Retention and dispersion of HTO in snow

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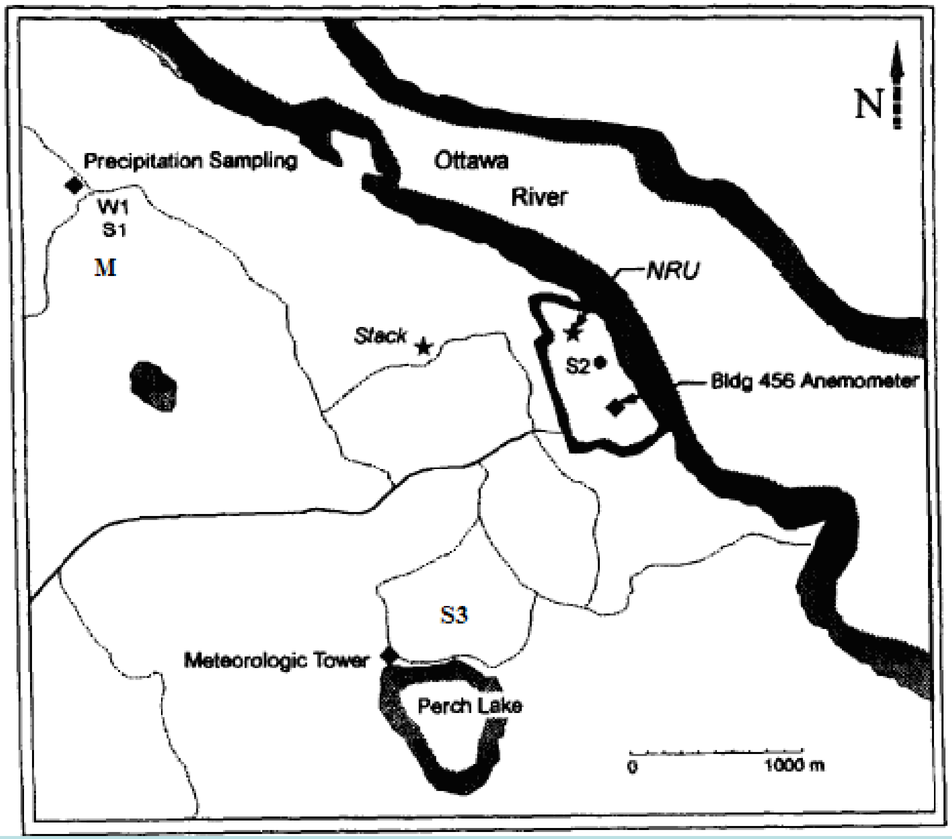
Submitted to JER

EMRAS II
Approaches for Assessing Emergency Situations
Working Group 7
“Tritium” Accidents
Vienna 25-29 January 2010

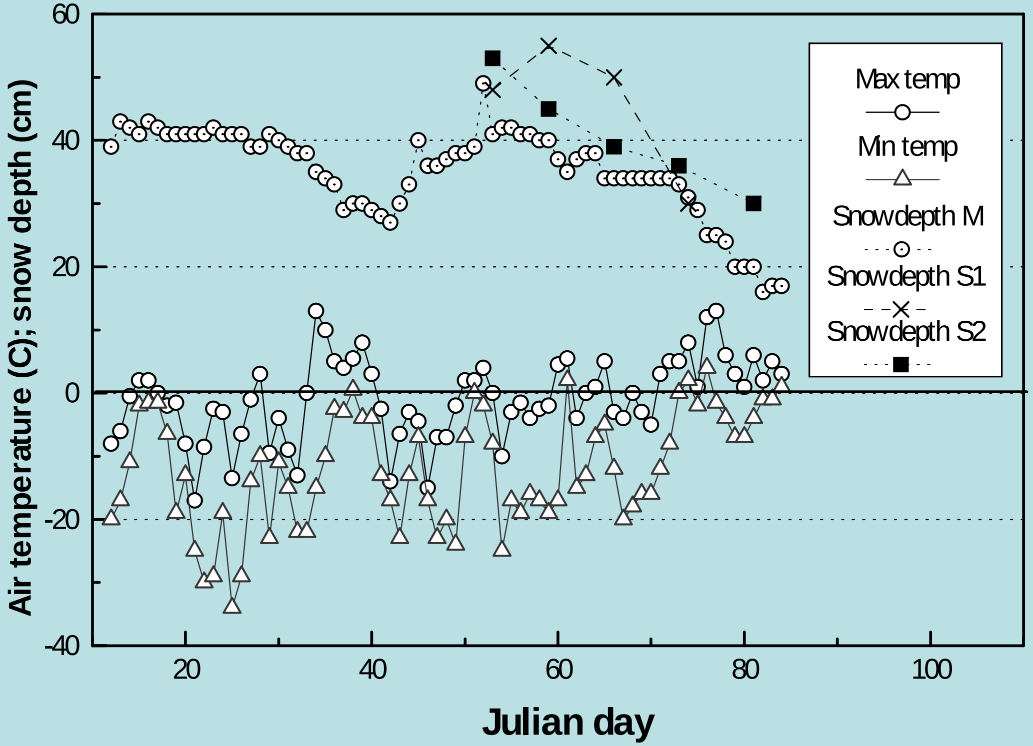
SNOW NOT IMPORTANT (U.K.),
WINTER IS A PROBLEM (Canada))

- In this CONTRIBUTION, 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.

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



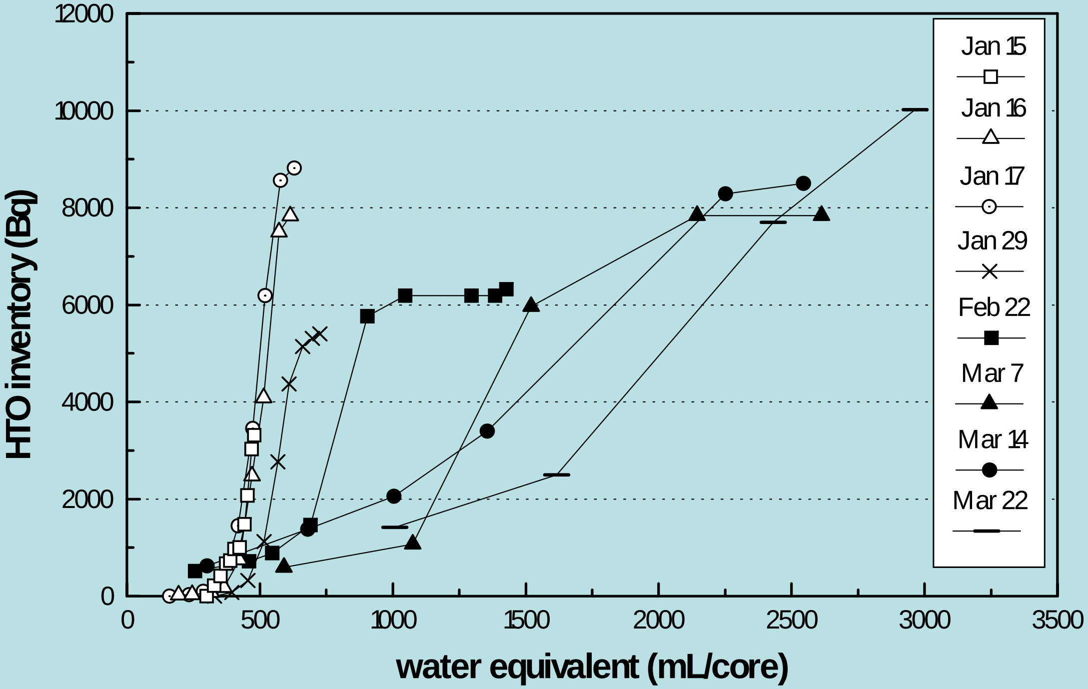
Time evolution of minimum and maximum air temperatures and snowpack depth. Snowpack depths for sites S1 and S2 are given only when the complete snowpack was sampled and recorded.



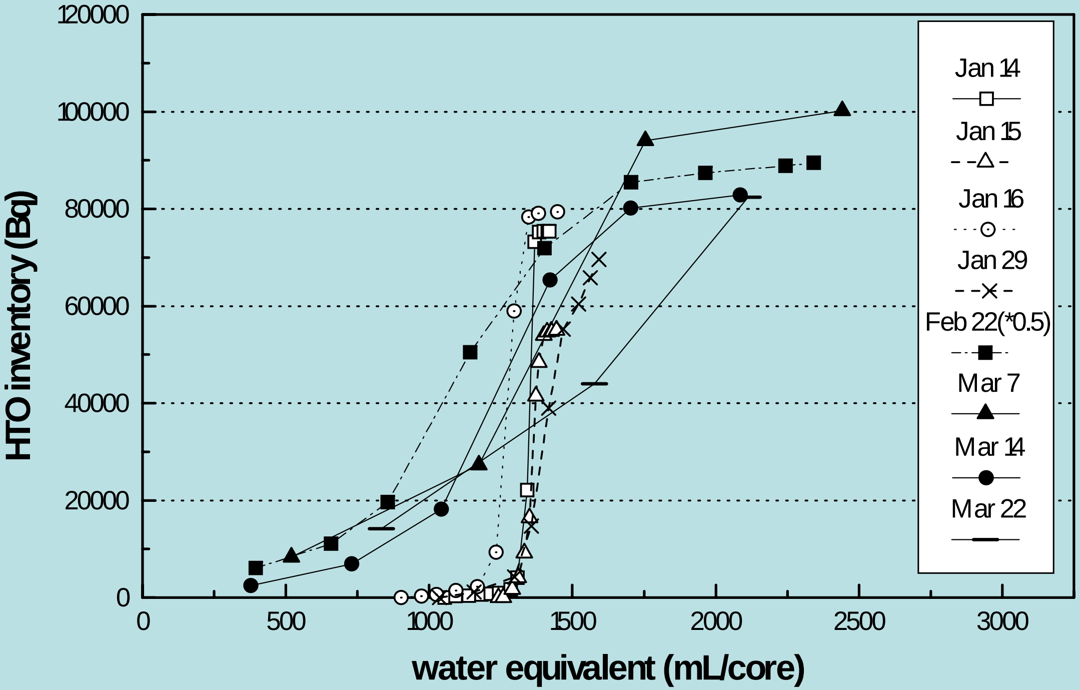
Material and data represenatation

- In order to minimize sampling errors, we will analyze 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 of as the depth of water that would result if the layer melted instantaneously. Adding layers from bottom to top, we obtain the cumulative water equivalent.
- For each layer we measure the inventory of HTO. The cumulative inventory up to some height is the sum of inventories from bottom to top.
- In a plot of cumulative inventory versus cumulative water equivalent, a short-term acute deposition of HTO-contaminated snow, followed by uncontaminated snowfall and no subsequent HTO transport in the core, will appear as a step function. Any dispersion of HTO will alter the shape and slope of the function near the deposition layer, and/or change the height of the step.

Cumulative HTO content as a function of cumulative water equivalent in the cores at site S1. The origin is at the bottom of the core.



Cumulative HTO content as a function of cumulative water equivalent in the cores at site S2. The origin is at the bottom of the core. Plotted data for February 22 are half the observed values.



theory

An approximate way to obtain information on the effective tritium diffusion coefficient in snow (D) is to apply the analytical solution of the classical diffusion equation, considering the concentration pattern near the maximum of the profile and, assuming an infinite medium so that boundary conditions can be ignored. With a known initial concentration distribution C(x,0) = F(x), symmetrically centred, the solution of the diffusion equation at a distance x from the maximum and at time t is (Moltyaner and Paniconi 1984):

$$C(x,t) = (4\pi Dt) - 1/2 \int_{-\infty}^{+\infty} \exp[(x-z)^2 /(4Dt)]F(z)dz. \tag{1}$$

The initial condition, normalized to the total inventory, was chosen to reproduce the measured concentration profile immediately after the acute deposition on January 15-16, and is given by:

$F(x) = 0.5\ \alpha \exp(-\ \alpha|x|)$

Values of the effective diffusion coefficient D from 2 x 10-11 to 2 x 10-10 m2/s

were obtained from analysis

It is known (Gray and Male 1981, Bales 1991) that the diffusion coefficient of a pollutant in snow, carried in the air phase, depends on the snow temperature gradient and density, and increases in the warm period before snow melt (up to its value in water).

At any time, the system of coupled differential equations is:

dy1/dt = R(y2-y1) (3a)

dyk/dt = R(yk+1 + yk-1) -2Ryk k=2 to (n-1), where n is the surface layer (3b)

dyn/dt = Ryn-1 -2Ryn + vdCa (3c)

where yk is the activity in layer k and the layers are counted from bottom to top in the core. The interlayer transfer rate R is linked with the effective diffusion coefficient D and layer thickness z:

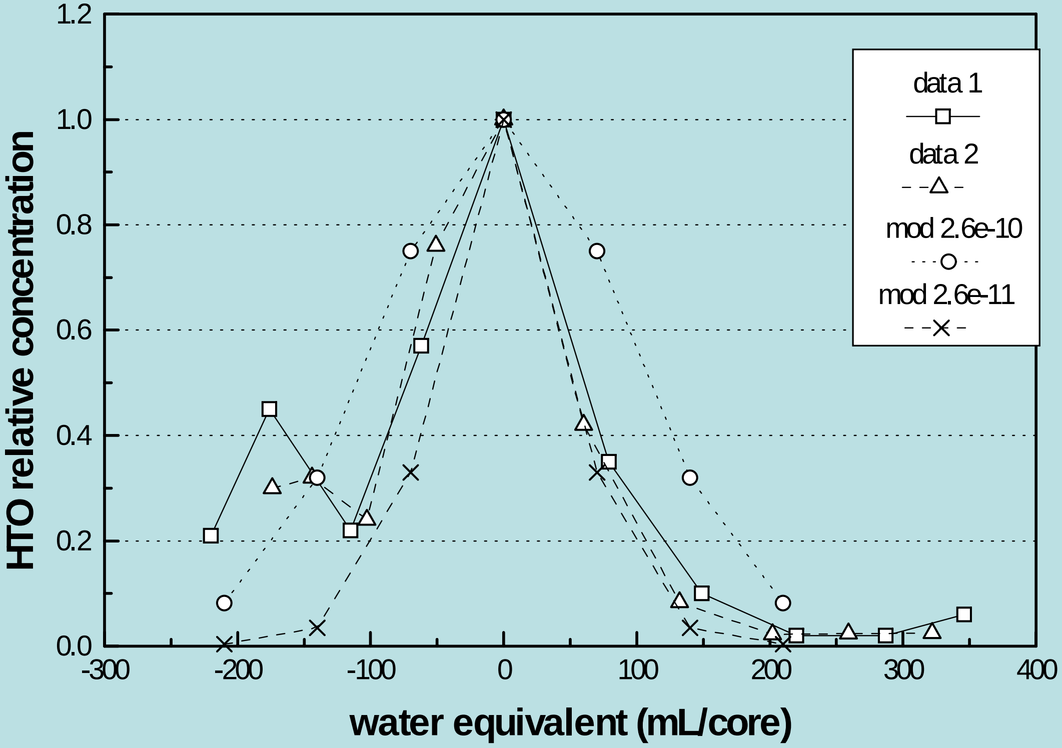
R = D/z2.

Theory 2

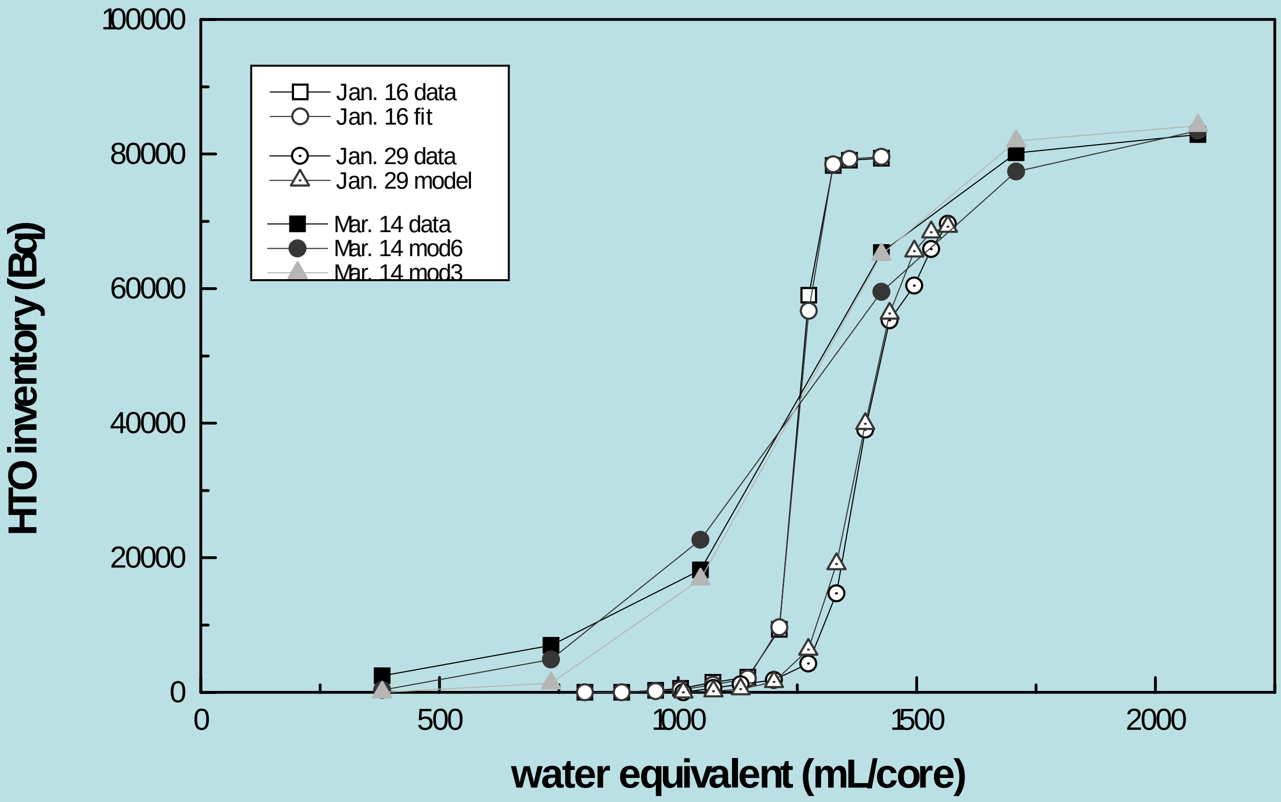
- The one-dimensional transport equation for a non-decaying chemical species (Bales 1991) then reduces to:
- $\theta\ dCa/dt + (1-\theta)\ dCi/dt = \theta\ Dv\ d2Ca/dx2$ (6)
- where θ is the snow porosity (air volume fraction), Ca and Ci are the solute concentrations in air and ice, and Dv is the diffusion coefficient of water vapour in the air pore space. Due to the rapid exchange between tritium and hydrogen, we can assume that the ratio between the concentrations in air and ice is given by the absolute humidity q (m3 water per m3 air) and we obtain
- $dCi/dt = [\theta q/(\theta q+1-\theta)]\ Dv\ d2Ci/dx2 = Ded2Ci/dx2$ (7)
- which is the classical diffusion equation with an effective diffusion coefficient
- $De = [\theta q/\ (\theta q+1\ -\theta)\ ]\ Dv$ (8)

Relative HTO concentration as a function of depth at site S2. The concentration has been normalised to the maximum observed value and depth is expressed as cumulative water equivalent measured from the layer with the maximum concentration. Negative water equivalents correspond to the top of the snowpack, above the deposition layer. Shown are two replicate profiles from January 29, and

predictions of Eq. (1) for diffusion coefficients of 2.6 x 10-11 and 2.6 x 10-10 m2/s.

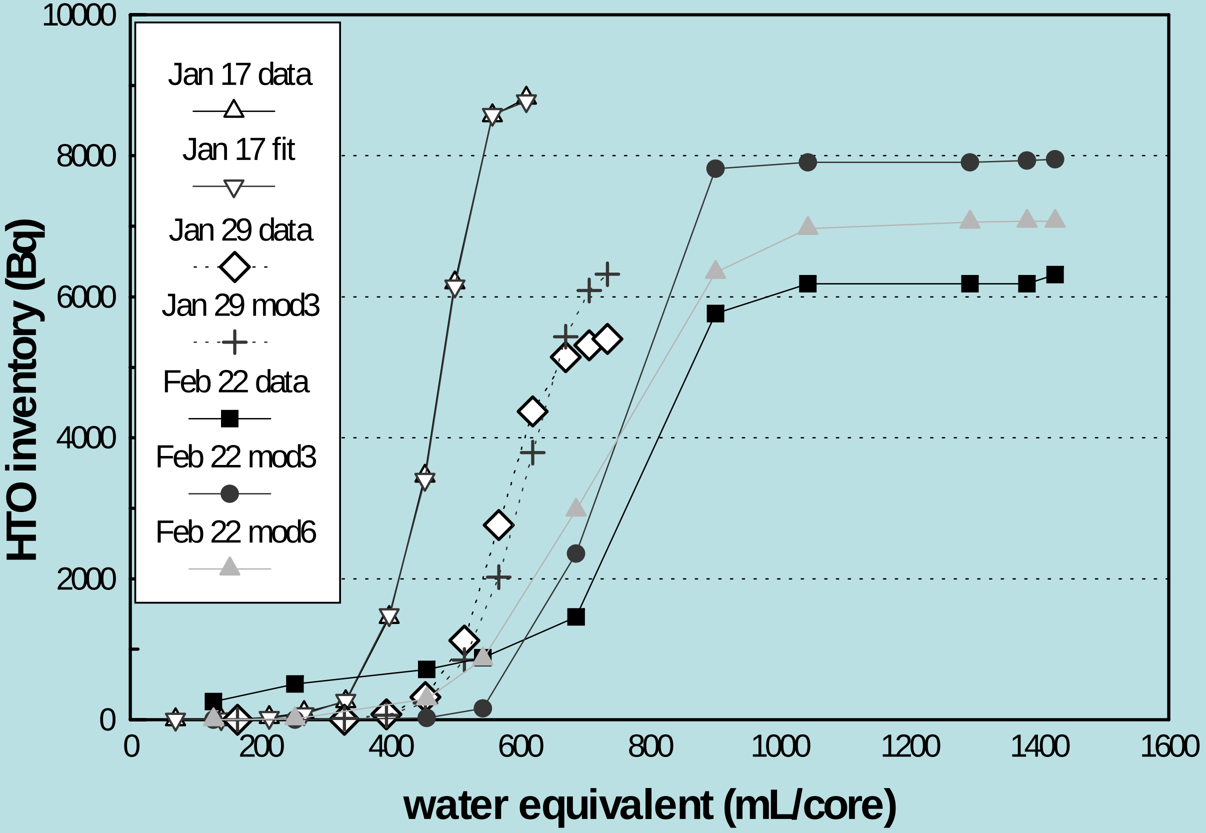


Comparison of observed S2 cumulative profiles with calculations from Eq. (3). The diffusion coefficient for January was 1.8 x 10-10 m2/s; for February and March it was three times or six times greater (mod3 and mod6 results respectively).

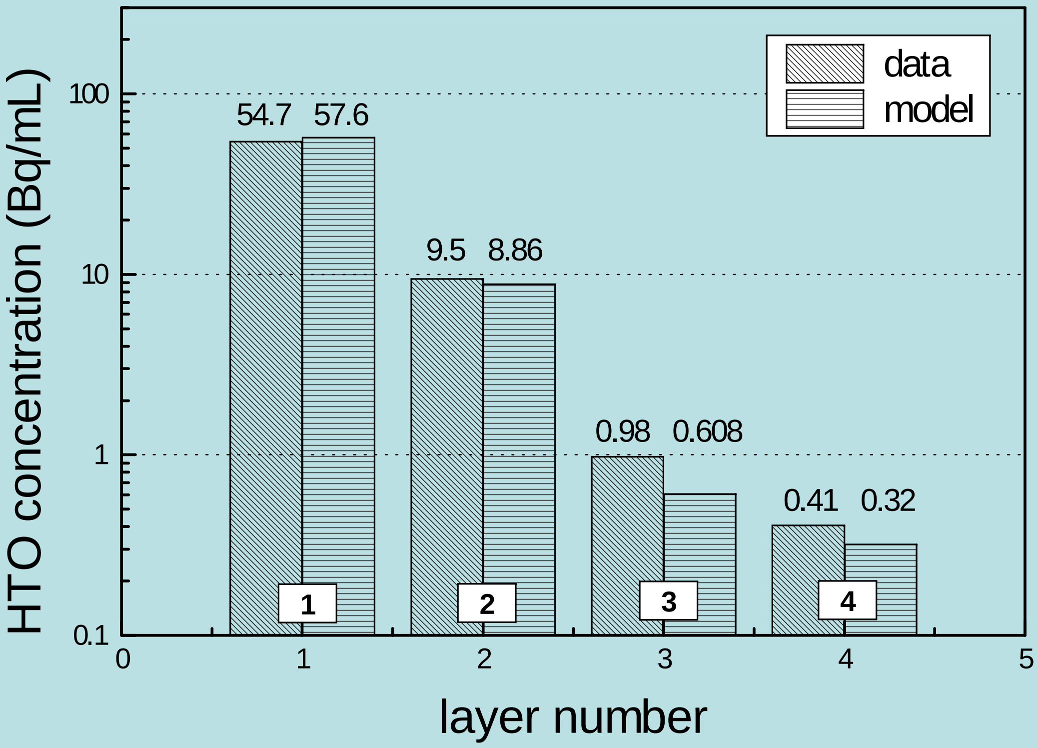


Comparison of observed S1 cumulative profiles with calculations from Eq. (3). The diffusion coefficient for January was 1.2 x 10-10 m2/s; for February and March it was t

t**hree times or six times greater (mod3 and mod6 results respectively).**



Predicted and observed HTO concentrations as a function of depth at site S3 twelve days after snowfall. The HTO concentration in ice beneath the snowpack was set to 200 Bq/mL, the central value in the range of experimental data. Layer 1 is adjacent to the underlying surface and layer 4 is adjacent to the air.



conclusions

- Data obtained during an acute release of HTO in winter and above a snow-covered, contaminated surface were analyzed to determine the diffusion coefficient of HTO in snowpacks. Although the processes at the snow-atmosphere surface were not fully characterised, the analysis was nevertheless possible since the deposited HTO was isolated from the atmosphere by new, uncontaminated snowfalls. With the exception of the pre-melt period and the warmer time sequences (snow temperatures >-5°C), we can conclude that the diffusion coefficient for HTO in snow has a range (1-2) x 10^{-10} m^2/s, quite close to the theoretical predictions of Bales (1991) when uncertainties in the data and model are taken into account.
- **A large fraction of the initial HTO inventory, greater than 70%, was preserved in the snow cores studied here until spring thaw.** This is likely an upper limit to the amount that could be retained. For snowfall histories in which the acute deposition is followed by a dry period, the persistence of HTO in the snowpack will depend strongly on surface processes. The HTO will be released from the snowpack during spring thaw. The consequence for tritium contamination of ground water, soil water and crops should be assessed, through either field experiments or the use of environmental tritium models.

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