.1	0.3	4.0
Pem-08-12	Soil 2008-2	0.24	3.8	4.3	0.8	14.9	1.0	0.4	3.5
Pem-09-12	Soil 2009-2	0.24	3.8	6.3	0.9	55.2	2.6	0.5	8.8
Vegetation - garden produce									
Pem-09-01	Apple 2009	0.87	0.4	224.0	3.7	234.0	96.1	3.1	1.0
Pem-08-01	Carrot 2008	0.91	0.4	75.5	1.9	69.1	22.1	3.1	0.9
Pem-09-01	Carrot 2009	0.9	0.4	116.0	2.5	103.0	25.4	2.4	0.9

Table A3 (continued)

Sample ID (year-location)	Type of sample	Water content	Distance from facility (km)	HTO		OBT			OBT/HTO ratio
				Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	
Pem-08-01	Potato 2008	0.77	0.4	76.0	1.9	206.0	81.8	1.7	2.7
Pem-09-01	Potato 2009	0.75	0.4	105.0	2.3	90.0	42.9	2.2	0.9
Pem-08-05	Tomato 2008	0.94	2.0	15.3	1.0	66.3	7.9	6.6	4.3
Pem-09-05	Tomato 2009	0.94	2.0	17.7	1.1	40.3	10.4	3.4	2.3
Pem-09-09	Cucumber 2009	0.95	4.8	7.6	0.9	117.0	7.9	6.3	15.4
Vegetation – animal feed									
Pem-09-10	Grass 2009	0.75	4.1	7.3	0.9	31.3	13.0	9.8	4.3
Pem-08-13	Corn Leaves 2008	0.77	2.7	7.0	0.8	34.1	7.7	2.4	4.9
Pem-09-13	Corn Leaves 2009	0.79	2.7	9.2	1.2	13.9	5.2	1.9	1.5
Pem-08-13	Corn Cob 2008	0.77	2.7	8.3	0.9	15.8	7.2	1.9	1.9
Pem-09-13	Corn Cob 2009	0.85	2.7	12.7	1.2	18.4	8.7	2.1	1.4

Table A4

HTO and OBT measurements in soil and vegetation samples in the vicinity of SSI, a tritium processing facility.

Sample ID (year-location)	Type of sample	Water content	Distance from facility (km)	HTO		OBT			OBT/HTO ratio
				Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	
Soils									
Pt-08-01	Soil 2008-1	0.19	3.4	49.7	3.8	21.9	0.7	0.3	0.4
Pt-09-01	Soil 2009-1	0.14	3.4	56.2	0.9	127.0	7.0	0.5	2.3
Pt-08-03	Soil 2008-2	0.14	3.4	60.0	0.9	22.7	0.4	0.0	0.4
Pt-09-03	Soil 2009-2	0.09	3.4	103.0	0.9	167.0	2.8	0.4	1.6
Pt-08-10	Soil 2008-3	0.25	5.6	58.3	1.0	21.9	1.3	0.4	0.4
Pt-09-10	Soil 2009-3	0.17	5.6	69.9	1.0	245.0	12.1	0.6	3.5
Vegetation – garden produce									
Pt-08-10	Apples 2008	0.84	5.6	40.4	0.8	55.6	12.2	7.6	1.4
Pt-09-10	Apples 2009	0.86	5.6	24.8	0.8	72.0	37.1	3.2	2.9
Pt-08-01	Beans 2008	0.91	3.4	21.8	1.2	16.9	7.7	9.6	0.8
Pt-09-01	Beans 2009	0.91	3.4	19.6	1.4	94.1	40.4	2.6	4.8
Pt-08-01	Tomato 2008-1	0.93	3.4	26.9	1.4	23.6	8.4	2.9	0.9
Pt-09-01	Tomato 2009-1	0.93	3.4	41.3	1.8	74.6	30.9	3.1	1.8
Pt-08-03	Tomato 2008-2	0.95	3.4	27.4	3.5	25.8	9.5	5.3	0.9
Pt-09-03	Tomato 2009-2	0.96	3.4	30.8	1.6	38.9	11.9	5.9	1.3
Pt-08-05	Potato 2008	0.84	0.9	456.0	6.3	170.0	72.7	14.2	0.4
Pt-09-05	Potato 2009	0.79	0.9	1010.0	14.2	451.0	208.0	6.3	0.4
Pt-08-09	Carrot 2008-1	0.89	3.3	52.4	1.6	43.5	13.0	6.0	0.8
Pt-08-09	Carrot 2008-2	0.85	3.3	52.7	1.6	48.9	22.8	5.2	0.9
Vegetation – animal feed									
Pt-08-07	Corn Leaves 2008-1	0.79	8.1	11.1	1.0	13.5	5.9	1.9	1.2
Pt-09-07	Corn Leaves 2009-1	0.78	8.1	24.5	1.5	106.0	41.1	4.7	4.3
Pt-08-07	Corn Cob 2008-1	0.87	8.1	12.7	1.0	24.8	10.5	7.2	2.0
Pt-09-07	Corn Cob 2009-1	0.60	8.1	25.6	1.5	103.0	51.1	3.6	4.0
Pt-08-14	Grass 2008-1	0.72	5.6	22.4	1.2	12.0	4.2	2.4	0.5
Pt-09-14	Grass 2009-1	0.71	5.6	37.5	1.7	141.0	52.0	3.2	3.8
TPD-08-04	Grass 2008-2	0.62	16.0	4.5	0.8	12.9	4.6	2.6	2.8
TPD-09-04	Grass 2009-2	0.73	16.0	6.2	0.9	14.2	4.7	3.1	2.3
TPD-08-04	Corn Leaves 2008-2	0.78	16.0	6.7	0.9	8.2	3.4	2.2	1.2
TPD-09-04	Corn Leaves 2009-2	0.78	16.0	4.7	1.5	6.6	2.7	2.2	1.4
TPD-08-04	Corn Cob 2008-2	0.83	16.0	6.6	0.9	10.2	5.1	1.5	1.6
TPD-09-04	Corn Cob 2009-2	0.70	16.0	5.2	0.9	6.5	3.5	1.3	1.2

Table A5

HTO and OBT measurements in soil and vegetation samples in the vicinity of the Darlington nuclear facility, a nuclear power plant and a tritium removal facility.

Sample ID (year-location)	Type of sample	Water content	Distance from the facility (km)	HTO		OBT			OBT/HTO ratio
				Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	
Soils									
Dar-08-02	Soil 2008-1	0.16	2.6	18.4	1.1	28.9	0.5	0.3	1.6
Dar-09-02	Soil 2009-1	0.15	2.6	11.4	1.0	12.0	0.3	0.4	1.1
Dar-08-07	Soil 2008-2	0.20	5.2	9.8	1.0	1.8	0.1	0.3	0.2
Dar-09-07	Soil 2009-2	0.16	5.2	7.9	0.9	4.7	0.2	0.3	0.6
Dar-08-11	Soil 2008-3	0.17	5.7	16.3	1.1	26.8	0.9	0.4	1.6
Dar-09-11	Soil 2009-3	0.14	5.7	10.1	1.0	19.6	0.3	0.3	1.9
Vegetation – garden produce									
Dar-08-02	Apple 2008	0.86	2.6	15.7	1.1	16.9	7.5	1.5	1.1

(continued on next page)

Table A5 (continued)

Sample ID (year-location)	Type of sample	Water content	Distance from the facility (km)	HTO		OBT			OBT/HTO ratio
				Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	
Dar-09-02	Apple 2009	0.86	2.6	14.1	1.0	16.0	7.3	1.2	1.1
Dar-08-06	Tomato 2008	0.95	6.1	10.1	1.0	26.4	6.9	3.5	2.6
Dar-09-06	Tomato 2009	0.93	6.1	5.9	0.9	6.8	2.7	1.2	1.2
Dar-09-07	Zucchini 2009	0.95	5.2	10.8	1.0	9.3	3.9	1.9	0.9
Dar-08-08	Potato 2008	0.76	4.1	18.2	1.1	20.9	9.8	2.1	1.1
Dar-09-08	Potato 2009	0.74	4.1	21.1	1.2	10.2	5.0	1.7	0.5
Dar-08-11	Carrot 2008	0.90	5.7	8.1	0.9	17.0	5.9	2.3	2.1
Dar-09-11	Carrot 2009	0.87	5.7	11.6	1.0	12.1	5.3	1.4	1.0
Vegetation – animal feed									
Dar-08-14	Corn Leaves 2008	0.76	4.2	11.4	1.0	4.1	1.8	0.2	0.4
Dar-09-14	Corn Leaves 2009	0.79	4.2	15.9	1.1	11.1	4.3	2.0	0.7
Dar-08-14	Corn Cob 2008	0.80	4.2	10.7	1.0	16.8	7.4	1.8	1.6
Dar-09-14	Corn Cob 2009	0.72	4.2	15.8	1.1	12.7	5.0	1.1	0.8

Table A6

HTO and OBT measurements in soil and vegetation samples in the vicinity of the Gentilly-2 facility, a nuclear power plant.

Sample ID (year-location)	Type of sample	Water content	Distance from facility (km)	HTO		OBT			OBT/HTO ratio
				Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	
Soils									
Gen-08-02	Soil 2008-1	0.22	2.9	15.6	1.0	9.7	0.4	0.3	0.6
Gen-09-02	Soil 2009-1	0.16	2.9	6.6	1.1	7.4	0.2	0.3	1.1
Gen-08-06	Soil 2008-2	0.17	14.7	4.3	0.8	10.3	0.4	0.3	2.4
Gen-09-06	Soil 2009-2	0.13	14.7	3.9	1.4	16.2	0.5	0.3	4.2
Gen-08-17	Soil 2008-3	0.25	7.0	11.6	1.0	3.0	0.1	0.3	0.3
Gen-09-17	Soil 2009-3	0.23	7.0	8.8	1.0	10.3	0.4	0.3	1.2
Vegetation – garden produce									
Gen-08-17	Apple 2008	0.84	7.0	20.7	1.1	10.6	4.7	1.4	0.5
Gen-09-17	Apple 2009	0.85	7.0	15.5	1.4	11.0	5.3	1.6	0.7
Gen-08-01	Tomato 2008	0.95	1.5	11.3	1.0	12.2	4.6	2.2	1.1
Gen-09-01	Tomato 2009	0.92	1.5	12.2	1.0	11.8	5.2	1.4	1.0
Gen-08-02	Carrot 2008	0.88	2.9	6.9	0.9	17.0	3.8	4.8	2.5
Gen-09-20	Beans 2009	0.92	8.8	4.5	0.8	12.0	2.7	3.2	2.7
Gen-09-19	Potato 2009	0.78	9.4	4.3	0.8	3.3	0.8	-0.2	0.8
Vegetation – animal feed									
Gen-08-01	Corn Leaves 2008	0.77	1.5	10.6	1.0	14.1	5.4	2.2	1.3
Gen-09-01	Corn Leaves 2009	0.74	1.5	21.9	1.2	21.6	7.3	2.1	1.0
Gen-08-01	Corn Cob + Kernels 2008	0.84	1.5	13.5	1.0	193.0	29.5	6.3	14.3
Gen-09-01	Corn Cob + Kernels 2009	0.85	1.5	24.6	1.2	19.9	10.1	1.3	0.8
Gen-09-01	Grass 2009	0.76	1.5	10.0	1.0	16.7	6.7	3.7	1.7

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