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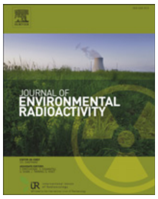

Tritium profiles in snowpacks

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ABSTRACT

The release of tritiated water (HTO) to the atmosphere during the winter can contribute significantly to snow contamination and to water–soil–plant contamination after the spring thaw. The dose significance of such a release depends on the persistence of tritiated water in the snowpack, which is primarily controlled by the HTO diffusion process in snow and the rate of re-emission into the atmosphere from the snowpack surface. Monitoring data collected after an acute winter release at Chalk River Laboratories and data obtained in winter over a chronically contaminated area were analyzed to estimate the diffusion coefficient of HTO in the snowpack. Under conditions of cold and dry snow, the diffusion coefficient lay in the range $1–2 \times 10^{-10}$ m^2 s^{-1}, an order of magnitude lower than diffusion in water but an order of magnitude higher than self-diffusion in ice. These results confirm the theoretical predictions (Bales, 1991). Values up to six times higher were found for warmer periods and just before spring melt, when other processes contribute to profile evolution. The low diffusion rate of tritium in cold, dry snow means that tritium remains in the snowpack throughout the winter, to be released during spring thaw to potentially contaminate surface water, soil and crops.

1. Introduction

The dose impact of tritium released to the atmosphere in winter is far from being understood. Some experimental data on dry and wet deposition of HTO to snow were obtained in the past (Barry, 1964; Papadopoulos et al., 1986; Davis, 1997), but little experimental information is available on the persistence of tritiated water in the snowpack. Also no data exist on re-emission of tritium into the atmosphere from the snow–atmosphere interface. It is generally assumed that the re-emission rate is very low and, as a first approximation, the snow can be considered a perfect sink. It is not clear if this assumption is valid in the long term in northern climates where the snowpack can persist for up to six months. For dose assessment, the processes contributing to the time evolution of the contaminated snowpack and the fraction of deposition still present in the spring are of relevance, as snow-melt releases tritium to the land surface where it can potentially enter groundwater, soil water and crops. The data pertaining to bomb fallout (Jouzel et al., 1979) are of limited usefulness in studying these processes, due to the quasi-continuous character of the source and the exotic site (Antarctica). Recently, theoretical work on pollutant transport in snow was published by Bales (1991) which emphasises pollutant diffusion, and includes the role of chemical transformation, the mobile air phase and the immobile water and ice phases. This theory can be used to obtain an effective diffusion coefficient for HTO if all key parameter values are known or can be estimated.

In this paper, we will present experimental information on HTO transport in the snowpack, using a subset of the monitoring data accumulated after an accidental release of HTO at Chalk River Laboratories (CRL) (Davis et al., 1992), as well as winter data from a survey of a chronically contaminated surface (Workman et al., 1993). Both sets of data were obtained with other purposes in mind and are not ideal for a full quantitative analysis. We will restrict our study to the process of HTO diffusion in the snowpack itself, and use simplifying assumptions in order to test Bales' (1991) theory. Due to limits on the experimental data, processes at the snow–atmosphere interface are not considered in detail, but are included in a simplified manner. It is unlikely that re-emission played a major role in the evolution of the tritium profiles since subsequent falls of snow uncontaminated by tritium isolated the bulk of the HTO from the atmosphere.

2. Material and methods

2.1. Release and sampling information

The HTO release began about midday, 1991 January 12, following a pipe break in the NRU (National Research Universal) reactor building that resulted in a spill of tritiated heavy water. A site map revealing the tritium sources and relevant

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monitoring stations is provided in Fig. 1. About 700 TBq of HTO were released to the atmosphere over the period January 12 to January 30, with two thirds of the total released in the first 3 d. Approximately half of the HTO was emitted through various roof and side vents of the reactor building itself, and the remainder through a 50-m exhaust stack located on a ridge 1 km to the west of NRU and 35-m higher in elevation. The first snow following the start of the release fell in the morning of January 14. Frequent short periods of light snow occurred between January 14 and January 19, and from January 28–29. Thus the highly HTO-contaminated snow that fell in the first few days was covered in the following days by relatively uncontaminated snow, and the main bulk of the HTO was consequently isolated from the free atmosphere.

An intense monitoring program was started soon after release began and continued until spring thaw (Davis et al., 1992). Snow samples were collected beginning January 13 and ultimately more than 200 samples were gathered at distances from 50 m to 40 km from NRU. The samples were collected as cores using Lucite tubes with an area of 0.01716 m^2, and by cutting a fresh face in the snowpack and taking out successive layers with a flat trowel of known area. The samples were sealed in plastic bags and returned in coolers to the laboratory, where they were weighed, melted and counted by liquid scintillation.

Two of the monitoring sites yielded time series of HTO profiles in the snowpack that were of sufficient quality to provide information on HTO diffusion in snow. The first site (S1) was located in a meadow in the forest, approximately 2.4 km from the exhaust stack and 3 km from the reactor building. This site had the advantage of being close to a permanent monitoring station (Station M) where snowfall, rainfall and snowpack height are routinely measured. The second sampling site (S2) was situated 400 m from the reactor building. At these sites, sectioned cores were taken every 24 h from January 14–17, and then at one or two week intervals until complete snow thaw at the end of March. The top 20 cm of snow was collected in 1 cm sections in the first 2 d but later in January in 2 cm sections in order to minimize the sampling errors. Later the sampling was in 5 or 10 cm sections from mid-February to the end of March. The uncertainty in the total HTO inventory was estimated to be less than 20%, with the error in individual sections somewhat higher, up to 30%. In the present paper the data in the first 2 d were omitted, as sampling errors were high.

From the point of view of studying HTO movement through snow, both sites are far from ideal. S2 is situated on relatively flat terrain with slope less than 5°, but nearby buildings can induce turbulence and temperature effects that influence the snowpack depth history (e.g., frequency and amount of deposition, erosion and melting). At site S1, the sampling was done in a small area at the bottom of a depression about 10 m in diameter and 1 m in depth where off-site snow and snow-melt were collected and mixed with on-site snow and snow-melt. Also, nearby bushes and trees likely induced local turbulence affecting the transport and deposition of the snow. For the time period when the complete snowpack depth was sampled (end January–end March), local turbulence effects can be observed from the differences in snowpack height (Fig. 2) measured at the two study sites (S1, S2) relative to that at the monitoring site (M). Generally the site data are 10–70% higher than the monitoring station values with no correlation. This site-specific difference in snowpack height, and the unavailability of the total core in January, make our analyses more uncertain, as the snow-ground interface is the only fixed boundary and reference for any dynamic description.

Air temperatures measured at a site about 500 m southeast of NRU are also shown in Fig. 2. The warm periods in the first two weeks in February and March resulted in partial melt, affecting the apparent snowpack profile of HTO. As diffusion is temperature controlled, changing snow temperature must be considered in the analysis of diffusion coefficients.

The other data in the diffusion analysis were taken from snow samples collected above a wetland (site S3) chronically contaminated by an underground tritiated water plume (Workman et al., 1993). Here, 12 d after the ground became snow-covered in 1992, the snowpack was sampled in four layers of 2 cm each. The core showed a strong decrease in HTO concentration from bottom to top, apparently due to diffusion of tritium from the frozen surface into the snowpack above.

3. Results

3.1. Time-dependent HTO profiles

For the thinner snow layers of 1 or 2 cm sampled in January, the relative error in sample thickness was quite large, and this resulted in a large variability of computed snow density. This in turn induced large errors in the local volumetric concentration of HTO. In order to minimize such sampling errors, we analyzed and present the data in terms of cumulative inventory as a function of cumulative water equivalent of snow layers. The water equivalent of a given layer is the amount of water contained within the layer. It can be thought as the depth of water that would result if the layer

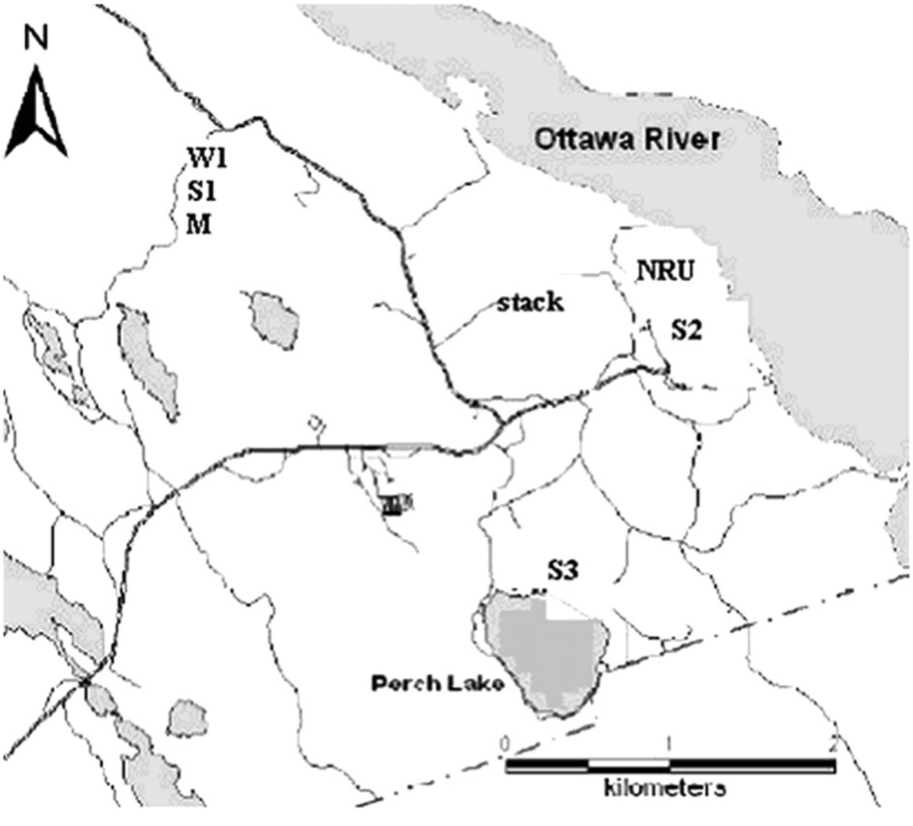

Fig. 1. Site map showing tritium sources NRU and stack, sampling sites S1, S2, S3, W1 and M, and the location of meteorological measurements.