Synchrotron Radiation for Detecting Movement of a Volatile Soil Liquid

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Introduction

In porous media coupled heat and water fluxes are important for engineering, environmental, agricultural, and food science applications. One example is analysis of heat dissipation from underground nuclear waste storage facilities. Other volatile liquids are also of interest, particularly with respect to environment.

Although numerous laboratory studies have been reported, they have been characterized by low spatial resolution or reliance on destructive sampling. Our work used synchrotron x-rays for evaluating redistribution of a volatile liquid in nonisothermal soil at high spatial resolution.

Dibromomethane (DBM) proved advantageous as the volatile liquid because of its high attenuation coefficient. While DBM clearly has properties differing greatly from water, this disadvantage was outweighed by the highly accurate, nondestructive concentration measurements possible with DBM. Important similarities of DBM and water are that both are pure, stable, volatile liquids over the 0-40°C temperature range. We mapped DBM concentration in a sand column while alternating a 1.1 K/mm temperature gradient in pursuit of our objective to determine transient volatile liquid transport with high resolution.

Methods and Materials

A sample cell 9.5 mm diameter by 25.4 mm long contained quartz sand, DBM, and air (volume fractions 0.691, 0.040, and 0.269). A temperature gradient was created by circulating water at 5 and 35°C through heat exchange blocks at the ends. The sample cell was mounted on a motion control stage. Concentration of DBM was calculated from the attenuation of a 44 keV beam of 28 (horizontal) by 4.8 mm cross section. X-rays were detected via a scintillation crystal and CCD camera. Data were gathered in each of four vertical positions of the stage. The 'white' data, I_o, was gathered in a position that allowed the beam to pass completely above the sample. Images of the cell were taken at three stage heights 4 mm apart, with 0.8 mm overlap.

The experiment was conducted over a 22 h period on Dec. 8 and 9, 2000. At time zero, the cell, initially at ambient temperature, received 5.0° and 35.0°C water circulated through its heat exchangers. These initial temperatures were maintained until steady-state DBM distribution was observed. The temperature gradient was then reversed. Movement of DBM then resumed until steady state was reached in the opposite direction. We moved the DBM four times starting from steady-state configurations. Movements were left, right, left, right so we correspondingly designated them L1, R1, L2, and R2.

We used 30 s exposures for all the images. Two white (I_o) exposures and two complete cell scans were done each half hour. Data were evaluated via the exponential law, which states

$$I = I_o \exp[-m^*t], \qquad (1)$$

where I and I_o are the transmitted and incident intensities, m* is the linear attenuation coefficient, and t is the absorber path length. For multiple absorbers the thickness of DBM is

$$t = (\ln(I_o) - \ln(I) - A)/m^*, \qquad (2)$$

where A is the sum of m*t for all other materials. Intensities I and I_o were found by subtracting CCD dark current from the original CCD data. Preprocessing removed 'zingers,' large CCD signals due to x-rays directly striking the CCD array, from the data.

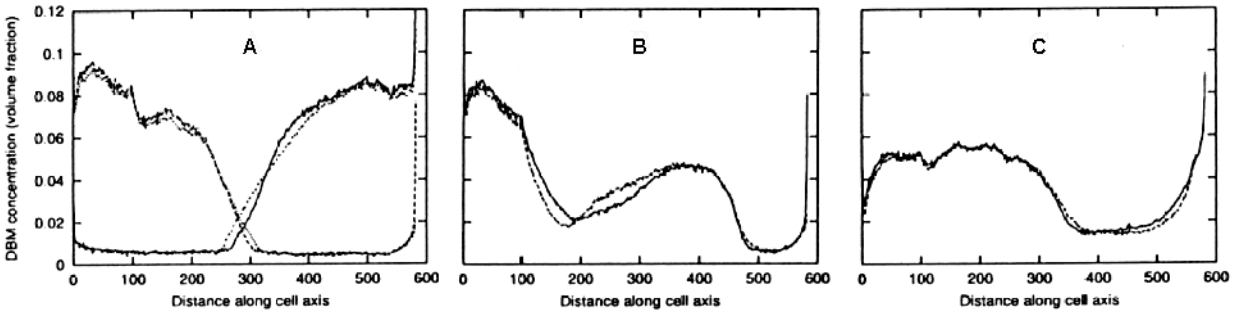

FIG. 1. Average DBM concentration along cell length, in 42.5 μm pixels; (A) L1, L2, R1, and R2 at 0.0 h, (B) L1 and L2 at 2.5 h; (C) R1 and R2 at 1.25 h.

Results

It took 5.1 hours for complete movement from one steady-state condition to the opposite one. The data contained a vertical gradient, which was eliminated by normalizing each row of pixels to the overall DBM concentration. Loss of DBM from the cell during the experiment was determined gravimetrically at less than 4.2%.

Spatial distribution of DBM concentration was determined at 86 times. Average DBM concentration along the axial direction of the sample was plotted at selected times (Fig. 1A-C). The left and right steady-state conditions are each represented by sets of two lines (Fig. 1A), which nearly overlie one another. Comparison of the two left movements at 2.5 h into the movement (Fig. 1B) shows highly similar profiles. Comparison of the R1 and R2 movements (Fig. 1C) at 1.25 h into the movement also shows excellent agreement.

Discussion

We found nearly identical movements of DBM occurring during L1, L2 and R1, R2 (Fig. 1). There is also excellent agreement (not shown) between the L and R movements as seen, for instance, comparing L and R movement plots at near-equal times into the movement. When left-right direction is reversed in one of the two and they are overlaid, the agreement is striking. At the same time, however, there is evidence of longitudinal structure in the cell packing in the steady-state distributions. When DBM is concentrated at the left steady-state configuration in Fig. 1A, the peak near 30 pixels and the dip near 100 pixels indicate packing differences.

Although parameter values may differ, the theory of coupled transport of heat and water in soil should apply for volatile liquids other than water, also. The theory is based on liquid and vapor transport driven by simultaneous gradients of potential energy and temperature. Certain features of the present results suggest that DBM vapor flow dominates at particular times and locations. Consider, for instance, Fig. 1C. The buildup of DBM between 500 and 580 pixels probably resulted mostly from vapor movement through a 'valley' of concentration, which is between 350 and 500 pixels at the moment of Fig. 1C. In Fig. 1B, where movement is to the left, the 'valley' extent is greatly reduced, with its low point at about 190 pixels. Still, liquid movement is theoretically relatively restricted in this low-concentration region.

The basic advance reported here is capability to nondestructively sense volatile liquid content at much finer spatial resolution than before. This is important because reproducibility of previous experiments is generally inadequate to allow detailed model testing. That is, model outputs usually provide detail far greater than experimental data. The data of Fig. 1 appear to hold promise of providing more complete and easier comparison to model output.

If scale independence applies, results can be extrapolated to larger scales. This is of interest because an experiment completed in time t in a laboratory column 0.025 m long, as ours, will require $(2.5/0.025)^2\ t = 10^4\ t$ to complete in a field soil column 2.5 m long. Thus, what took about 5 h in our experiment would require 50,000 h, or 5.71 yr, at field scale.

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