



Environmental Fate of Tritium in Soil and Vegetation

Part of the Tritium Studies Project



December 2013



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PWGSC catalogue number CC172-101/2013E-PDF
ISBN 978-1-100-22687-3

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Également publié en français sous le titre de : Le devenir environnemental du tritium dans le sol et la végétation

Document availability

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Executive Summary

Background

Tritium is a radioactive isotope of hydrogen that occurs both naturally and as a by-product of the operation of nuclear and research reactors. It emits low-energy beta radiation, which can be absorbed by paper, plastic, glass or metal. Tritium can pose a health risk if it is ingested by drinking water or eating food that contains it, or if it is inhaled or absorbed through the skin in large quantities.

In Canada, the control of tritium releases to the environment is important, as Canadian-designed CANDU (CANada Deuterium Uranium) reactors produce more tritium than most other types of reactors. Some of the tritium generated in power reactors is recovered and used to produce self-luminescent lights and paints. Tritium is processed commercially in products such as exit signs, airport runway lights, watch dials and gun sights.

The Canadian Nuclear Safety Commission (CNSC) regulates and carefully monitors environmental releases of man-made tritium in order to protect the health and safety of Canadians and the environment. In 2007, the CNSC launched the Tritium Studies Project, a series of research studies on tritium releases in Canada to expand the body of knowledge on the subject and to further enhance regulatory oversight of tritium-related activities. One of those studies, *Tritium Activity in Garden Produce from Pembroke in 2007 and Dose to the Public*, revealed unexpected results regarding the ratio of organically bound tritium (OBT) to tritiated water (HTO) in soil and in a small number of food items – the OBT/HTO ratios were greater than 1, higher than the expected finding. Although the radiation doses from tritium are very low and orders of magnitude below doses known to cause health effects, the CNSC launched another study to better understand the mechanisms underlying the observed OBT/HTO ratios greater than 1.

This report describes the results of CNSC-funded research, conducted by the University of Ottawa, on the distribution of tritium in soils, garden produce, animal fodder and animal products near sites of long-term, sustained atmospheric releases of tritium gas and/or tritiated water near four nuclear facilities: SRB Technologies, Shield Source Incorporated, the Darlington Nuclear Generating Station and the Gentilly-2 Nuclear Generating Station.

Main findings

The findings of this research are summarized as follows:

- In general, HTO and OBT levels decrease as distance from a facility increases. This trend was most pronounced in environmental samples obtained near nuclear processing facilities, and was less pronounced near nuclear power plants.
- The results of this study do not indicate that soil type influences the partitioning of atmospheric tritium.
- The experimental approach did not provide insight into whether OBT was derived from atmospheric HTO or from tritiated hydrogen gas converted to HTO in soils.

- The OBT activity in tree rings may be a useful indicator of historical emissions under certain circumstances (i.e., at close distances to nuclear facilities).
- OBT activities in garden produce sampled in this study reflected the year-to-year changes in stack emissions.
- There was a large variation in OBT/HTO ratios for soils, vegetation, animal products and animal fodder.
- OBT/HTO ratios measured in environmental samples were generally greater than 1 near the various nuclear facilities.
- The dose from consuming produce with OBT/HTO ratios greater than 1 represents a small fraction of the annual public dose, and is orders of magnitude (between 250 and 2000 times) below doses known to cause health effects.

Conclusions

This CNSC-funded study has expanded the suite of measurements of HTO and OBT near nuclear facilities. The research found that, in general, OBT/HTO ratios measured in environmental samples were greater than 1 at the various nuclear facilities. This was consistent with the range of OBT/HTO ratios previously measured as part of the 2007 Pembroke study.

Because additional research questions remain, a follow-up research project has been undertaken. This project will involve performing detailed tritium monitoring experiments to address uncertainties in the cycling and behaviour of tritium released to the atmosphere.

The knowledge gained from this additional research will help improve model predictions and the monitoring of tritium in the environment, in order to better regulate nuclear facilities.

1. Introduction

1.1 Tritium in the environment

Tritium is a radioactive isotope of hydrogen that emits low-energy beta radiation, which can be absorbed by paper, plastic, glass or metal. It can pose a health risk if it is ingested by drinking water or consuming food, or if it is inhaled or absorbed through the skin in large quantities.

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 environment 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, which are used to generate electricity in Canada. Some of the tritium produced by nuclear reactors is recovered and used by processing facilities for the manufacture of non-electrical self-luminescent lights and paints (e.g., those used in exit signs, airport runway lights, watch dials and gun sights).

Tritium occurs in the atmosphere in the same chemical forms as hydrogen. As such, tritium easily forms water molecules and is found in the environment wherever water is present. Once tritium is in the air it rapidly mixes with air moisture and exchanges with water in other environmental reservoirs such as soil, plants and animals. The most important atmospheric forms of tritium are tritiated hydrogen gas (HT) and tritiated water (HTO). Natural tritium is primarily found in the environment as HTO. CANDU reactors release greater amounts of tritium in the form of HTO whereas tritium removal facilities and tritium processing facilities emit a larger fraction of HT.

HT is up to 25,000 times less harmful than HTO because it is only weakly absorbed by the body. HT can be slowly converted to HTO in the environment through isotopic exchange with moisture. Tritium gas can also be oxidized in plant foliage or it can diffuse into soil where it is quickly oxidized to HTO by microorganisms.

Organically bound tritium (OBT) is another important form of this radionuclide that is formed by the conversion of HTO in biological systems and organisms through various metabolic processes including photosynthesis in plants and decomposition of organic matter in soil. OBT is incorporated into organic compounds such as sugars, proteins, starches, lipids, cell structural materials and amino acids. HTO is very mobile compared to OBT. Furthermore, OBT has a significantly longer retention time in biological organisms and systems than HTO. The longer retention time leads to higher dose consequences of OBT relative to HTO.

1.2 Tritium monitoring at nuclear facilities

The Canadian Nuclear Safety Commission (CNSC) regulates and carefully monitors environmental releases of man-made tritium from nuclear reactors and tritium processing facilities in order to protect the health and safety of Canadians and the environment. The main objective of this study was to investigate the behaviour of tritium released from nuclear facilities in the terrestrial environment in order to validate and/or improve tritium monitoring strategies for human foods and the environment.

Typically, the CNSC's licensee compliance programs for monitoring food focus on measuring HTO in garden produce or natural vegetation. OBT/HTO ratios are generally used to predict OBT activities in human food based on measured and modelled HTO levels in the environment. The OBT/HTO ratio describes the likelihood of tritium to accumulate in the organic fraction (Okada and Momoshima, 1993). The current environmental risk and public dose estimates models rely on the assumption that OBT forms directly from HTO and that under equilibrium conditions the OBT/HTO ratio should be the same in all environmental compartments. The OBT/HTO ratio is expected to be in the range of 0.6–0.8 near atmospheric tritium emission sources based on experimental data in the literature (CSA, 2008).

However, some recent studies at background locations in Ontario have reported that the OBT/HTO ratios in garden produce were considerably greater than expected (Thompson et al, 2011; OPG 2006, 2008). Further, OBT data are sparse and there is a lack of experimental data under conditions of chronic tritium exposure. Consequently, there are gaps in the current knowledge regarding the biological aspects of tritium uptake and incorporation in plants (Boyer et al., 2009).

The results presented in this study aim to address the gaps in the literature described above by expanding on the suite of measurements of HTO and OBT in environmental samples collected near nuclear facilities in Canada. This work expands on the measurements summarized in a report entitled *Tritium Activity in Garden Produce from Pembroke in 2007 and Dose to the Public*, which was also published by the CNSC (CNSC, 2010).

2. Study Description

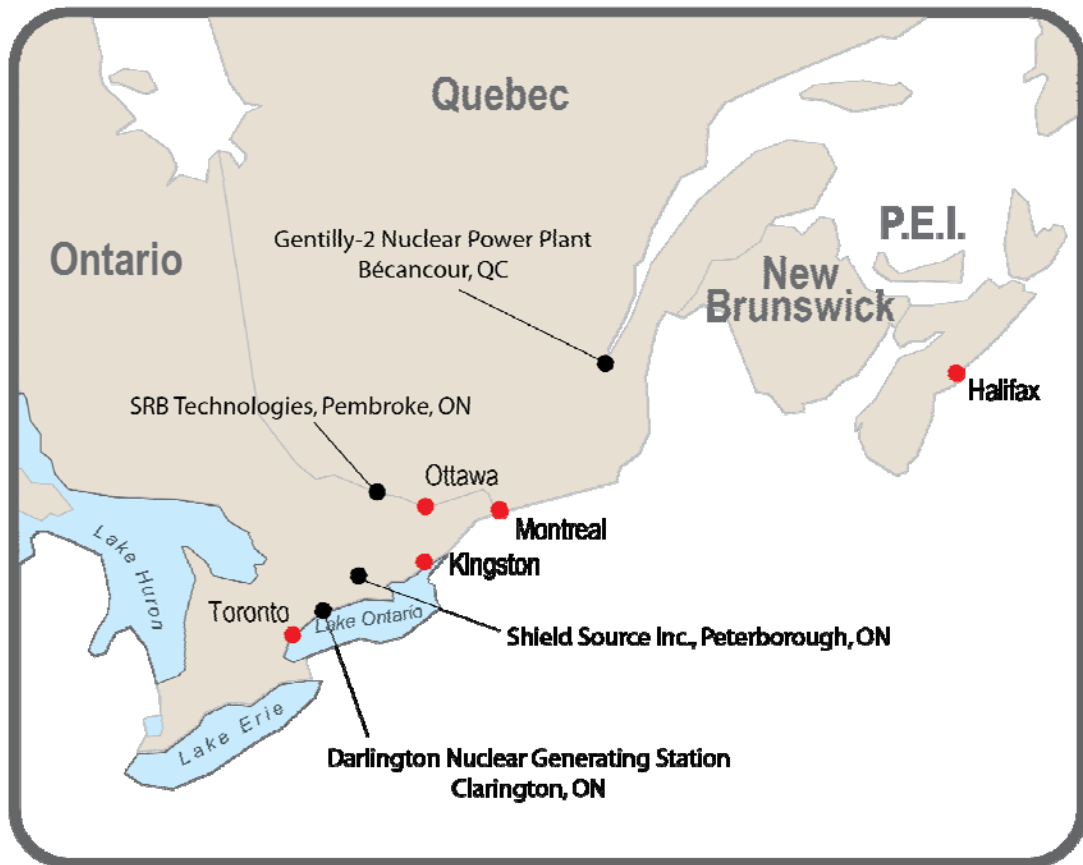
2.1 Study overview and objectives

The CNSC issued a contract to the University of Ottawa to measure the concentrations of tritium in diverse media in the terrestrial environment to determine how tritium releases enter the biosphere and the food supply. The study was conducted near four representative nuclear facilities which release tritium. Air, soil, garden produce, animal fodder and animal product samples were collected over two growing seasons (2008 and 2009) and were analyzed for various forms of tritium. The specific objectives of the study were to:

- obtain an integrated set of measured HTO and OBT concentrations in soil, garden produce, animal fodder and animal products measured near representative tritium-releasing facilities
- investigate the factors that influence the fate of tritium gas that is released to the atmosphere and deposited onto the ground surface
- assess the validity of determining the historical record of tritium releases by measuring OBT in the growth rings of representative long-lived trees
- document the recovery of the environment after a major reduction in HT and HTO releases from a tritium processing facility
- interpret the dynamics of OBT as it cycles between different environmental compartments under conditions of long-term, sustained releases of tritium from atmospheric sources

2.2 Study site description

Air, soil, garden produce, animal fodder and animal product samples were collected over two field growing seasons (2008 and 2009) near four Canadian nuclear facilities known to have a long history of tritium releases to the atmosphere (see figure 1). This section provides a brief description of the four nuclear facilities and two background locations included in this study. Table 1 summarizes the emissions from the four nuclear facilities between 2007 and 2010.

Figure 1 Locations of the four nuclear facilities included in this study

2.2.1 Nuclear facilities

Darlington Nuclear Generating Station

The Darlington Nuclear Generating Station (NGS) is a CANDU pressurized heavy water nuclear generating station that has been operating continuously since the early 1990s. This facility releases HTO from four CANDU reactors and HT from a tritium removal facility. The Darlington NGS is located in Clarington, Ontario, approximately 50 km southwest of Shield Source Incorporated in Peterborough, Ontario.

Shield Source Incorporated

Shield Source Incorporated (SSI) is a tritium processing facility located in Peterborough, Ontario. Emissions of tritiated hydrogen gas make up the majority of HTO emissions at SSI, versus emissions from the nuclear generating stations that are primarily HTO.

SRB Technologies

SRB Technologies (SRBT), located in Pembroke, Ontario, is another tritium processing facility. This facility, which was temporarily shut down in 2007, was included in this study because it provided an opportunity to investigate the response of the environment to the shutdown and restart of operations. Like SSI, SRBT releases more HT than HTO.

Gentilly-2 Nuclear Generating Station

The Gentilly-2 NGS, located in Bécancour, Quebec, has one CANDU power reactor that releases HTO. This facility was chosen to contrast the other facilities since it is far from the tritium releases from other nuclear facilities in southeastern Ontario.

Table 1 **Summary of HTO and HT emissions from nuclear facilities included in this study¹**

Year	HTO	HT
	(TBq)	(TBq)
Darlington		
2007	160.0	130.0
2008	160.0	83.0
2009	170.0	54.0
2010	140.0	21.0
SRB Technologies		
2007	5.8	36.1
2008	6.4	33.7
2009	14.3	26.3
2010	9.2	27.3
Gentilly-2		
2007	150.0	N/A
2008	170.0	N/A
2009	190.0	N/A
2010	170.0	N/A

2.2.2 Background locations

Two natural background areas served as control sites. The site in Russell, Ontario is located in the same province as several man-made sources of tritium. The other control sites are near the towns of Warman and Langenburg, Saskatchewan. The background sites in Saskatchewan are far from the influences of nuclear facilities in Ontario and any other man-made sources of tritium.

¹ Tritium (HT and HTO) emissions from the Shield Source Facility are not included in table 1 as the emission data is under regulatory review.

3. Experimental Methods

Sampling and analytical methods are described in detail in the report by the University of Ottawa (Clark et al., 2010). HTO and OBT concentrations were measured in soil, garden produce, animal fodder and animal product samples.

3.1 Environmental sampling

A sampling network involving approximately 10 vegetable gardens was selected over the 2008 and 2009 growing seasons at various distances at each of the four nuclear facilities. Sampling at each of the four sites provided a full range of materials including soils, garden produce, animal fodder (corn and grass), and animal products (meat, milk, eggs and honey). Efforts were made to choose sampling sites according to the prevailing winds. In addition, one or two gardens were sampled during both growing seasons (2008 and 2009) at each control site. All samples were stored in a refrigerator in their original containers prior to freezing for storage until analysis.

The vegetable gardens were selected at each site with increasing distance from the nuclear facilities. Seasonal vegetables were grown in gardens by local residents living near the nuclear facilities. The selection of vegetables depended on what was being cultivated in the gardens, with an emphasis on both above- and below-ground vegetables. Garden produce was also sampled at the two control sites. In Russell, Ontario, garden produce were collected from a local garden and additional vegetables were obtained from a market where local growers sell their produce. Locally grown fruits and vegetables were also obtained from the gardens near the towns of Warman and Langenburg in Saskatchewan.

Animal fodder (grass and corn) was sampled in farmers' fields in close proximity with the four nuclear facilities. Grass and corn were obtained from farms near SSI, Darlington NGS, SRBT, Gentilly-2 NGS and the two control sites in Russell, Ontario, as well as from Warman and Langenburg, Saskatchewan.

Honey was obtained from local producers living in close proximity to the four nuclear facilities, and the control site in Russell, Ontario. The sampling of dairy was challenging because of the restrictions on access to raw milk. Nonetheless, dairy farms located in close proximity to the nuclear facilities were identified and producers were queried on the feed supply for their cows. Farms where cows are fed locally grown fodder and grain were selected for milk samples.

3.2 Analytical methods

The following section provides a summary of the analytical methods used by the University of Ottawa to analyze HTO and OBT in environmental samples obtained for this project. Details of quality assurance and quality control measures are also described.

3.2.1 Extraction of HTO in environmental samples

The HTO measured in vegetation is often referred to as "tissue-free-water-tritium", which will be referred to as HTO in this report. The vegetation samples were ground to extract water free of plant tissue and to increase the surface area-to-volume ratio. Soil samples were used as received. Samples were then weighed to confirm original fresh weights.

Once the tissue-free water tritium was extracted from the sample, the remaining portion was dried to a constant weight. On average, approximately 65 percent of the remaining portion of the sample was dried. The final dried organic material was then re-saturated once or twice, as needed, to reach background with low-tritium water and to remove any remaining “exchangeable” tritium. The rinse water was periodically analyzed by liquid scintillation counting to ensure that the exchangeable tritium had been removed and to confirm that the tritium content was at background values. Samples were kept frozen until analysis. Losses during storage were negligible (< 1 percent as measured by differences between fresh weight and weight after retrieval from frozen storage).

3.2.2 Extraction of OBT in environmental samples

The specific activity of OBT can be measured in the dry matter that remains after the water free of plant tissue has been extracted. The dry sample was washed with tritium-free water to remove any remaining tritium. The measured activity represents activities of “non-exchangeable” tritium that is not removed by rinsing with tritium-free water and not necessarily bound to carbon.

The most widely used method for measuring OBT is conventional Parr bomb combustion of dry matter and subsequent liquid scintillation counting of OBT specific activity in the collected combustion water. The OBT levels in garden produce and animal fodder samples were analyzed by the Parr bomb combustion method. The low organic content in most soils precluded OBT analysis by conventional Parr bomb combustion; therefore, a separate line was used for OBT soil analysis. The separate line was based on the combustion stream of dry air with secondary catalytic conversion of volatile organic gases to water vapour. The captured water was weighed for yield, and then diluted with tritium-free glacier water and counted for tritium activity.

All samples and selected rinse waters were analyzed for tritium activity by liquid scintillation counting using a Perkin Elmer Quantulus 1220 low-background liquid scintillation counter. The average activity of three cycles was noted. A suite of standards was used for calibration.

A non-destructive method to measure OBT in samples is the ingrowth of the stable isotope of helium (^3He) from tritium decay (Faure and Mensing, 2005). The concentration of tritium in a sample, which has been encapsulated under vacuum, is determined from the amount of ^3He measured after a specified period of time according to the parent-daughter decay law for tritium. Tritium was estimated from the measured ^3He using the following formula:

$$T_o = {}^3\text{He}_{\text{excess}} / (1 - e^{-\lambda t}) \quad (1)$$

where: ${}^3\text{He}_{\text{excess}}$ is the accumulated ^3He from ingrowth; T_o represents tritium at the time of sample encapsulation; λ is the decay constant for tritium ($\ln(2)/T_{1/2} = 0.694/12.3 = 0.0563$); and t is the time of decay (period of encapsulation).

The helium ingrowth method was initially used to measure OBT in tree ring samples collected during this study, but was replaced by the conventional combustion method due to instrument failure. A small set of samples were analyzed by ^3He ingrowth. No samples were analyzed for OBT using both methods. The remaining samples were analyzed by combustion and liquid scintillation counting.

3.2.3 Corrections to tritium concentrations

Measured tritium concentrations were corrected to account for the difference between the sample collection date and the date of sample analysis. The following equation was used to calculate the tritium activities at the time of sample collection:

$$T_o = T / e^{-\lambda t} \quad (2)$$

where T_o is tritium activity at the time of sample collection; T is the measured activity (at the time of sample analysis); t is the time elapsed between sample collection and sample analysis; and λ is the decay constant for tritium.

3.2.4 Quality assurance and quality control

Intra- and inter-laboratory replicates were undertaken to assure appropriate quality control of analytical procedures. An inter-laboratory comparison was made with the Environmental Technologies Branch, Atomic Energy of Canada Limited's Chalk River Laboratories. Results demonstrate that replicates were within the limits of quantification.

The limit of detection and the limit of quantification for tritium measurements made by liquid scintillation counting are presented with analytical results in the appendices of the report by Clark et al (2010).

4. Environmental Fate of Tritium

Four experimental approaches were used to investigate the fate of tritium in soil and vegetation near nuclear facilities in Canada. The subsequent sections of the report are broken down as follows:

- HTO and OBT measurements were obtained at various distances from four nuclear facilities (see section 4.1). The results were compared to determine the effect of tritium releases on different environmental compartments.
- An experimental garden was set-up near SSI to investigate the HT–HTO–OBT conversion in garden produce (see section 4.2).
- Growth rings in tree samples were explored as records of tritium emissions near nuclear facilities (see section 4.3). The viability of this methodology for routine environmental monitoring was assessed.
- Measurements of HTO and OBT in environmental samples taken near SRBT over a three-year period (2007–09) were obtained to determine how the environment responds to abrupt changes in tritium emissions (section 4.4).

4.1 Partitioning of tritium in air, soil, vegetation and animal produce

An integrated set of measured concentrations of HTO and OBT in soil, garden produce, animal fodder and animal products obtained at close proximity to four nuclear facilities are presented below.

4.1.1 Background sites

In general, the HTO activities in soils, garden produce, animal fodder and animal products were between 0.73 and 4.6 Bq/L for the three control sites. The lowest value was observed in a crabapple sample in Warman, Saskatchewan, and the highest value was observed in corn leaves from Russell, Ontario. The HTO activity in garden produce is the most commonly monitored form of tritium in the environment. Ontario Power Generation (OPG) and Bruce Power (BP) have reported background HTO levels between 3 and 7 Bq/L for vegetables for five locations in Ontario in 2009: Lakefield, Bancroft, Sarnia, Barrie and Picton (OPG, 2010; BP, 2010). The background activities reported by OPG and BP are higher than background activities at the two Saskatchewan sites (0.73–2.6 Bq/L) and in agreement with the HTO activities in background vegetation in Russell, Ontario (1.2–4.6 Bq/L).

The OBT activities at all three background sites were higher than the HTO activities (see figures 2 and 3). This was unexpected because it was anticipated that OBT and HTO activities would be similar, based on the experimental data in the literature and current environmental risk assessment and public dose estimate models. The range in OBT concentrations in soils, garden produce, animal fodder and animal products was between 0.96 and 23.7 Bq/L. The highest OBT activities were observed in soil and tomato samples from the two Saskatchewan sites (19.9 and 23.7 Bq/L, respectively). Background OBT levels of 19.1 Bq/L in tomatoes from Lakefield, Ontario (OPG, 2003) and 7 Bq/L in carrots from Killaloe, Ontario (Kim and Davis, 2008) have been measured and are within the background range of this study. Higher OBT activities of 36.1 Bq/L and 65 Bq/L were reported for apple samples from Bancroft, Ontario, and bean samples from Kingston, Ontario, respectively (Kim and Davis, 2008).

The OBT/HTO ratio is assumed to be between 0.6 and 0.8 near atmospheric tritium emission sources, based on experimental data in the literature (CSA, 2008). In this study, the OBT/HTO ratios in all environmental samples from background locations were between 0.3 and 20.1, with the highest value observed in a tomato sample from Langenburg, Saskatchewan. OBT/HTO ratios measured for samples from the Saskatchewan site were higher (between 3.1 and 20) than the Ontario background site (0.29–5.0). The higher ratios observed at the background sites in Saskatchewan may be a result of lower HTO activities relative to Ontario. At this time, the exact mechanism responsible for this phenomenon is not well understood.

Figure 2 HTO and OBT activities in environmental samples from the background location at Russell, Ontario

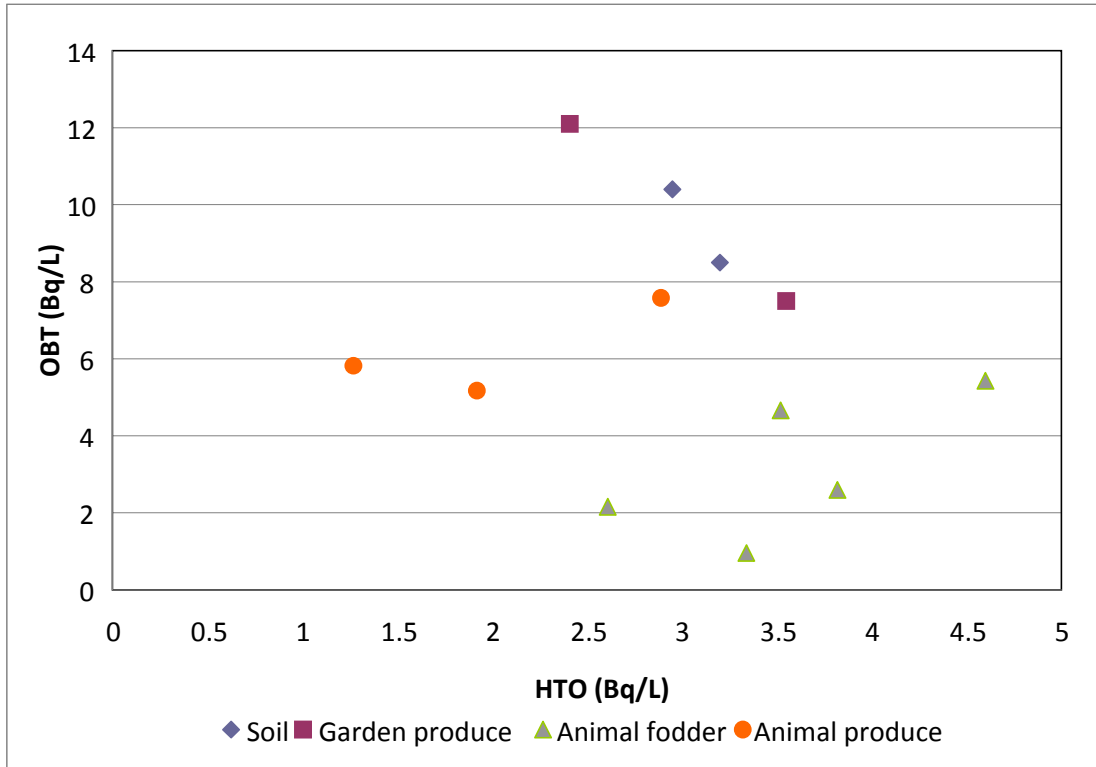
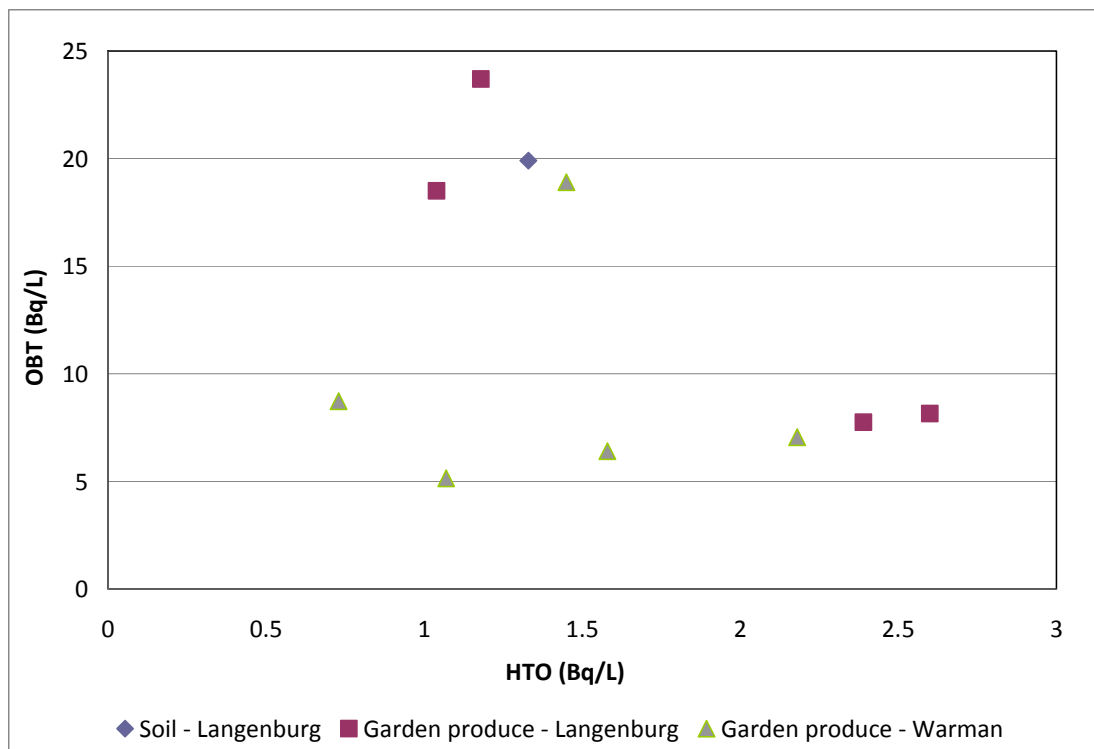


Figure 3 HTO and OBT activities in environmental samples from background locations near Warman and Langenburg, Saskatchewan



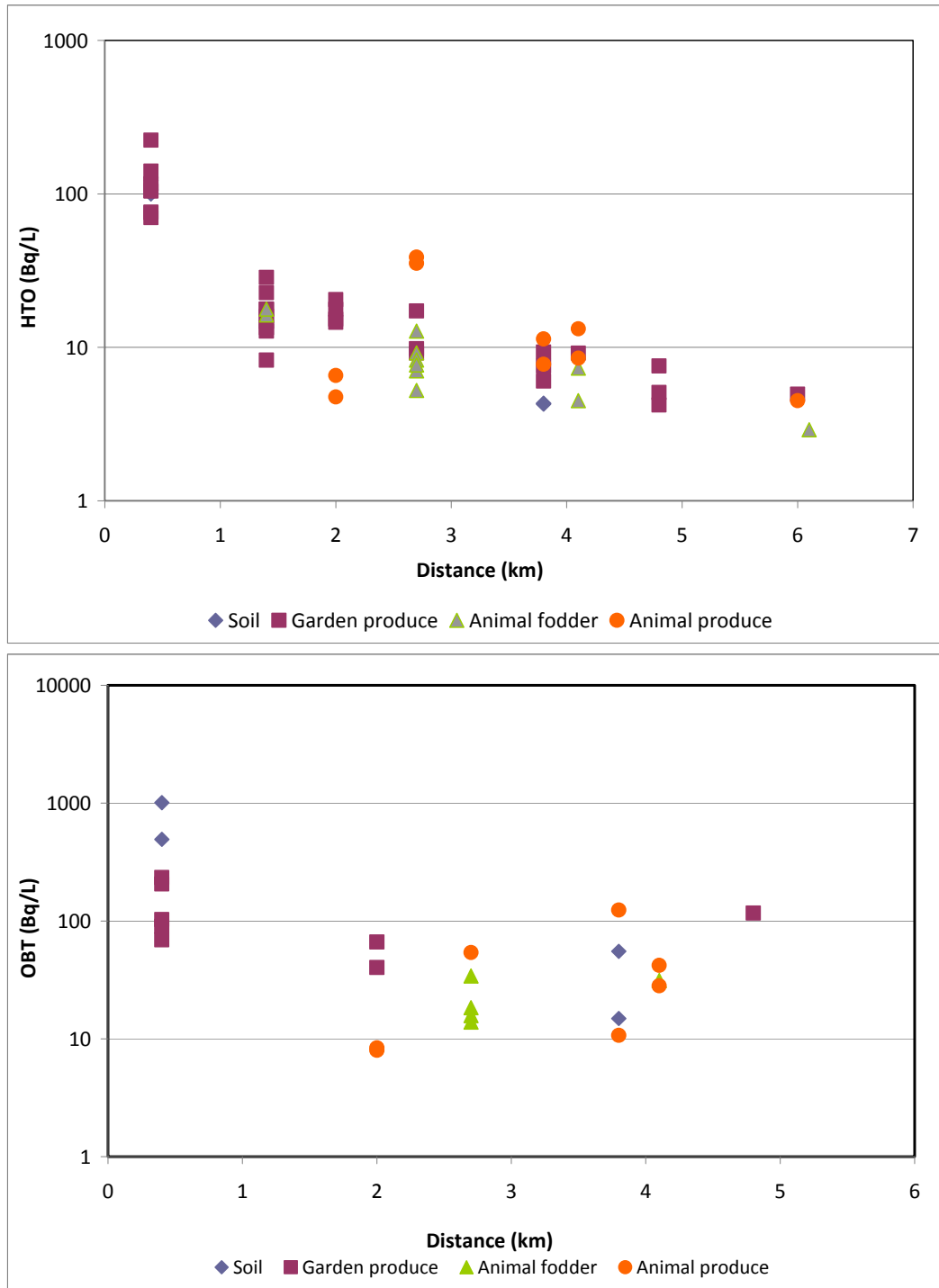
4.1.2 SRB Technologies

Soil, garden produce, animal fodder and animal product samples were collected from between 0.4 and 4.8 km from the facility. The HTO activities measured in all environmental compartments were between 224 Bq/L near the facility (0.87 km) to values less than 15 Bq/L at 2 km from the facility and beyond (see figure 4). Activities were observed to decrease with increasing distance from the facility's stack. HTO activities were highest in garden produce (4.21–222 Bq/L) and soil samples (4.31–122 Bq/L) and lowest in animal products (4.5–38.7 Bq/L) and animal fodder (2.9–17.7 Bq/L). In 2005, HTO activities of about 5000 Bq/L were measured in fruits and below-ground garden produce (e.g., root vegetables such as potatoes) (CNSC, 2010).

OBT activities in samples obtained near SRBT appear to decrease slightly with increasing distance from the facility (see figure 4). OBT activities in soil, animal fodder and garden produce samples range from 1010 Bq/L near the facility to 10.7 Bq/L at 4 km from the facility.

The mean OBT activities in soil and vegetation were 393 Bq/L and 115 Bq/L, respectively. OBT activities in soil samples were enriched over OBT activities in vegetation. Further, OBT activities in animal produce were higher than OBT activities in animal fodder, where average OBT levels in the two compartments were 39 and 23 Bq/L, respectively. The high OBT activities in animal produce compared to animal fodder most likely reflect differences in food supply. There is no evidence for preferential enrichment (i.e., biomagnification) through the food chain from soil to plants to animals.

Figure 4 SRB Technologies. Top: HTO activities for environmental samples collected at farms near SRBT, Pembroke, Ontario. Concentrations are plotted as a function of distance from the facility. Bottom: OBT concentrations from environmental samples as a function of distance from the facility



OBT/HTO ratios are expected to be close to 1, in accordance with current models, in which OBT is assumed to form directly from HTO under equilibrium conditions. The OBT/HTO ratios in environmental samples collected near SRBT were between 0.6 and 15.5. The maximum OBT/HTO ratio was observed in a cucumber sample obtained 4.8 km from the facility. The ratios observed in samples from SRBT are within the range of ratios observed at the background site. The enrichment of OBT compared to HTO in the environment near SRBT may be related to OBT remaining in the older soil reservoir following the decrease in emissions after the shutdown at SRBT in 2007.

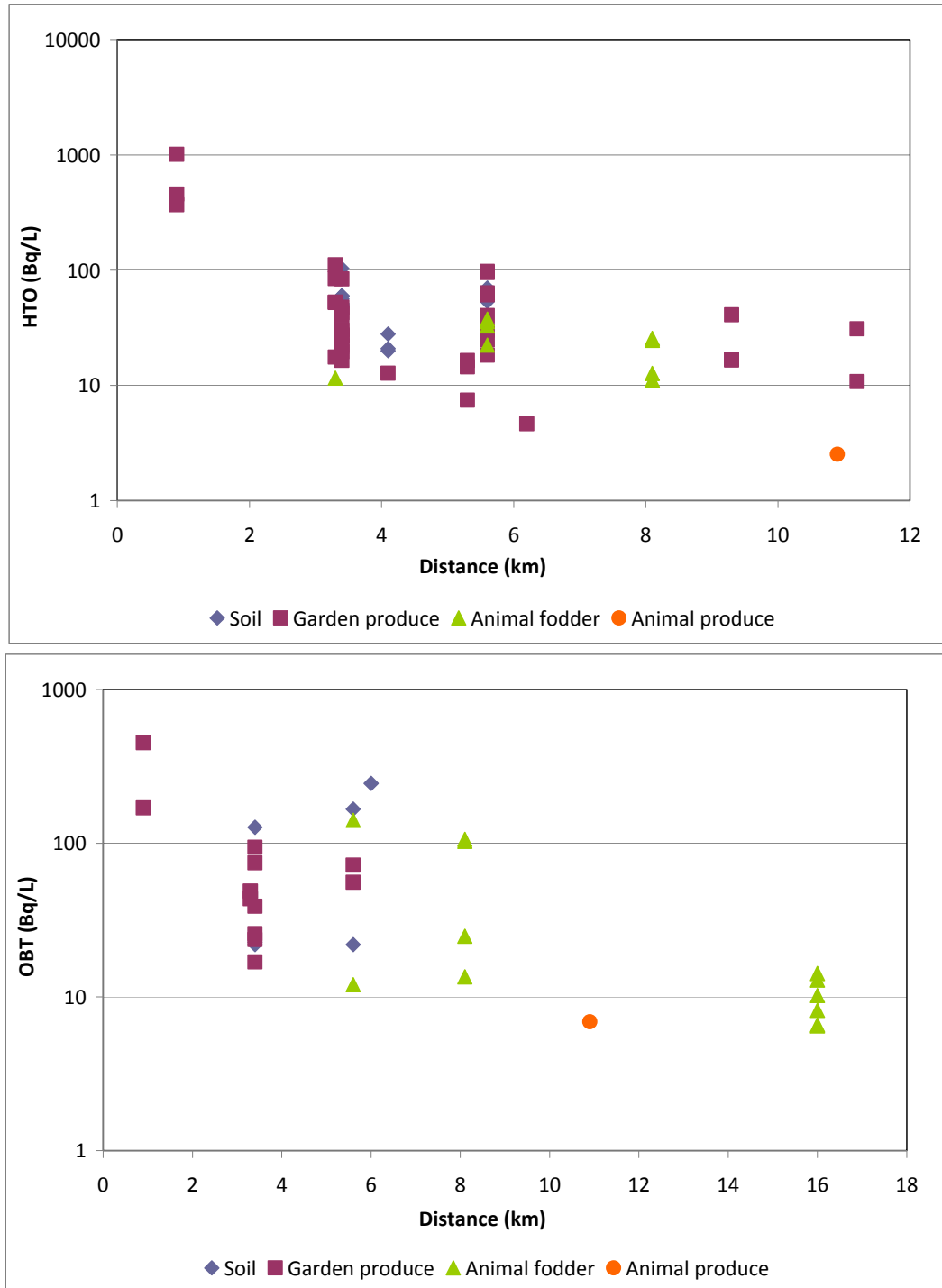
4.1.3 Shield Source Inc.

The HTO activities in soil, garden produce and animal fodder demonstrate a decrease to near background levels within 3 km of the facility. Activities range from 1000 Bq/L near the facility (0.8 km) to values less than 40 Bq/L, at more than 10 km from the facility (see figure 5). Soil, garden produce and animal fodder all show similar ranges in HTO activities. The maximum HTO activity was 1010 Bq/L for a potato sample collected 0.80 km from the facility. The decrease in OBT activity with distance was less pronounced than the trend observed for HTO samples collected near SSI (see figure 5). OBT activities varied between 451 Bq/L near the facility (0.79 km) to 6.5 Bq/L away from the facility (16 km).

The soil samples obtained near SSI showed a systematic increase in OBT activities from 2008 to 2009. Garden produce and animal fodder samples also displayed the same pattern of increased OBT between the two years. This increase is not observed in HTO samples. In general, HTO reflects current atmospheric conditions to within hours to days of sampling, whereas, OBT represents integrated values over a growing season. Analytical artifacts cannot explain the increases in OBT activities observed in the 2009 data. The increase in OBT may reflect a fluctuating emission source during the 2009 growing season. The absence of this trend in HTO activities may be due to the rapid exchange between environmental compartments and atmospheric HTO.

Overall, OBT/HTO ratios were between 0.4 and 4.8 for all environmental samples that were analyzed. The maximum OBT/HTO ratio was observed in a bean sample obtained 3.4 km from the facility. The increase in OBT between 2008 and 2009 was reflected in the OBT/HTO ratios, with higher ratios for the 2009 samples.

Figure 5 Shield Source Inc. Top: HTO concentrations from environmental samples collected at farms near Shield Source Inc, Peterborough, Ontario. Concentrations are plotted as a function of distance from the facility. Bottom: OBT concentrations from environmental samples as a function of distance from the facility



4.1.4 Darlington Nuclear Generating Station

Environmental samples were obtained between 2.6 and 6.3 km from the Darlington NGS. Many samples obtained near SRBT and SSI were already near background levels at 2 km from the facility. As a result, the observed effect of distance on tritium activities is weaker for the Darlington samples.

The HTO activities in soils, garden produce, animal fodder and animal products range from 25 Bq/L near the facility (2 km) to activities less than 12 Bq/L further away from the facility (6 km) (see figure 6). The animal fodder samples were all obtained at the same distance; approximately 4 km from the facility. The Darlington NGS measures tritium as part of its environmental compliance program. The measurements obtained in this study are within the range of HTO activities reported by OPG for Darlington NGS in the same year (OPG, 2010).

Similar to the measured HTO activities, no trend between OBT activity and distance was observed. One animal produce sample (milk) from 2008, obtained at a distance of 4 km from the facility, had an OBT concentration of 228 Bq/L (see figure 6). However, the following year, an animal sample from the same site had an OBT concentration of 11.9 Bq/L. The food supply may have affected OBT activities in animal produce. Food supply was not constrained or monitored during this study.

Many garden produce samples had comparable OBT and HTO activities, suggesting a steady-state loading of tritium to this reservoir from a steady-state source term. This is consistent with the stable HTO activities in vegetation and air samples that have been measured and reported by OPG at Darlington NGS since 1994. There was enrichment in OBT over HTO in the beef, milk and egg samples obtained in 2008. This enrichment was not observed in the 2009 samples. The higher OBT levels observed in the 2008 samples cannot be reconciled by fractionation or bioaccumulation of tritium during transformations in the food chain. Feed supply from high-tritium animal fodder cannot be discounted as a possible explanation for the excess OBT.

The OBT/HTO ratios in environmental samples varied, measuring between 0.2 and 45.7. The maximum ratio for animal produce was due to the large excess of OBT in a milk sample from 2008, as discussed above. If the maximum OBT/HTO ratio is excluded, the range in the environmental samples is between 0.2 and 13.0, similar to the range in OBT/HTO ratios observed at SRBT. The OBT/HTO ratios for soil, garden produce and animal fodder were between 0.18 and 2.61.

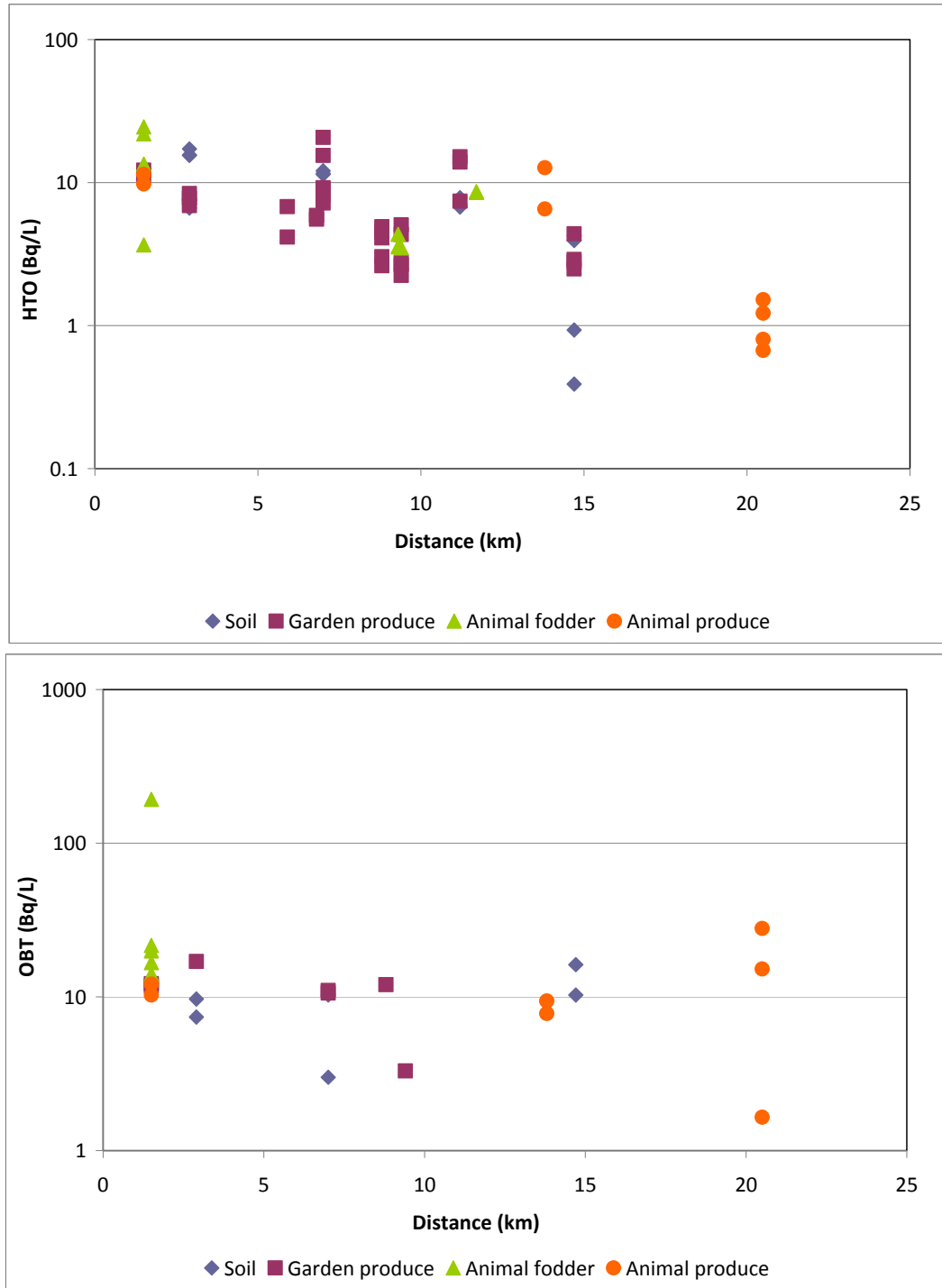
4.1.5 Gentilly-2 Nuclear Generating Station

The range in HTO concentrations at the distances sampled near Gentilly-2 was similar to the HTO measurements obtained near SRBT and Darlington NGS for similar distances from the facility. This similarity in the pattern of tritium concentration with distance suggests a similar mechanism of atmospheric dispersion of tritium near similar types of facilities.

The measured HTO activities in the different samples decreased with distance (see figure 7). There was no similar trend observed for OBT activities and distance. Environmental samples were obtained between 1.5 and 20.5 km from Gentilly-2 NGS. HTO activity values varied from 0.7 Bq/L to 24.6 Bq/L. OBT activities in the four environmental compartments were between 3 Bq/L and 193 Bq/L. There was no significant enrichment of OBT over HTO in any environmental reservoir, except for one anomalous animal produce sample (beef) from 2008. This sample was obtained 20.5 km from the facility and likely represents a background value.

OBT/HTO ratios were generally between 0.4 and 4.8 for samples in all environmental compartments. There were no significant year-to-year variations for OBT or HTO in any of the samples. The maximum OBT/HTO ratio at Gentilly-2 NGS was 41.6 from a beef sample obtained in 2008.

Figure 7 Gentilly-2 Nuclear Generating Station. **Top: HTO concentrations from environmental samples collected near the Gentilly-2 NGS. Concentrations are plotted as a function of distance from the facility. Bottom: OBT concentrations from environmental samples as a function of distance from the facility**



4.1.6 Summary

In general, HTO activities decreased with distance from a nuclear facility. This correlation was most pronounced at SRBT and SSI, where samples were obtained at closer distances to the facility and over a wider distance range. Samples could not be obtained at distances close to either the Gentilly-2 NGS or the Darlington NGS, because of an exclusion zone of 914 m around nuclear power plants. As a result, the observed effect of distance was less pronounced near nuclear power plants than near the processing facilities. The relationship between HTO and distance suggests that atmospheric tritium plays a role in HTO accumulation in soil and vegetation because tritium levels in air are also known to decrease rapidly with distance from the source of atmospheric tritium emissions (Ilin et al., 2005).

The relationship of OBT and distance was less pronounced at all nuclear facilities studied. At the nuclear processing facilities, there was a weak inverse correlation between measured OBT activity and distance. In contrast, there appeared to be no direct relationship between OBT activity and distance near nuclear power plants. Observations of higher than expected OBT values at background locations and at distances from facilities indicate that further investigations are required to determine the mechanism of OBT partitioning in the terrestrial environment.

Table 2 summarizes the average OBT/HTO ratios at the four nuclear facilities studied. There was no systematic enrichment of OBT over HTO at the two nuclear generating stations (Darlington NGS and Gentilly-2 NGS), except for some animal produce samples. The average OBT/HTO ratios from animal produce at Darlington NGS and the Gentilly-2 NGS were 11.1 and 8.2, respectively (see table 2). The measured ratios were higher than expected based on current physiological models of HTO and OBT behaviour in plants and animals (CSA, 2008).

Table 2 Average OBT/HTO ratios from environmental samples taken near nuclear facilities

Environmental compartment	Average OBT/HTO ratios			
	SRB Technologies	SSI	Darlington NGS	Gentilly-2 NGS
Soil	6.5	1.4	1.2	1.6
Garden produce	3.6	1.4	1.3	1.3
Animal fodder	2.8	2.2	0.9	3.8
Animal produce	4.1	2.8	11.1	8.2
Overall	4.0	1.8	3.6	3.8

For samples obtained near SRBT, OBT is enriched over associated HTO in all environmental samples. The enrichment of OBT over HTO is likely a result of a high inventory of OBT that resides in the region; perhaps due to historical emissions. Environmental samples obtained near SSI, are slightly enriched in OBT over HTO. The average OBT/HTO ratios for samples collected near SSI are lower than the ratios obtained for SRBT.

OBT/HTO ratios measured in this study were greater than 1. These results are consistent with the results obtained as part of the 2007 environmental survey and other background sites in Ontario and Saskatchewan. The consumption of produce with OBT/HTO ratios greater than 1 should only

contribute a small fraction to the public dose, and therefore is not considered an additional risk (CNSC, 2010).

4.1.7 Dose to members of the public

In the work summarized in *Tritium Activity in Garden Produce from Pembroke in 2007 and Dose to the Public* (CNSC, 2010), the public dose for members of the public living near SRBT in Pembroke was estimated using measured tritium activity levels in the food products sampled as well as site specific information regarding sources of food and food consumption rates. The conclusion of this previous work was that the dose from consuming tritium in fruits and vegetables grown in Pembroke was well below the public dose limit of 1 mSv per year and below doses known to cause health effects.

The dose to members of the public from consuming food products with high OBT/HTO ratios was determined using the same approach as described in the 2007 study in Pembroke. The dose from consuming food products with an OBT/HTO ratio of 0.8 is approximately 0.0001 mSv per year. An OBT/HTO ratio of 0.8 is the expected value near atmospheric tritium emission sources based on experimental data in the literature (CSA, 2008). The dose from consuming cucumber with an OBT/HTO ratio of 15.4 results in a dose to public of 0.0005 mSv per year. The dose from consuming milk with an OBT/HTO ratio of 45.7 results in a dose to public of 0.005 mSv per year. These values are well below the public dose limit of 1 mSv per year.

The dose to public from the consumption of produce with OBT/HTO ratios greater than 10 represents a small fraction of the annual public dose limit and is orders of magnitude below doses known to cause health effects. It should be noted that the total dose near a facility or background location would be greater than this because there are also contributions from other sources (e.g., consumption of other food products, inhalation) and from other radionuclides (e.g., uranium, radium and polonium).

The average effective dose from natural background radiation in Canada is 1.8 mSv per year. The dose from tritium around the four nuclear facilities in this report represent a small fraction of natural background radiation.

4.2 HT–HTO–OBT conversion

The conversion of atmospheric HT to HTO is known to occur predominantly in soil where microorganisms are available and capable of oxidizing hydrogen to water. Experiments were carried out to investigate the mechanism and factors that influence the fate of HT released to the atmosphere and the rate of conversion of HTO to OBT in soils. In 2009, an experimental garden was set up at SSI to examine HT–HTO–OBT conversion in tomatoes, cucumbers, radishes and beans.

4.2.1 Experimental garden

An experimental garden was established in a fully enclosed modular greenhouse located 25 m from the SSI stack. Three types of soil were used including: mushroom compost; standard potting soil, consisting of a mixture of peat, compost manure and black loam; and organic-matter-rich potting soil. All three soils were characterized as organic-rich silty sand. Low tritium water from the Otonabee River, the source of municipal water at SSI, was used to water the plants.

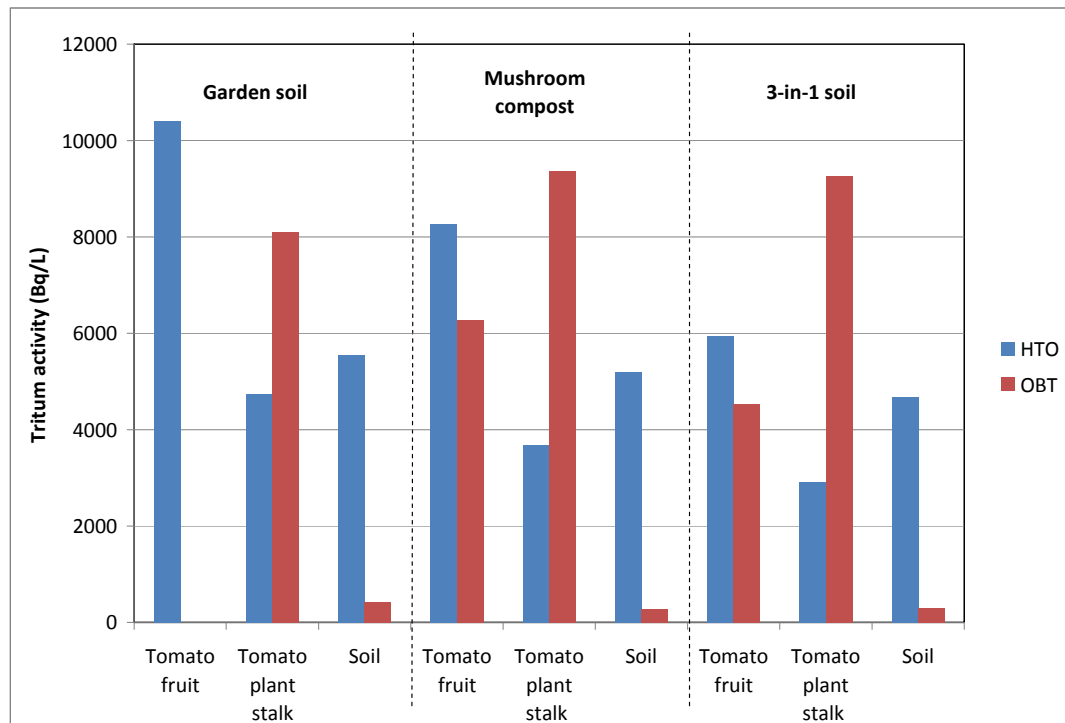
Tomatoes, cucumbers, radishes and beans were grown in pots. The type of tomato chosen for the experiment was obtained from local gardeners and is resistant to common local diseases.

Cucumber was chosen because it is a high-water-content green vegetable. Royal purple beans (*vicia faba*) are a high source of protein for humans and a common vegetable in regional gardens.

Of the four vegetable types grown in the three soils, only the tomato plant produced enough sample for analysis. The soil failed to support the other plant types well enough to produce vegetables. As a result only a small amount of plant material was produced for HTO analysis.

The tomato plant was analyzed in three parts: the tomato fruit, the tomato plant stalk, and soil (see figure 8). The tomato fruit had the highest HTO concentration, for all three soil types. The plant stalk had the lowest HTO concentration. In contrast, OB T was highest in plant stalks and lowest in soil. Furthermore, OB T was enriched in the tomato plant stalks over the corresponding fruit. HTO levels are greater than OB T activities in the tomato fruit, whereas OB T was greater in the tomato plant stalk.

Figure 8 HTO and OB T activities in tomatoes grown in three different types of soil in a greenhouse located 25 m from the SSI facility



4.2.2 Summary

The possible impact of soil type on tritium uptake was examined by growing the same plant types in different commercial soil types. These results provide no indication that soil type influences the partitioning of atmospheric tritium into OB T. The experimental approach did not provide insights into whether OB T was derived from atmospheric HTO or from HT converted to HTO in soils.

4.3 Tree rings as records of atmospheric tritium

Evidence of historical tritium releases from nuclear facilities may be recovered from the growth rings of trees (Love et al., 2002; Kozak et al., 1993). The validity of OB T measurements in tree

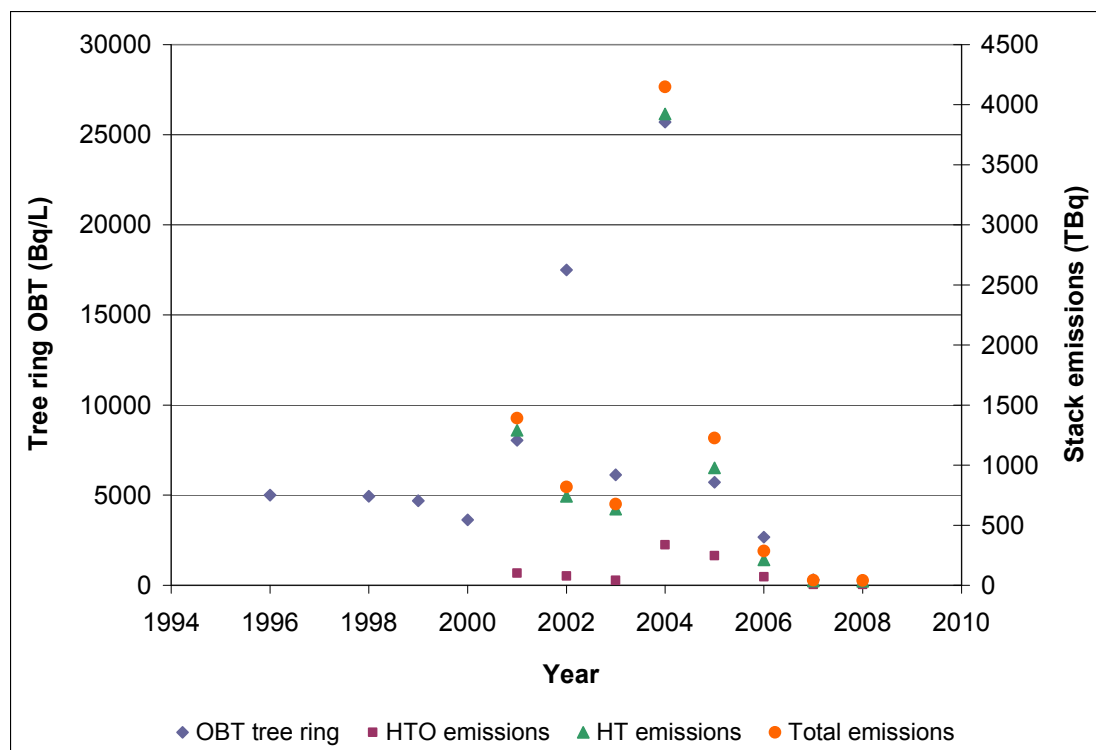
rings as an indication of historical releases of tritium (HT and HTO) was assessed by taking tree sections acquired near SRBT and the Darlington NGS.

4.3.1 SRB Technologies

A section from the trunk of a recently fallen aspen tree with a 14-year growth period was collected approximately 20 m southwest of the SRBT stacks in the general area of recent tritium in groundwater investigations.

Emissions from SRBT from 2001 to 2008 are presented in figure 9 along with measured tree ring data over the same period. The OBT activity in tree rings peaked at 27 500 Bq/L in 2004. The emissions data was obtained from SRBT's annual compliance reports. No emission records are available before 2000. Year-to-year variability was observed in the OBT tree ring data. The tree ring data was partially consistent with the emissions trend at SRBT.

Figure 9 OBT activities in tree rings from a poplar tree located 20 m from SRB Technologies

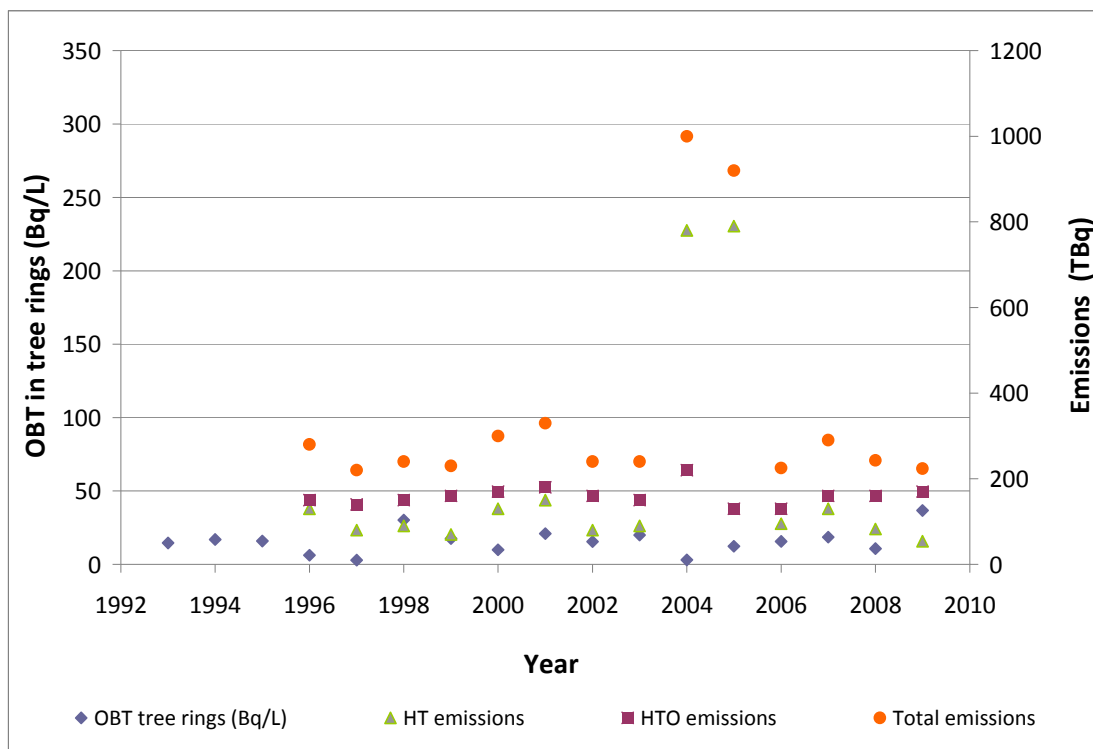


4.3.2 Darlington Nuclear Generating Station

A section of a poplar tree was obtained in November 2009 from a fallen tree located 1,760 m northwest of the Darlington NGS. Chain saw cuttings remaining on the stump indicate that the tree had recently been cut and therefore the outermost ring was taken to represent the 2009 growth season. The growth rings counted back to the year 1987.

The OBT levels in tree rings are plotted in figure 10 along with HT and HTO emissions from the Darlington site. The OBT tree ring activities for the tree sampled near the Darlington site were much lower than the activities of tree ring OBT for samples near SRBT. The average OBT measured in the tree ring samples near Darlington was approximately 15 Bq/L.

Figure 10 OBТ activities in tree rings from a poplar tree located 1760 m from the Darlington site



The Darlington site emissions were not reflected in the tree ring record. Furthermore, the two peak emission years (2004 and 2005) attributable only to higher-than-normal HT emissions are not reflected in the tree ring OBТ.

4.3.3 Summary

The tree ring records from SRBT captured the major multi-year trends of the associated HT-dominated emissions records reasonably well. The tree ring records recovered from near the Darlington NGS showed a poor correlation with emission trends.

The OBТ values in the tree record at SRBT (see figure 9) were an order of magnitude greater than OBТ levels measured in the tree ring of the sample collected near the Darlington NGS (see figure 10). The tree samples recovered at SRBT were closer to the stack than the tree sample collected near Darlington. Tritium emissions are known to decrease with distance approaching background within a few kilometers. The poor correlation near the Darlington NGS may have been due to the lower atmospheric tritium concentrations that occur further from the facility.

Measurements of OBТ activity from tree ring samples may be a useful indicator of historical emissions in certain contexts. For example, tree samples obtained at closer distances to the emission source better reflected the emission record than samples obtained further away.

4.4 Environmental response to changes in emissions

SRBT has been processing tritium since the early 1990s. The facility was temporarily shut down from January 31, 2007 to July 1, 2008 by the CNSC. SRBT was included in this study because

the temporary 2007–08 shutdown allowed an investigation of how the environment responded to the shutdown and restart of operations. The emission record for SRBT is summarized in figure 9. Since 2007, emissions have remained constant at approximately 40 TBq per year (see table 3) but the relative contribution of HTO to the total increased in 2009.

Table 3 Tritium emissions from SRBT between 2007 and 2009

Year	HTO (TBq)	HT (TBq)	Total tritium (TBq)	HTO/Total tritium (%)
2007	5.8	36.0	41.8	13.9
2008	6.4	33.6	40.1	16.0
2009	14.3	26.2	40.5	35.3

4.4.1 Environmental recovery at SRB Technologies

Figure 11 compares HTO and OBT concentrations in various garden produce (tomatoes, apples and potatoes) grown near SRBT for 2007, 2008 and 2009. For all three produce types, OBT concentrations decreased over the three consecutive years, with maximum values observed in 2007. Table 4 summarizes the OBT/HTO ratios for garden produce collected near SRBT between 2007 and 2009.

Figure 11 HTO and OBT activities in garden produce collected near SRB Technologies between 2007 and 2009

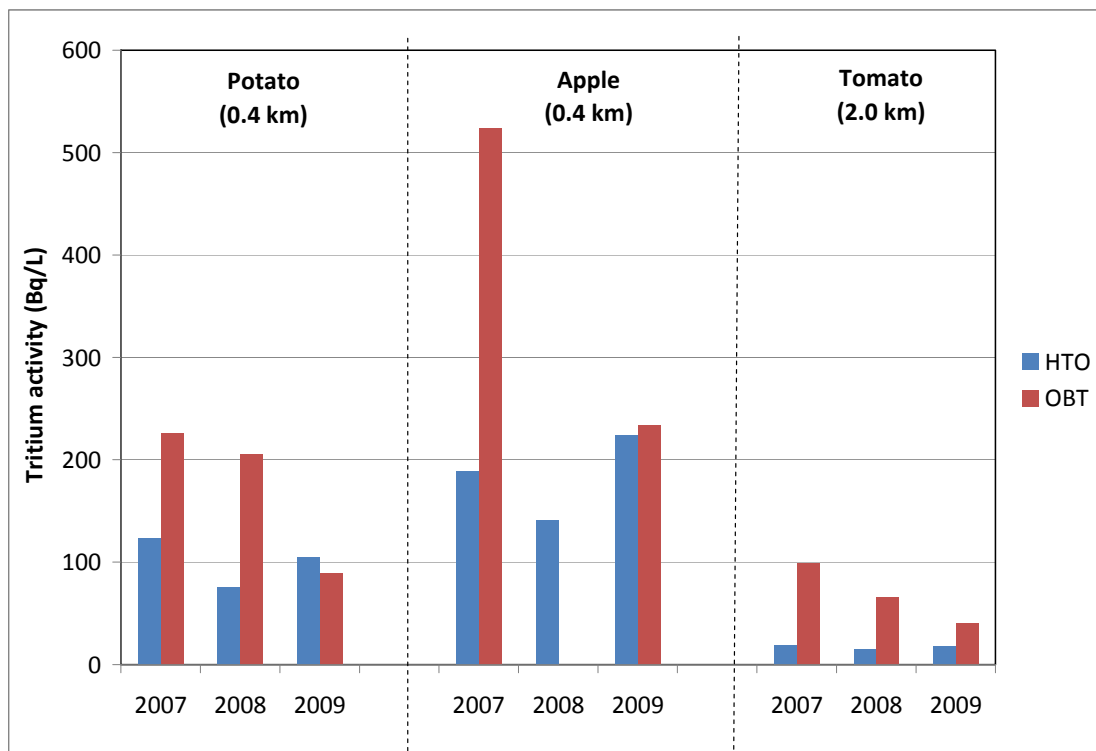


Table 4 OBT/HTO Ratios in garden produce collected near SRBT between 2007 and 2009

Produce (Distance from stack)	Year		
	2007	2008	2009
Potato (0.4 km)	1.8	2.7	0.9
Apple (0.4 km)	2.7	--	1.0
Tomato (2.0 km)	5.1	4.3	2.3

The decrease in OBT levels during this three-year period is inconsistent with the relatively constant total tritium emissions released from the facility during the three-year period (see table 3). HT, due to its low solubility, is not absorbed by vegetation. Therefore, the decrease in HT emissions (see table 3) cannot explain the decrease in plant OBT levels. The decreasing trend in plant OBT may be due to a decrease in soil OBT, which was not measured as part of this investigation.

4.4.2 Summary

The OBT activities in vegetation collected between 2007 and 2009 reflected changes in tritium emissions from SRBT. OBT reflects the tritium levels over a growing season; therefore, OBT activities from vegetation may be representative of the tritium levels for a given growing season. In this study, the HTO activities in vegetation over the same three-year period did not vary as much as the OBT activity in vegetation. Based on this study, OBT activities in vegetation may respond to changes in emissions. However, the exact mechanism of the formation of OBT from HT or HTO is still not well understood.

5. Overall Summary

Main findings

The CNSC issued a contract to the University of Ottawa to investigate the fate of tritium in the terrestrial environment. The objective of this research was to gain insight into how tritium releases enter the terrestrial environment and the human food supply. The results presented in this work are based on an integrated set of measured HTO and OBT activities in soil, garden produce, animal fodder and animal product samples measured near facilities that release tritium.

The findings of this research are summarized as follows:

- In general, HTO and OBT levels decrease with increasing distance from a facility. This trend was most pronounced in environmental samples obtained near nuclear processing facilities and less pronounced near nuclear power plants.
- The results of this study do not indicate that soil type influences the partitioning of atmospheric tritium.
- The experimental approach did not provide insight into whether OBT was derived from atmospheric HTO or from HT converted to HTO in soils.
- The OBT activity in tree rings may be a useful indicator of historical emissions under certain circumstances (i.e., at close distances to nuclear facilities).
- There was a large variation in the OBT/HTO ratio for soils, vegetation, animal products and animal fodder.
- OBT/HTO ratios measured in environmental samples were generally greater than 1 near the various nuclear facilities.
- The dose from consuming produce with OBT/HTO ratios greater than 1 represents a small fraction of the annual public dose and is orders of magnitude (between 250 and 2000 times) below doses known to cause health effects.

Further research

The data presented in this report provide additional information regarding the distribution of tritium in the terrestrial environment. However, questions remain regarding tritium activities in different environmental compartments and the rate and mechanism of OBT formation in soils. To this end, the CNSC has contracted the University of Ottawa to conduct an additional research project to help better understand the mechanisms of how tritium emissions are transported and transformed through the atmosphere to the terrestrial environment. The objectives of this follow-up research project include:

- methods for measuring different forms of tritium in air
- the evolution of chemical forms of tritium in air

- the transfer of tritium from air to the human food chain
- the kinetics of the formation of OBT

The knowledge gained from this additional research will be used to improve predictions and monitoring of tritium in the environment in order to better regulate nuclear facilities.

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7. Acronyms

BP	Bruce Power
CANDU	CANada Deuterium Uranium
CNSC	Canadian Nuclear Safety Commission
HT	tritiated hydrogen gas
HTO	tritiated water
NGS	nuclear generating station
OBT	organically bound tritium
OPG	Ontario Power Generation
SRBT	SRB Technologies
SSI	Shield Source Incorporated

Appendix A: Laboratory Results Obtained by the University of Ottawa

Background location: Russell, Ontario

			HTO	HTO	OBT	OBT	OBT	
Sample ID (year-location)	Type of sample	Water content	Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	OBT/HTO ratio
Soils								
Rus-08-01	Soil 2008	0.21	2.95	0.73	10.4	0.783	0.31	3.53
Rus-09-01	Soil 2009	0.25	3.20	0.81	8.5	0.533	0.38	2.66
Garden produce								
Rus-08-01	Carrot 2008	0.88	3.55	0.74	7.5	2.63	1.74	2.11
Rus-09-01	Carrot 2009	0.90	2.41	0.79	12.1	3.52	0.059	5.02
Animal fodder								
Rus-09-01	Grass 2009	0.79	3.52	0.82	4.66	1.70	0.023	1.32
Rus-08-02	Corn leaves 2008	0.77	2.61	0.77	2.16	0.80	0.022	0.83
Rus-09-02	Corn leaves 2009	0.77	4.60	0.84	5.43	2.23	1.73	1.18
Rus-08-02	Corn cob + kernels 2008	0.72	3.34	0.79	0.96	0.0483	1.58	0.29
Rus-09-02	Corn cob + kernels 2009	0.75	3.82	0.83	2.60	1.31	1.01	0.68
Animal products								
Rus-08-03	Milk 2008	0.86	1.27	0.73	5.82	3.77	2.08	4.58
Rus-09-03	Milk 2009	0.81	1.92	0.78	5.17	3.09	1.35	2.69
Rus-08-05	Honey 2008	0.10	2.89	0.81	7.58	4.06	1.87	2.62

Background location: Langenburg and Warman, Saskatchewan

			HTO	HTO	OBT	OBT	OBT	
Sample ID (year-location)	Type of sample	Water content	Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	OBT/HTO ratio
Soils								
2009-Langenburg	Soil 2009	0.17	1.33	0.89	19.9	0.59	0.007	15.0
Garden produce								
2008-Langenburg #1	Tomato 2008	0.92	1.18	0.74	23.7	5.04	1.52	20.0
2009-Langenburg #1	Tomato 2009	0.91	2.60	0.95	8.15	3.72	1.74	3.1
2008-Langenburg #3	Cucumber 2008	0.97	1.04	0.73	18.5	4.29	2.73	17.8
2009-Langenburg	Squash 2009	0.90	2.39	0.93	7.75	3.59	1.45	3.2
2008-Warman #4	Tomato 2008	0.93	1.45	0.75	18.9	6.2	2.17	13.0
2008-Warman Park #5	Crabapple 2008	0.82	0.73	0.72	8.73	3.61	1.30	12.0
2009-Warman	Apple 2009	0.85	1.58	0.91	6.40	3.15	1.26	4.0
2009-Warman	Apple Duplicate 2009	0.86	1.07	0.90	5.14	2.39	1.15	4.8
2009-Warman	Berries 2009	0.80	2.18	0.91	7.05	3.27	1.46	3.23

SRB Technologies

				HTO	HTO	OBT	OBT	OBT	
Sample ID (year-location)	Type of sample	Water content	Distance from facility (km)	Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	OBT/HTO ratio
Soils									
Pem-08-01	Soil 2008-1	0.19	0.40	102	2.26	1010	53.1	1.24	9.9
Pem-09-01	Soil 2009-1	0.22	0.40	122	2.54	493	8.09	0.33	4.0
Pem-08-12	Soil 2008-2	0.24	3.80	4.30	0.83	14.9	1.01	0.38	3.5
Pem-09-12	Soil 2009-2	0.24	3.80	6.30	0.86	55.2	2.64	0.51	8.8
Garden produce									
Pem-09-01	Apple 2009	0.87	0.40	224	3.69	234	96.1	3.11	1.0
Pem-08-01	Carrot 2008	0.91	0.40	75.5	1.88	69.1	22.1	3.12	0.9
Pem-09-01	Carrot 2009	0.90	0.40	116	2.49	103	25.4	2.39	0.9
Pem-08-01	Potato 2008	0.77	0.40	76.0	1.89	206	81.8	1.73	2.7
Pem-09-01	Potato 2009	0.75	0.40	105	2.32	90.0	42.9	2.22	0.9
Pem-08-05	Tomato 2008	0.94	2.00	15.3	1.00	66.3	7.87	6.63	4.3
Pem-09-05	Tomato 2009	0.94	2.00	17.7	1.09	40.3	10.4	3.39	2.3
Pem-09-09	Cucumber 2009	0.95	4.80	7.60	0.89	117	7.94	6.33	15.4
Animal fodder									
Pem-09-10	Grass 2009	0.75	4.10	7.3	0.88	31.3	13.0	9.83	4.3
Pem-08-13	Corn leaves 2008	0.77	2.70	7.0	0.84	34.1	7.71	2.40	4.9
Pem-09-13	Corn leaves 2009	0.79	2.70	9.2	1.15	13.9	5.19	1.91	1.5
Pem-08-13	Corn cob 2008	0.77	2.70	8.3	0.91	15.8	7.19	1.86	1.9
Pem-09-13	Corn cob 2009	0.85	2.70	12.7	1.23	18.4	8.71	2.11	1.4
Animal products									
Pem-08-06	Honey 2008	0.13	2.00	4.8	1.0	8.4	4.63	3.19	1.8
Pem-09-06	Honey 2009	0.13	2.00	6.6	1.2	8.0	3.38	0.68	1.2

				HTO	HTO	OBT	OBT	OBT	
Sample ID (year-location)	Type of sample	Water content	Distance from facility (km)	Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	OBT/HTO ratio
Pem-08-10	Egg 2008	0.70	4.10	8.5	0.87	42.0	19.4	0.85	4.9
Pem-09-10	Egg 2009	0.71	4.10	13.2	1.01	28.2	11.7	9.48	2.1
Pem-08-12	Milk 2008	0.88	3.80	7.8	0.86	124	78.8	2.17	15.9
Pem-09-12	Milk 2009	0.83	3.80	11.4	0.97	10.7	4.92	1.25	0.9
Pem-08-13	Ground beef 2008	0.67	2.70	35.3	1.31	54.1	34.3	1.08	1.5

Shield Source Inc.

				HTO	HTO	OBT	OBT	OBT	
Sample ID (year-location)	Type of sample	Water content	Distance from facility (km)	Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	OBT/HTO ratio
Soils									
Pt-08-01	Soil 2008-1	0.19	3.4	49.7	3.84	21.9	0.69	0.29	0.44
Pt-09-01	Soil 2009-1	0.14	3.4	56.2	0.91	127	6.99	0.47	2.26
Pt-08-03	Soil 2008-2	0.14	3.4	60	0.92	22.7	0.38	0.02	0.38
Pt-09-03	Soil 2009-2	0.09	3.4	103	0.89	167	2.77	0.37	1.62
Pt-08-10	Soil 2008-3	0.25	5.6	58.3	1	21.9	1.25	0.36	0.38
Pt-09-10	Soil 2009-3	0.17	5.6	69.9	0.97	245	12.1	0.60	3.51
Garden produce									
Pt-08-10	Apples 2008	0.84	5.6	40.4	0.78	55.6	12.2	7.63	1.38
Pt-09-10	Apples 2009	0.86	5.6	24.8	0.77	72	37.1	3.16	2.90
Pt-08-01	Beans 2008	0.91	3.4	21.8	1.15	16.9	7.7	9.62	0.78
Pt-09-01	Beans 2009	0.91	3.4	19.6	1.39	94.1	40.4	2.62	4.80
Pt-08-01	Tomato 2008-1	0.93	3.4	26.9	1.43	23.6	8.4	2.85	0.88
Pt-09-01	Tomato 2009-1	0.93	3.4	41.3	1.81	74.6	30.9	3.13	1.81
Pt-08-03	Tomato 2008-2	0.95	3.4	27.4	3.45	25.8	9.5	5.28	0.94
Pt-09-03	Tomato 2009-2	0.96	3.4	30.8	1.62	38.9	11.9	5.93	1.26
Pt-08-05	Potato 2008	0.84	0.9	456	6.25	170	72.7	14.2	0.37
Pt-09-05	Potato 2009	0.79	0.9	1010	14.2	451	208	6.34	0.45
Pt-08-09	Carrot 2008-1	0.89	3.3	52.4	1.62	43.5	13	6.03	0.83
Pt-08-09	Carrot 2008-2	0.85	3.3	52.7	1.62	48.9	22.8	5.19	0.93
Animal fodder									
Pt-08-07	Corn leaves 2008-1	0.79	8.1	11.1	0.97	13.5	5.9	1.9	1.22
Pt-09-07	Corn leaves 2009-1	0.78	8.1	24.5	1.49	106	41.1	4.71	4.33

				HTO	HTO	OBT	OBT	OBT	
Sample ID (year-location)	Type of sample	Water content	Distance from facility (km)	Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	OBT/HTO ratio
Pt-08-07	Corn cob 2008-1	0.87	8.1	12.7	1.00	24.8	10.5	7.19	1.95
Pt-09-07	Corn cob 2009-1	0.60	8.1	25.6	1.52	103	51.1	3.57	4.02
Pt-08-14	Grass 2008-1	0.72	5.6	22.4	1.16	12	4.2	2.39	0.54
Pt-09-14	Grass 2009-1	0.71	5.6	37.5	1.72	141	52	3.17	3.76
TPD-08-04	Grass 2008-2	0.62	16.0	4.54	0.83	12.9	4.6	2.57	2.84
TPD-09-04	Grass 2009-2	0.73	16.0	6.16	0.87	14.2	4.67	3.12	2.31
TPD-08-04	Corn leaves 2008-2	0.78	16.0	6.67	0.87	8.17	3.44	2.2	1.22
TPD-09-04	Corn leaves 2009-2	0.78	16.0	4.69	1.46	6.57	2.67	2.24	1.40
TPD-08-04	Corn cob 2008-2	0.83	16.0	6.57	0.86	10.2	5.09	1.54	1.55
TPD-09-04	Corn cob 2009-2	0.7	16.0	5.22	0.86	6.47	3.49	1.26	1.24
Animal products									
Pt-08-06	Chicken breast 2008	0.75	10.9	2.5	0.76	6.9	4.3	3.94	2.76

Darlington Nuclear Generating Station

				HTO	HTO	OBT	OBT	OBT	
Sample ID (year-location)	Type of sample	Water content	Distance from the facility (km)	Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	OBT/HTO Ratio
Soils									
Dar-08-02	Soil 2008-1	0.16	2.60	18.40	1.11	28.90	0.52	0.30	1.57
Dar-09-02	Soil 2009-1	0.15	2.60	11.40	0.98	12.00	0.25	0.37	1.05
Dar-08-07	Soil 2008-2	0.20	5.20	9.80	0.96	1.80	0.08	0.35	0.18
Dar-09-07	Soil 2009-2	0.16	5.20	7.90	0.91	4.70	0.16	0.28	0.59
Dar-08-11	Soil 2008-3	0.17	5.70	16.30	1.08	26.80	0.88	0.45	1.64
Dar-09-11	Soil 2009-3	0.14	5.70	10.10	0.95	19.60	0.32	0.34	1.94
Garden produce									
Dar-08-02	Apple 2008	0.86	2.60	15.70	1.09	16.90	7.47	1.48	1.08
Dar-09-02	Apple 2009	0.86	2.60	14.10	1.03	16.00	7.29	1.20	1.13
Dar-08-06	Tomato 2008	0.95	6.10	10.10	0.95	26.40	6.92	3.51	2.61
Dar-09-06	Tomato 2009	0.93	6.10	5.90	0.86	6.80	2.65	1.20	1.15
Dar-09-07	Zucchini 2009	0.95	5.20	10.80	0.97	9.30	3.94	1.94	0.86
Dar-08-08	Potato 2008	0.76	4.10	18.20	1.10	20.90	9.83	2.08	1.15
Dar-09-08	Potato 2009	0.74	4.10	21.10	1.16	10.20	5.01	1.65	0.48
Dar-08-11	Carrot 2008	0.90	5.70	8.10	0.91	17.00	5.93	2.28	2.10
Dar-09-11	Carrot 2009	0.87	5.70	11.60	0.98	12.10	5.30	1.44	1.04
Animal fodder									
Dar-08-14	Corn leaves 2008	0.76	4.20	11.40	0.97	4.10	1.83	0.24	0.36
Dar-09-14	Corn leaves 2009	0.79	4.20	15.90	1.07	11.10	4.29	2.04	0.70

				HTO	HTO	OBT	OBT	OBT	
Sample ID (year-location)	Type of sample	Water content	Distance from the facility (km)	Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/kg)	Error (Bq/kg)	OBT/HTO Ratio
Dar-08-14	Corn cob 2008	0.80	4.20	10.70	0.97	16.80	7.36	1.82	1.57
Dar-09-14	Corn cob 2009	0.72	4.20	15.80	1.06	12.70	4.95	1.13	0.80
Animal products									
Dar-08-05	Honey 2008	0.10	6.30	22.10	1.20	26.50	11.20	3.66	1.20
Dar-08-01	Ground beef 2008	0.72	4.00	15.70	1.09	39.60	23.20	0.85	2.52
Dar-08-03	Milk 2008	0.89	4.20	5.00	0.89	228.00	137.00	2.76	45.60
Dar-09-03	Milk 2009	0.90	4.20	6.10	0.86	11.90	7.60	3.05	1.95
Dar-08-03	Egg 2008	0.70	4.20	3.10	0.85	40.30	16.80	0.83	13.00
Dar-09-03	Egg 2009	0.76	4.20	2.00	1.05	5.00	2.50	1.64	2.50

Gentilly-2 Nuclear Generating Station

				HTO	HTO	OBT	OBT	OBT	
Sample ID (year-location)	Type of sample	Water content	Distance from facility (km)	Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/Kg)	Error (Bq/kg)	OBT/HTO ratio
Soils									
Gen-08-02	Soil 2008-1	0.22	2.9	15.6	1.04	9.7	0.43	0.262	0.62
Gen-09-02	Soil 2009-1	0.16	2.9	6.6	1.08	7.4	0.24	0.27	1.12
Gen-08-06	Soil 2008-2	0.17	14.7	4.3	0.82	10.3	0.37	0.306	2.40
Gen-09-06	Soil 2009-2	0.13	14.7	3.9	1.41	16.2	0.45	0.271	4.15
Gen-08-17	Soil 2008-3	0.25	7	11.6	1	3	0.13	0.253	0.26
Gen-09-17	Soil 2009-3	0.23	7	8.8	0.97	10.3	0.42	0.301	1.17
Garden produce									
Gen-08-17	Apple 2008	0.84	7	20.7	1.14	10.6	4.69	1.42	0.51
Gen-09-17	Apple 2009	0.85	7	15.5	1.37	11	5.33	1.55	0.71
Gen-08-01	Tomato 2008	0.95	1.5	11.3	0.98	12.2	4.64	2.15	1.08
Gen-09-01	Tomato 2009	0.92	1.5	12.2	0.99	11.8	5.17	1.37	0.97
Gen-08-02	Carrot 2008	0.88	2.9	6.9	0.89	17	3.76	4.79	2.46
Gen-09-20	Beans 2009	0.92	8.8	4.5	0.83	12	2.68	3.23	2.67
Gen-09-19	Potato 2009	0.78	9.4	4.3	0.83	3.3	0.84	-0.159	0.77
Animal fodder									
Gen-08-01	Corn leaves 2008	0.77	1.5	10.6	0.97	14.1	5.36	2.16	1.33
Gen-09-01	Corn leaves 2009	0.74	1.5	21.9	1.17	21.6	7.33	2.09	0.99
Gen-08-01	Corn cob + kernels 2008	0.84	1.5	13.5	1.03	193	29.5	6.33	14.30
Gen-09-01	Corn cob + kernels 2009	0.85	1.5	24.6	1.22	19.9	10.1	1.29	0.81
Gen-09-01	Grass 2009	0.76	1.5	10	0.95	16.7	6.73	3.69	1.67

				HTO	HTO	OBT	OBT	OBT	
Sample ID (year-location)	Type of sample	Water content	Distance from facility (km)	Field tritium (Bq/L)	Error (Bq/L)	Field tritium (Bq/L)	Field tritium (Bq/Kg)	Error (Bq/kg)	OBT/HTO ratio
Animal products									
Gen-08-01	Milk 2008	0.85	1.5	11.4	0.99	12.1	6.36	1.89	1.06
Gen-09-01	Milk 2009	0.87	1.5	9.8	1	10.3	4.99	1.23	1.05
Gen-08-08	Ground beef 2008	0.73	20.5	0.7	0.75	27.9	14.6	0.662	39.86
Gen-09-08	Ground beef 2009-1	0.71	20.5	1.5	0.76	1.65	1.2	3.7	1.10
Gen-09-08	Ground pork 2009	0.76	20.5	1.22	0.75	15.2	7.58	3.5	12.46
Gen-08-09	Honey 2008	0.1	13.8	6.5	2.17	7.8	3.64	3.66	1.20
Gen-09-09	Honey 2009	0.1	13.8	12.7	1.22	9.39	4.42	3.15	0.74