EVALUATION OF LONG TERM ENVIRONMENTAL TRITIUM REEMISSIONS FOLLOWING REACTOR SHUT-DOWN

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Abstract

More than 60 years of heavy water-moderated reactor operations at Canadian Nuclear Laboratories' (CNL) Chalk River site, have led to low level, persistent airborne tritium releases and tritium contamination in near surface geophysical strata (atmosphere, vadose zone, shallow groundwater and vegetation). Decades ago, the long-term-average flux of tritium from the National Research Universal (NRU) reactor to the surrounding area approached a steady state marked by slightly but clearly elevated local tritium levels in all geophysical strata at the Chalk River site. This study considers these near-surface geophysical compartments as a future source of tritium releases (reemission) following planned NRU decommissioning in 2018. The reemission phase ecological half-life (or conversely transfer coefficient) has been derived from elevated tritium concentrations which persisted in plants collected at the experimental garden plot near the NRU stack at the Chalk River site during the 15 month-long NRU shutdown in 2009-2010.

1. Introduction

Planned NRU decommissioning in 2018 will include the prediction of the evolution of tritium concentration in the environment. This could be a challenging task, as environmental transfer of tritium on a landscape scale comprises many interrelated processes subject to large spatio-temporal variability and the influence of site-specific parameters. There is no simple model to predict tritiated water (HTO) recirculation on a landscape scale. On the other hand, there is neither significant sorption, nor accumulation (including bioaccumulation) known for tritium, and it is therefore possible to estimate reemission using a simple lumped parameter model based on ecological tritium half-life $\boldsymbol{T}_3$. The model is based on the traditional first-order kinetics equation:

$$d\boldsymbol{C}_{\mathrm{HTO}}/dt = \boldsymbol{C}_0 \, exp\,(-\boldsymbol{\lambda} t) \qquad (1)$$

where t is time (s), $\boldsymbol{C}_0$ is initial concentration (Bq/L), $\boldsymbol{\lambda} = 1/\boldsymbol{T}_3$, $\boldsymbol{\lambda}$ is a kinetic loss or transfer coefficient (1/s) and $\boldsymbol{C}_{\mathrm{HTO}}$ is the average activity concentration of HTO (Bq/L) persisting locally in the geophysical compartment of interest (e.g., in near-surface atmosphere, in the vegetation layer, etc.) at the time t.

This paper presents an evaluation of a landscape-scale ecological half-life for tritium ($\boldsymbol{T}_3$) that would be applicable for evaluation of consequences and post-closure assessments at CNL. The proposed approach based on tritium retention time is applicable to non-stationary conditions and provides an alternative to detailed modelling.

The current regulatory paradigm, pertaining to quasi-equilibrium associated with slowly varying routine releases of tritium, directly relates the annual average atmospheric HTO level to HTO concentration in plant water (tissue free water tritium (TFWT)). Subsequently, it uses an isotopic

discrimination factor of ~0.8 to calculate organically bound tritium (OBT) concentrations [1]; such relationships are assumed without any lag in time. Such direct (instantaneous) linking of atmospheric and plant tritium concentrations is called the Specific Activity (SA) paradigm.

The SA approach is not applicable for a transition post-closure period but unfortunately, during that time period, dynamical models developed for fast changing (accidental) atmospheric releases are not applicable either. This is because such models rely solely on fast transfer of tritium from the atmosphere and precipitations into (and out of) connected geophysical strata including the biosphere (vegetation layer with rhisosphere a.k.a. root zone), the whole unsaturated (vadose) zone down to watertable and the subsurface water in aquifer. The fast transfer could, however, be modulated using a lump parameter model based on ecological half-lives. Such an approach exists already and, in general, the ecological tritium half-life in vegetation and rhisoshpere comprises two components: a short one which ranges from a few hours to a few days (according to the water transfer cycle) and a long one which takes place over a few weeks, as per the assimilation and decomposition of OBT in mild climate known to have a ecological half life of ~40 days [2-5]. Mathematical models subsequently rely on two exponents: the fast (few hours/days, T1) and the slow one (a few weeks up to 40 days, T2):

$$\lambda = \lambda_1 + \lambda_2, \text{ where } \lambda_1 = 1/\ T_1 \text{ and } \lambda_2 = 1/\ T_2 \quad (2)$$

When the fast processes get extinct ($t > T_1$) the slow process takes over and governs the magnitude of tritium in the noted near surface geophysical strata. However, when the second (slow) process becomes extinct ($t > T_2$), the system does not reach quasi-equilibrium and, as such, still does not obey the parameterization corresponding to the SA paradigm. The problem stems from a generally overlooked third exponent, which encompasses the much slower processes which becomes dominant after a month or two. This third component takes into account the slow HTO reemission processes with an ecological half-life approaching, and even exceeding, one year [6-10]:

$$\lambda = \lambda_1 + \lambda_2 + \lambda_3, \text{ where } \lambda_3 = 1/\ T_3 \quad (3)$$

This half-life, T_3 remains to be quantified at Chalk River Laboratories (CRL).

The reason for such a long ecological half-life could be attributed to our climate where processes like one dimensional (vertical) transfer in the presence of snow cover or to the effects of sequential freeze-thaw of the upper soil are taking place [11]. Another reason could be linked to landscape scale lateral subsurface transport of HTO and near-surface lateral migration of evaporated (re-emitted) atmospheric HTO. This latter possibility is tested in this study.

The study uses OBT levels found in agricultural produce, grown at the CRL experimental garden plot, located near the Acid Rain Site (ARS) [12]. A large swamp is located approximately 500 m from the garden plot. This is important at the landscape scale as the wind approaching the garden plot from all sectors between WSW and NW directions comes from the swamp area. This represents about 25% of all possible wind fetches. However, as these sectors contain winds prevailing at CRL, the garden plot effectively receives water vapour from the swamp 50% of the time. The swamp acts as a secondary HTO source with insignificant atmospheric dilution. This means that near surface atmospheric HTO concentrations do not change much over the distance

between the swamp and the garden plot. During the 15 months of NRU reactor shut-down in 2009-2010, the swamp was likely the predominant source of re-emitted tritium reaching the garden plot.

During the reactor shut-down period, steps were taken to confirm that the observations were not consistent with a fast transfer of tritium from the atmosphere and precipitations (as current dynamic models suggest), but rather followed a long lag function consistent with the slow HTO reemission processes ($t > \boldsymbol{T}_2$). Practically, we verified whether there was a lag between the signatures for the concentrations of HTO in precipitations, in soil porewater and in the concentration of OBT present in the agricultural produce at the garden plot. When a lag was observed, we evaluated whether it was consistent with a fast or a long time function.

After the NRU repair, the calandria vessel was refilled with new heavy water. Due to this refill, the routine atmospheric HTO emissions at CRL were reduced by a factor of approximately six. The post-repair drop in atmospheric HTO emissions presents a distinct signal traceable in ecological samples, OBT in vegetation being a good example. HTO activity concentrations also dropped in the waters collected from nine wells located close to the source, in an area which has no other contributing sources of HTO, except for atmospheric deposition. This well system was, therefore, used to further evaluate the effects of atmospheric deposition and lateral subsurface transport of HTO.

2. Methodology

A site description is presented in [12, 13]. The sampling methodology and the analytical techniques used to measure atmospheric HTO, HTO in precipitations, soil porewater HTO, plant TFWT and OBT are presented in [12].

The location of the NRU building, stack, ARS (experimental garden plot), Ottawa River and shoreline drilled wells are depicted in Figure 1. The precipitation sampling point is located at the Balmer Bay gate of the CNL site (denoted BB gate in Figure 1) to the northwest of the ARS approximately 5.5 km from the stack and approximately 3 km from ARS. The latter implies that precipitation HTO concentration at ARS is proportional to that sampled at the Balmer Bay gate.

Lag times between the source (atmospheric HTO) and the sink (produce OBT) were determined using a coefficient of determination (R2). Following a release, the time elapsed before the R2 value reaches a maximum was taken as the best estimate lag time. The same approach was used for the alternative analysis of lag time between precipitation HTO concentrations and produce OBT. Although the atmospheric HTO was changing smoothly, there was significant variability in the monthly precipitation HTO concentrations. Precipitation HTO levels decreased with the increased fraction of time that the wind was blowing towards the stack (and was delivering essentially uncontaminated precipitation). Because of this, before analysis, the precipitation HTO values were substituted with a 6-month moving average even if this introduced an inherent additional time lag. Due to this difference in methodology and to complex difference between HTO incorporation in plant TFWT from atmospheric and groundwater sources, alternative analysis based on precipitation HTO provided an independent evaluation of lag.

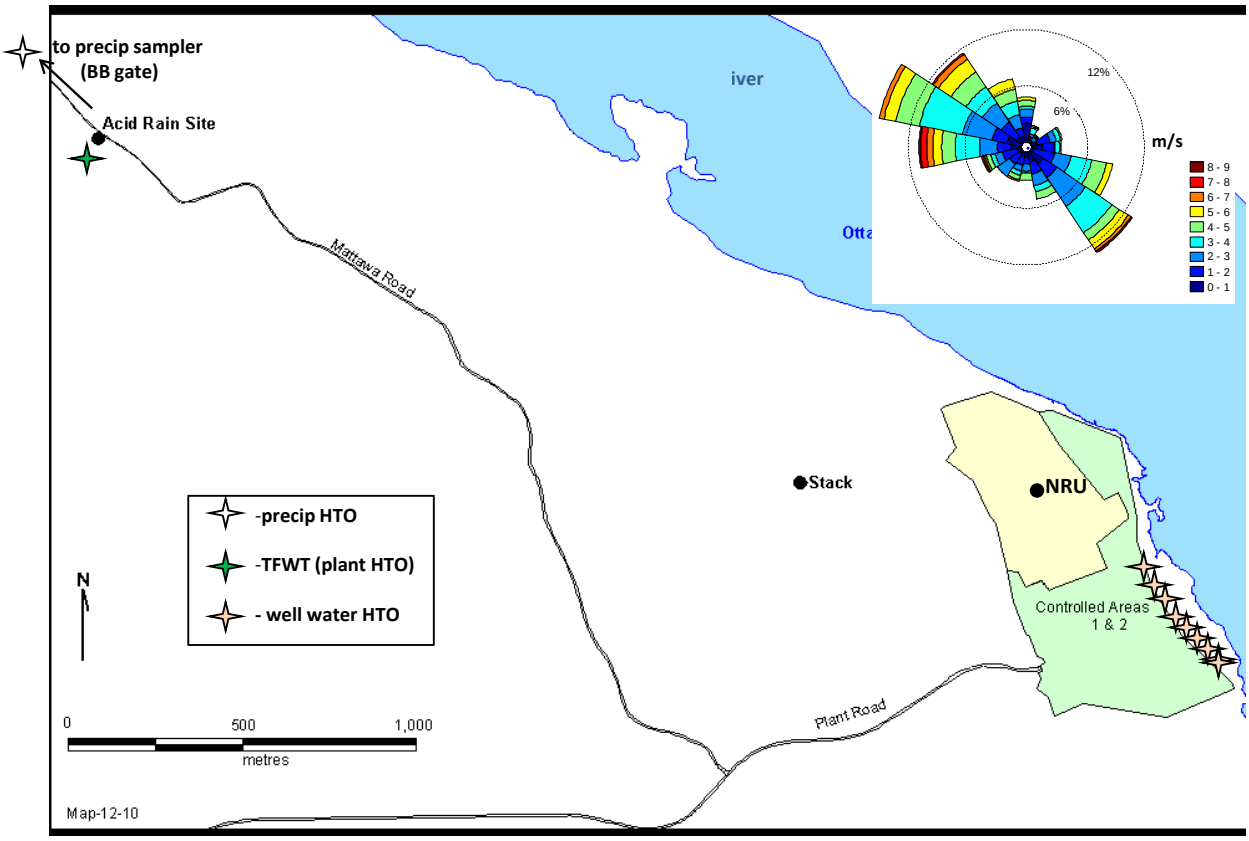

Figure 1. Location of NRU, stack, experimental garden plot at Acid Rain Site, Ottawa River and drilled wells at CNL and precipitation sampler. The wind rose measured at CNL tower at 60 m agl.

The approximately 6-fold drop in the magnitude of HTO atmospheric emission, due to the NRU calandria vessel repair in 2009, caused a significant drop in well water HTO within the same year. This implies that, at this location, a significant fraction of the well water is less than 1 year old. That being said, the slope of the water table near the NRU indicates that older well water is collecting in a nearby catchment location. Therefore, it is reasonable to assume that the HTO activity concentration in this older water is proportional to the atmospheric HTO concentration which existed at the well site 2 or more years prior. It can also be assumed that the levels were close to constant before the calandria vessel repair [14]. For the purposes of this study, we have assumed that the proportion of the old groundwater was minor in the water collected from the wells and we have neglected it. It is worth noting, that these assumptions simplify the analysis but are not critical for obtained results. We have analyzed wells 610-81 to 610-89 (Figures 1 and 2), which are in the vicinity of the Ottawa River shoreline and their hydro-geophysical characteristics can be found in [14]. They capture water moving approximately in a NE direction towards the shore of the Ottawa River, and most of their catchment is grass-covered with some impervious areas. The wells are drilled approximately to the same depth (~5 m to well screen) and through materials similar for all wells (as shown in Figure 2) and presumably for ARS and surrounding area. The wells were chosen to evaluate the travel time of atmospheric HTO. In these wells, the HTO activity concentrations dropped quickly following the drop in atmospheric HTO release in 2009. The time required to do so has been used to evaluate travel time [15], which appeared to be within the range of 5-12 months. We have used the well system to attempt to describe the transport of airborne HTO to well water using a single (landscape average) travel time.

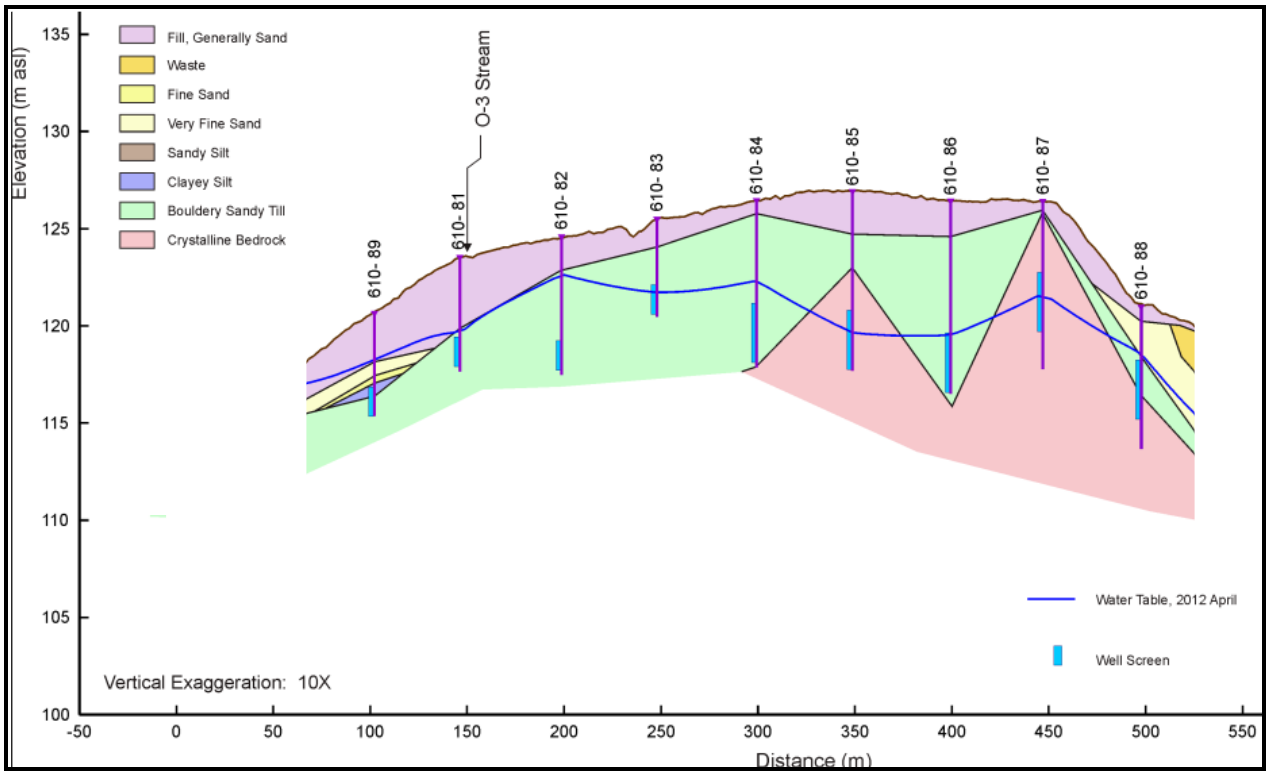

Figure 2. Stratigraphy along the cross section passing through the analyzed CNL wells.

3. Results and Discussion

The source (atmospheric HTO and HTO in precipitation) and the level of tritium in the sink (OBT) are related through deposition as illustrated in Figures 3 and 4. A similar relationship holds for the source (HTO in precipitation, lagged 6-months) and the activity concentrations measured in the 9 wells (sink) shown in Figure 5. The latter relationship provides the potential explanation (mechanism) of retarded atmospheric HTO transfer to OBT on a landscape scale. It also shows that a single (landscape average) travel time adequately describes transport of airborne HTO to well water.

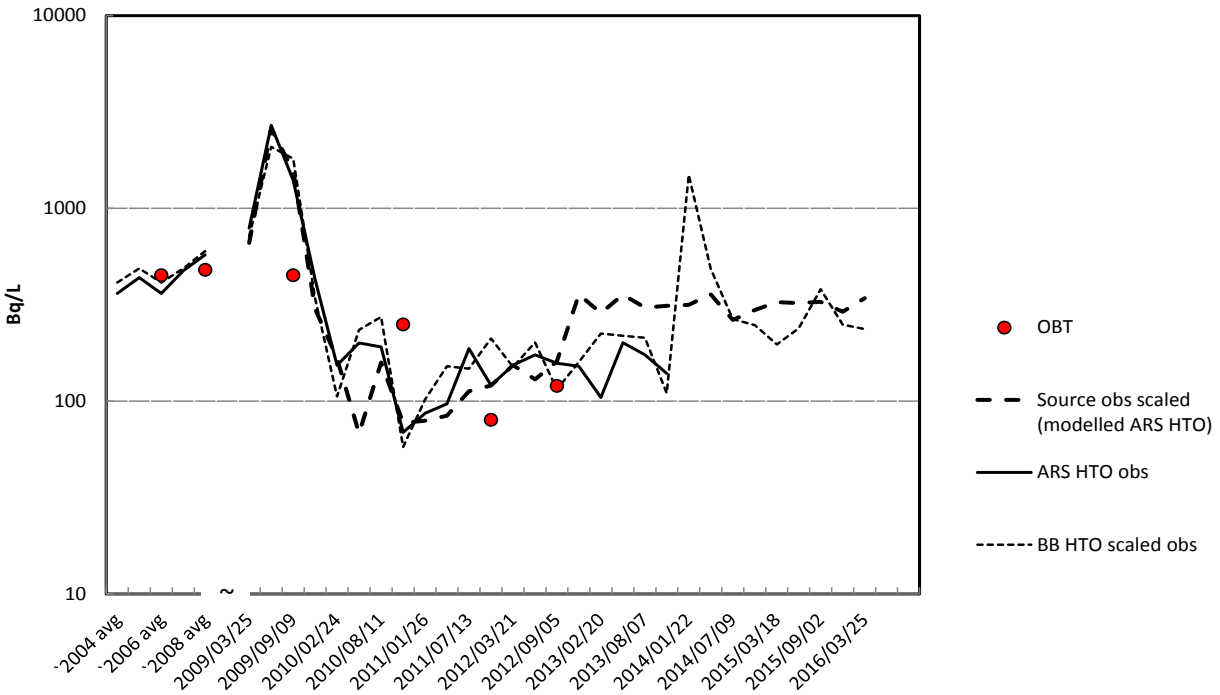

Figure 3. Timeline of atmospheric HTO: monitored at ARS garden plot (solid line), reconstructed by scaling down monitored HTO emissions at stack and NRU (thick dash line) and by scaling up of monitored at Balmer Bay (thin dash line). OBT in harvested vegetation is denoted by red markers.

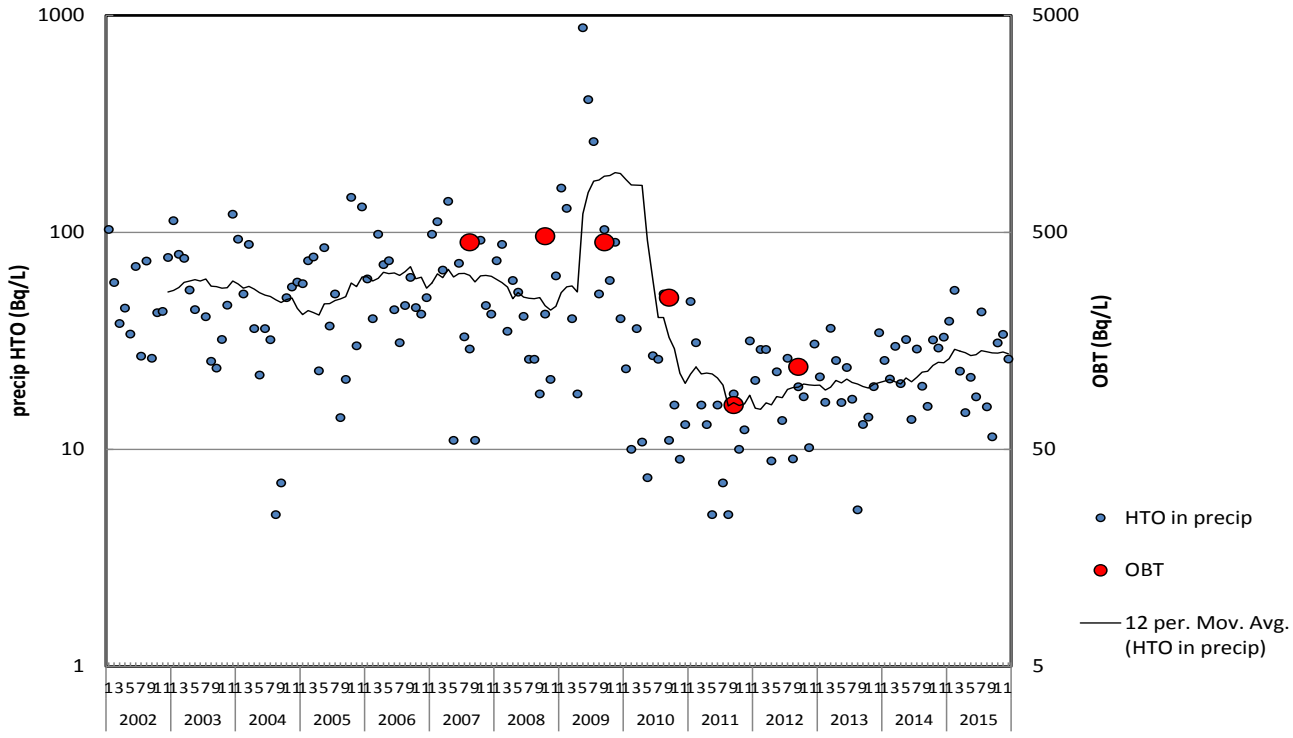

Figure 4. Timeline of monthly HTO in precipitation at Balmer Bay gate (small blue markers) and its 12-months moving average (solid line). Annual OBT measured at ARS is denoted by large red markers.

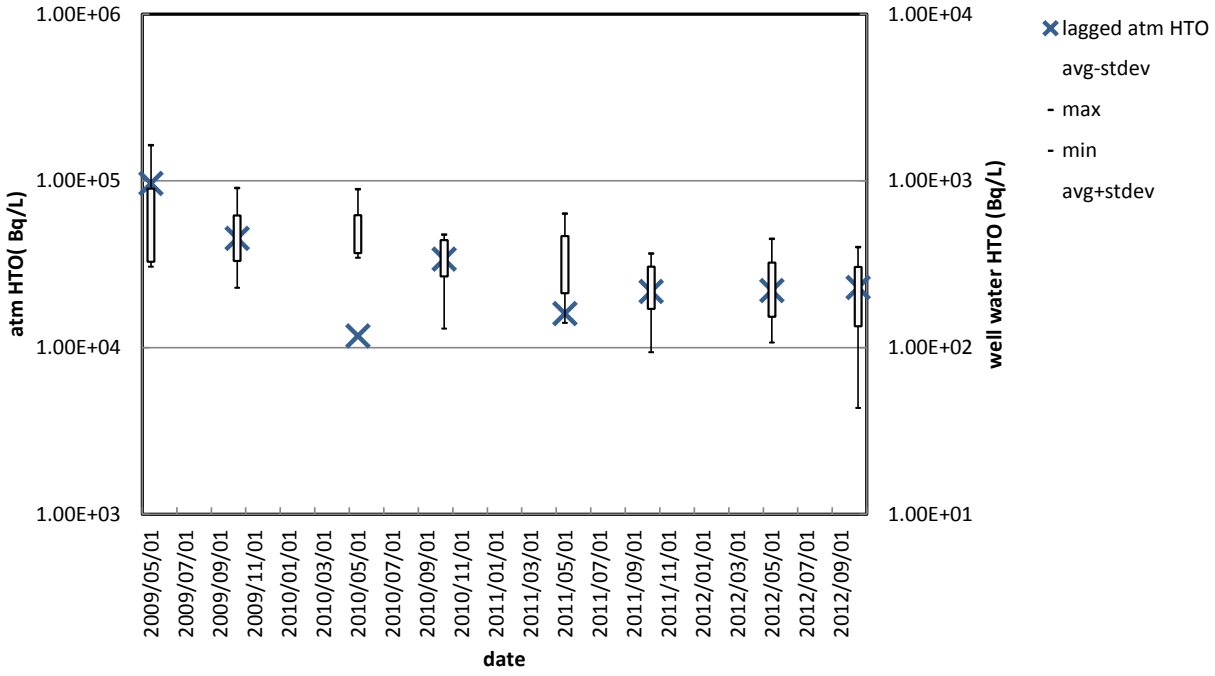

Figure 5. Timeline of water HTO activity concentration in nine on-shore wells represented by a boxplot (average plus-minus standard deviation with minimum and maximum marked by whiskers) and atmospheric HTO lagged 6-months upslope the well catchment area (denoted by crosses).

Precise correlations between the source and the sink emerge based on a total lag between the source and the sink approaching 12 months (see Figures 6 and 7). The total lag for analyzed monthly precipitation includes the contribution of the 6-month moving average used for HTO in precipitation. The best lag estimate was found using the functional dependence of the strengths of correlation between the source and the sink (using the coefficient of determination R2) on the lag. Results are presented in Figures 6 and 7. A one year long lag clearly provides the highest correlation between the measured atmospheric HTO at ARS experimental garden plot and the collected OBT (Figure 6). However, as noted maximum on quarterly scale is outstanding, it seems beneficial to repeat the same analysis for monthly observations to reduce uncertainty and get a better resolution of the peak. A similarly strong, yet less prominent, correlation between HTO in precipitation and OBT emerges for a total lag approaching 12 months (Figure 7). As noted, this lag takes into account the lag of 6 months applied to the moving average. At the landscape scale, our best ecological half-life estimate is therefore 1 year. The correlation between the properly lagged source and the sink (OBT in produce) is illustrated by scatterplots in Figure 8 (for atmospheric HTO) and Figure 9 (for HTO in precipitation).

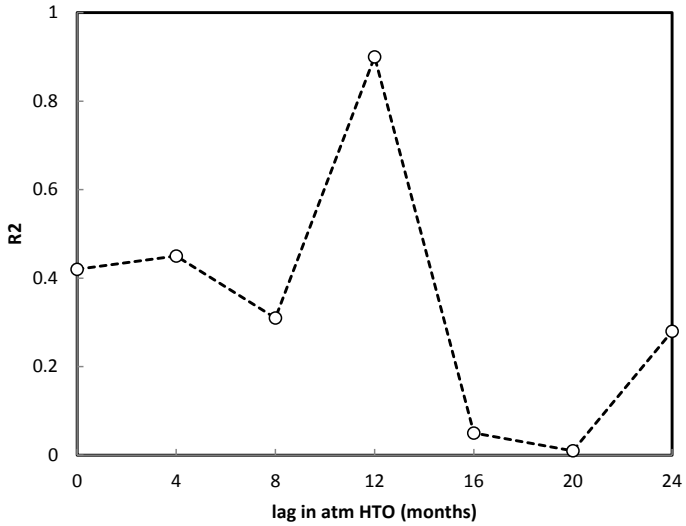

Figure 6. Coefficient of determination R2 calculated for correlation between OBT and lagged atmospheric HTO at ARS garden plot. Shown are calculations for different additional lag times.

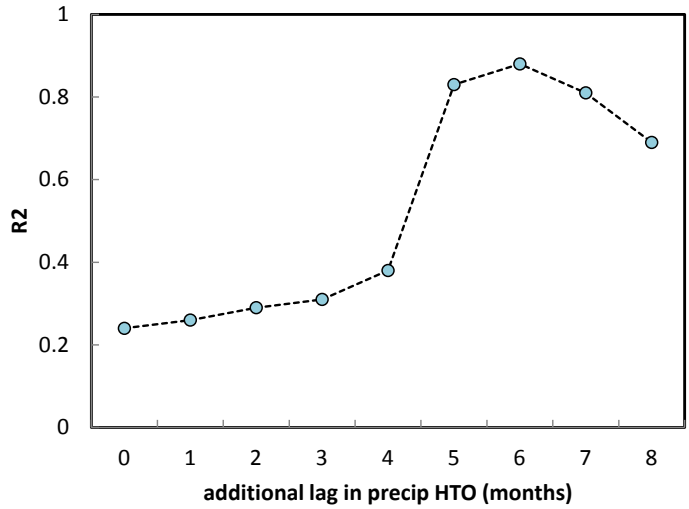

Figure 7. Coefficient of determination R2 calculated for correlation between OBT and 6-months moving average HTO in precipitation which is additionally lagged. Shown are calculations for different additional lag times.

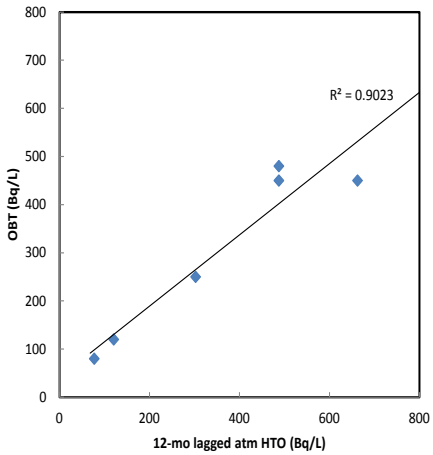

Figure 8. Relationship between OBT and atmospheric HTO lagged 12 month as per the best (the largest) coefficient of determination R2.

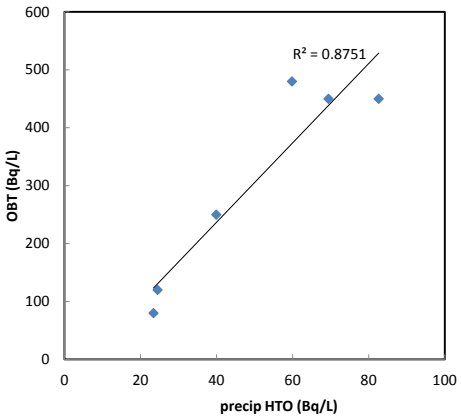

Figure 9. Relationship between OBT and lagged HTO in precipitation as per the best (the largest) coefficient of determination R2 (total lag comprised of 6 months plus the lag implied in the 6-month moving average).

This ecological half-life can be hypothesized to be linked to local recirculation of HTO through subsurface waters to local swamps and back to atmosphere, where it is relatively evenly dispersed in the surface layer on the local scale. This hypothesis finds support in lateral subsurface migration of HTO that was recently evaluated to be in the range of 6 months at CRL (on the basis of the HTO data collected from the nine drilled wells located sufficiently close to NRU to respond to the 2009 signal in atmospheric emissions) (see Figure 5). Due to the absence of significant differences in geomorphology at different well locations, we were able to define the mean travel time (estimated at about 6 months) which could be applied to all wells and thus to the whole section of catchment. Since residence time differs between wells due to the factors usually unavailable on a landscape scale (and as such ignored here), the coefficient of determination never surpassed the relatively small maximum value of 0.47 illustrated in Figure 10.

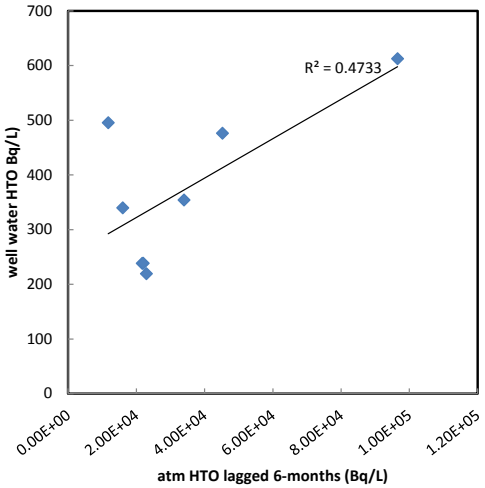

Figure 10. Relationship between well-water HTO concentration and 6-months lagged atmospheric HTO in the catchment area.

The lag associated with HTO lateral transport in shallow aquifer can be attributed to HTO's retention in the environment. However, other mechanisms can contribute to HTO's retention. For example, mechanisms linked to the freeze-thaw cycle could play a role. HTO retention in the root zone of mature trees could be contributing, as well as parameters like water table depth and interaction between mobile and stagnant water in soil pores [16, 17, 20, 21-23]. The decomposition of OBT in the organic layer could also merit investigation.

4. Conclusion

In the range of analyzed conditions encountered at CNL it was established that the ecological half-life of HTO $\boldsymbol{T}_3$ approaches one year. This value is in line with literature results [7, 10]. It is demonstrated, that the responsible mechanism could be the retention of HTO in shallow groundwater followed by lateral transport and subsequent release to the environment via evaporation from the surface of nearby swamps. This provides a direction for detailed hydrological analysis accounting for water table depth, the root depth, and other site specific parameters in the future. It is also established that this lateral transport can persist at the landscape scale. This implies the need of $\boldsymbol{T}_3$ for proper quantification of the transfer coefficient λ. Other mechanisms (e.g. freeze-thaw cycle) could also play a role in the retention of HTO. Determining if these are important contributors requires further investigation.

5. References

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