



## The effects of matrix diffusion on solute transport and retardation in undisturbed peat in laboratory columns

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Received 13 March 1995; revised 18 September 1996

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### Abstract

Experiments were performed to assess the nature of solute transport in peat by using step-inputs of a NaCl solution in laboratory columns of undisturbed peat. Peat has a dual-porosity matrix with inter-connected pores that actively transmit water, and dead-end and closed pores formed by the remains of plant cells. The proportion of dead-end and closed pores increased at depth, where the state of decomposition of organic material is more advanced. These dead-end and closed pores act as a sink for solute. Breakthrough at  $C/C_0 = 0.5$  occurred much later than the total active pore volume in the column, indicating that solute retardation occurred. This retardation was attributed to diffusion of the flowing solute into the closed and dead-end pores (matrix diffusion). Greater retardation occurred at depth, increasing from 2.7 at 0.20 m to 7.3 at 0.62 m, corresponding to the greater volume of closed and dead-end pores there. Retardation was also velocity dependent, with higher velocity resulting in less retardation of solute since there was less time available for solute to be abstracted from the flowing water into closed pores. Matrix diffusion was shown to enhance dispersion at lower flow velocities, and dispersion increased with depth. Peat effectively attenuated the conservative solute through matrix diffusion, and heterogeneity in peat properties influenced the effectiveness of this retardation. © 1997 Elsevier Science B.V.

*Keywords:* Matrix diffusion; Solute transport; Retardation; Peat

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## 1. Introduction

Although wetlands are increasingly being used as waste repositories and for wastewater treatment, the physical properties of peat which govern contaminant attenuation are poorly understood. Peat is an organic soil composed of partially to well-decomposed plant remains, which accumulate in a wet environment. Peat can effectively attenuate the movement of contaminants from groundwater by adsorption (Viraraghavan and Ayyaswami, 1987), including metals (Dumontet et al., 1990; Machermer and Wildeman, 1992), organic contaminants (Viraraghavan and Ayyaswami, 1989; Zytner et al., 1989), and divalent cations (Heathwaite, 1991). Peat can also remove contaminants through biochemical reactions (Bowmer, 1987) and filtration of suspended material (Brown and Stark, 1990). However, the understanding of the physical properties of peat which result in contaminant retardation is limited. Studies have attributed solute retardation to the above processes, while the properties of the peat matrix and its contribution to retardation have been largely ignored.

Peat, like fractured geologic media, has a dual porosity matrix (Loxham, 1980). The effects of “matrix diffusion” in fractured geologic media (Feenstra et al., 1984; among others) and in porous media with layers of contrasting hydraulic conductivity (Gillham et al., 1984) are well documented. In a layered porous medium, if the hydraulic conductivity of a layer is much higher than that of the adjacent layers, then groundwater flow and advection of contaminants will occur entirely within the high hydraulic conductivity layer. Concentration gradients between the high and low hydraulic conductivity layers will result in the diffusion of contaminants into the dead zone, causing decreased concentration of contaminants within the actively flowing layer. Grisak et al. (1980) found that matrix diffusion in fractured till can significantly retard weakly sorbing species in groundwater. The matrix in fractured rock can trap a significant amount of solute (Feenstra et al., 1984). Matrix diffusion in fractured media (Neretnieks et al., 1982) and dual-porosity porous media (Gillham et al., 1984; Price and Woo, 1988) enhances dispersion owing to solute loss at the front.

Limited knowledge exists regarding the effects of matrix diffusion on solute retardation and transport in peat. Some theoretical studies have been undertaken (Loxham, 1980), and column tests in the laboratory have also been conducted (Loxham and Burghardt, 1983; Price and Woo, 1988). These studies show that peat can attenuate contaminants through matrix diffusion into closed and dead-end pores. In addition, Price and Woo (1988) determined that the amount of retardation could possibly be dependent on water flow velocity. In a field-scale experiment (Hoag and Price, 1995), solute retardation was manifested as a plume with a near-stationary peak proximal to the solute injection point. Additionally, most of the solute transport occurred in the upper 40 cm of the peat, where water flow velocity was highest.

In order to understand the field-scale problem, the effects of matrix diffusion and water flow velocity on retardation and dispersion must be explained. The objectives of this paper, then, are to determine: (1) how the structure and properties of peat affect solute retardation and dispersion, and (2) how water flow velocity affects retardation.

## 2. Solute transport in peat

Peat is a porous medium with two major hydrological zones. The acrotelm (Ingram, 1978) is the upper, hydrologically active layer which comprises living and poorly decomposed plant material. Hydraulic conductivity and total porosity decrease significantly with depth in the acrotelm, and bulk density increases (Boelter, 1965). Below a depth of approximately 0.3 to 0.5 m, catotelm peat (Ingram, 1978) displays little variation in hydraulic properties (Boelter, 1965). Consequently, most of the solute advection will occur in the upper 0.3 to 0.5 m (Dumontet et al., 1990; Hoag and Price, 1995), although vertical solute migration can be important on the time scale of several, to several tens of years (Siegel et al., 1995; Charman et al., 1994).

Peat is a dual-porosity medium, with two main types of pores present: (1) interconnected pores which can actively transmit water and advect solute, and (2) closed and dead-end pores formed by partially intact to intact plant cell remains (see Hayward and Clymo, 1982) (Fig. 1). The active porosity of peat (the proportion of the total volume of peat which is pores that actively transmit water) varies significantly with depth, and can be as high as 0.8 near the surface and as low as 0.1 in the catotelm (Siegel et al., 1995; Romanov, 1968). The active porosity tends to decrease with depth due to compaction of the peat matrix by the weight of the overlying plant material and water trapped in closed pores (Romanov, 1968), as well as its increased state of decomposition. The remaining volume is occupied by closed and dead-end pores, as well as plant (solid) material.

Solute retardation may arise from adsorption, biological uptake, chemical reactions, and matrix diffusion. Price (1994) and Damman (1987) note that sodium is very mobile in peat, and is not affected by uptake by plants, although high sodium concentrations may result in some sorption (Chason and Siegel, 1986; Price, 1994). While the closed pores may not actively transmit water, they can trap a significant amount of solute from the actively flowing solution (Price and Woo, 1988). When a concentration gradient exists between the active pores and the closed pores, solute can enter the closed pores by molecular diffusion. The solute becomes temporarily entrapped, and will remain in the closed pores until the concentration gradient reverses. This matrix diffusion results in retardation, causing the bulk of the solute to move at a lower average velocity than the flowing groundwater (Feenstra et al., 1984).

As in granular geologic material, solute transport processes in peat are governed by advection, dispersion and retardation. Advection is the movement of solute owing to groundwater flow, with the centre of mass of the solute plume traveling at the average linear flow velocity ( $v$ ). For one-dimensional flow in a column,  $v$  can be estimated using

$$v = \frac{Q}{An_a} \quad (1)$$

where  $Q$  is the volumetric discharge from the column,  $A$  is the cross-sectional area of the column, and  $n_a$  is the active porosity of the peat. Pore-scale variations in velocity, tortuosity of the flow path, and heterogeneity result in the mechanical dispersion of the solute.

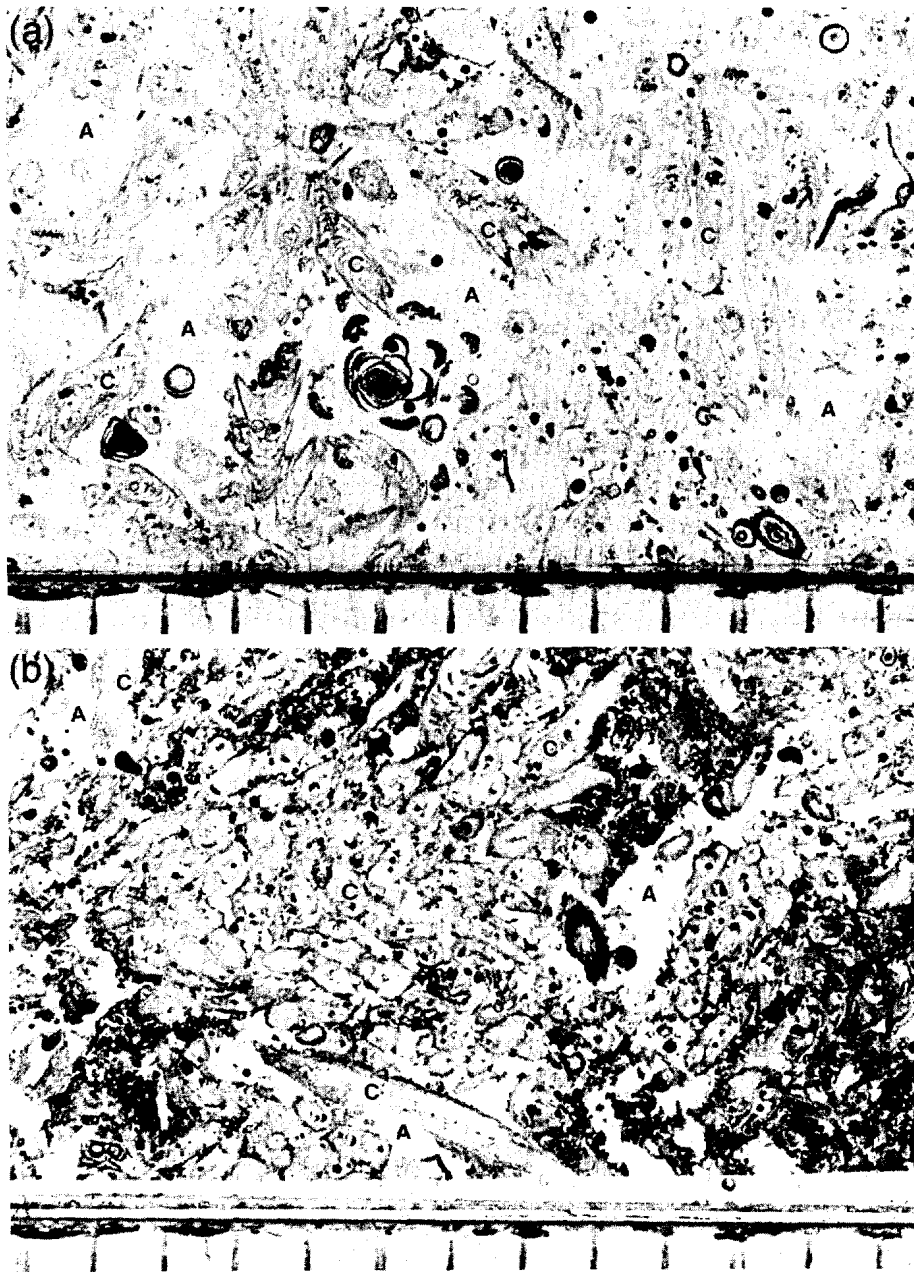


Fig. 1. Structure of peat at (a) 0.20 m depth, and (b) 0.55 m depth; showing interconnected pores which can actively transmit water (A), and dead-end and closed pores caused by plant cell remains (C).

The solute distribution resulting from one-dimensional flow in a column is described by the advection–dispersion equation, which states that

$$D_l \frac{d^2 C}{dl^2} - v \frac{dC}{dl} = \frac{dC}{dt} \quad (2)$$

where  $D_l$  is the coefficient of hydrodynamic dispersion,  $l$  is the distance along the flow path and  $t$  is time. The coefficient of hydrodynamic dispersion is defined as

$$D_l = \alpha_l v + D^* \quad (3)$$

where  $\alpha_l$  is the dispersivity of the peat in the direction of flow and  $D^*$  is the coefficient of molecular diffusion in the porous medium. If retardation occurs, then the average solute velocity will be less than the solution in which it is carried. A retardation factor ( $R$ ) can be used to quantify the effect such that

$$v_R = \frac{v}{R} \quad (4)$$

where  $v_R$  is the retarded velocity of the solute. If  $R > 1.0$ , then retardation occurs and the centre of mass or advective front of the solute will travel more slowly than the average linear flow velocity by a fraction proportional to  $R$ .

Eq. (2) is typically used for single-porosity, rigid porous media. Here, the approach mimics the influence of the immobile zone by incorporating the effects of matrix diffusion into the  $R$  factor. The single-porosity model provides a simpler approach, requiring fewer assumptions about the shape and size of the immobile zone and its parameterization. Dual-porosity models (e.g. Rasmuson, 1984; Gillham et al., 1984) are available, and can be useful in elucidating the micro-scale processes. Here, however, we have chosen the simplest model (Ogata and Banks, 1961) which can meet the objectives of the study (Anderson and Burt, 1985). Thus, for the initial boundary conditions  $C(0, t) = C_0$  and  $C(l, 0) = 0$ , the solution to Eq. (2) is the well-known Ogata–Banks (Ogata and Banks, 1961).

### 3. Methods

Three 0.80 m long undisturbed peat cores were taken from a Newfoundland blanket bog, near the location of the field-scale, natural-gradient solute transport experiment described by Hoag and Price (1995). The cores were frozen and shipped to the laboratory. The cores were frozen to prevent damage during shipping, and also to ensure that exposure to air did not result in further decomposition of the catotelm (anaerobic) peat prior to experimentation. Thirteen thin sections at 0.05 m intervals were prepared from one core by Spectrum Petrographics Inc. (South Jordan, Utah) using acetone displacement and resin impregnation. The active porosity ( $n_a$ ) was estimated using a point-count method. A  $30 \times 30$  grid with 900 points was superimposed over a magnified ( $10 \times$ ) image of each peat thin section ( $40 \times 25$  mm), and all points on the grid were

sampled. The number of points corresponding to open pores were then counted with the grid oriented parallel, diagonally and perpendicular to the long axis of the slide. Two counts were performed for each orientation, and the results were averaged. The coloured resin penetrated only the actively flowing pores, and not the closed pores and plant material, so the active pores were easily distinguishable from the matrix. While using the point-count method is subject to interpretive error, and only provides a two-dimensional representation of the pore structure, it is the most reasonable means of assessing active porosity in a highly deformable, dual-porosity medium like peat.

Another saturated core was used to estimate the dry bulk density and total porosity ( $n_t$ ) of the peat using the method described by Boelter (1976). The oven-dried weight of the peat was divided by the wet bulk volume of the sample to obtain dry bulk density. The wet bulk volume was used since peat significantly shrinks when dried. Closed/dead-end porosity ( $n_c$ ) was estimated by subtracting the active porosity from the total porosity.

The third core was cut into 0.13 m lengths and allowed to thaw until the edges of the core were nearly dry. The sections of core were then placed in a 0.12 m long, 0.16 m diameter pipe, which was filled with molten wax. Successive additions of molten wax were required to fill the space between the core and the pipe. The wax penetrated the peat by about 2 mm, ensuring a tight seal between the peat and the pipe. The column was then trimmed to a length of 0.10 m, a constant-head reservoir and bottom cap with a flexible rubber tube were affixed to the column, and all joints were sealed (Fig. 2). Following immersion in water for a period of five days, an upward gradient was

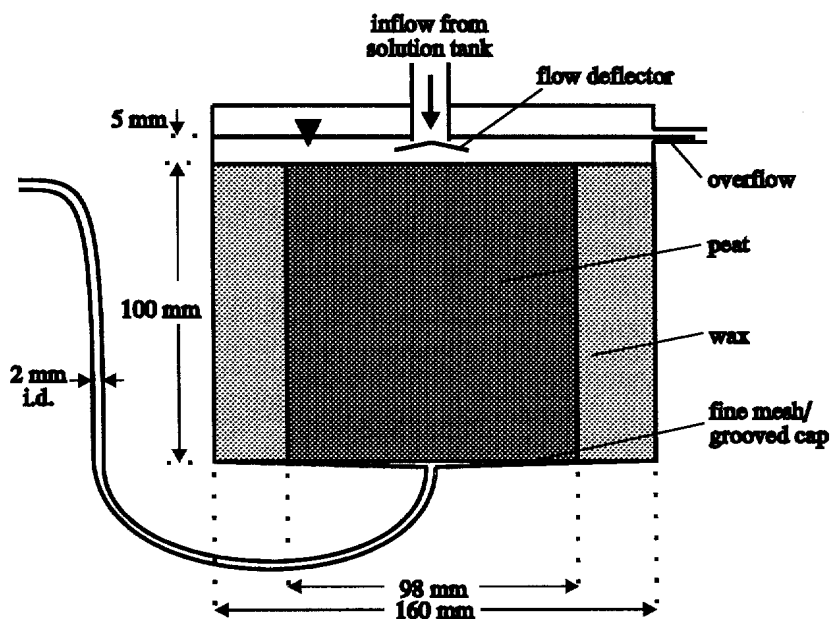


Fig. 2. Design of peat columns showing wax seal, constant-head reservoir and outflow tube used to control gradient.

established to ensure the column was completely saturated, and to displace any gas bubbles which may have formed. The source tank, distilled water and peat columns were kept together in a temperature-controlled room to maintain all components of the system at a constant temperature, to prevent degassing of the solution. The inflow tube from the source tank was placed below the level of the overflow to prevent aeration of the solution. A flow deflector was attached to the end of the tube to ensure a constant source concentration across the peat column, and a cover placed over the reservoir prevented evaporation. Depths are reported as the mid-point of the range (i.e. 0.62 m for a range of 0.57 to 0.67 m).

Flow through the column during each run was maintained at a constant rate by adjusting the hydraulic gradient with the flexible tube. The background electrical conductivity (EC) of the effluent, adjusted to 25°C, was measured prior to conducting each test. EC is directly proportional to the concentration of electrolytes in solution, and is therefore a quick and reliable way to measure solute (salt) concentration. A  $1.0 \times 10^3 \text{ mg l}^{-1}$  NaCl solution contained in a large tank was used as a tracer for all tests. Prior to each trial, the water in the reservoir was lowered until it just reached the surface of the peat. The solution was introduced as a step-input, and the discharge and EC of both the source and effluent were measured frequently during the tests. Source concentration remained constant during all tests. Following each run, the column was flushed with distilled water for several days using a downward gradient to ensure that the EC of the active- and closed-pore waters had returned to background levels. Two runs at different flow rates were conducted for each column (except at 0.50 m, where the column ceased functioning after one trial).

Solute breakthrough curves were plotted in terms of relative concentration (effluent concentration/input solution concentration or  $C/C_0$ ) versus pore volumes. Pore volumes were defined as the volume of discharge from the column divided by the estimated volume of the actively flowing pores. The retardation factor was determined by modelling the data and “backing out”  $R$ . The solution to Ogata–Banks  $\{C(l, t)\}$  was

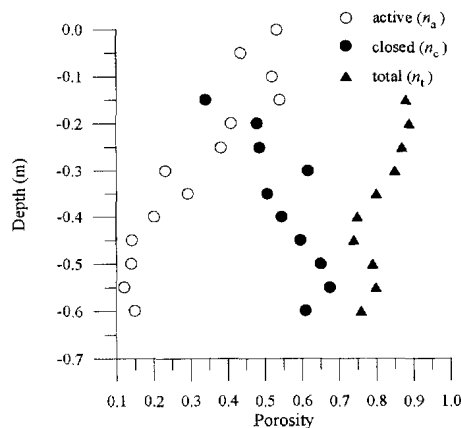


Fig. 3. Change in active porosity ( $n_a$ ), closed porosity ( $n_c$ ) and total porosity ( $n_t$ ) with depth.

Table 1

Hydraulic and physical characteristics of peat used in columns. Also shown is the change in hydraulic conductivity ( $\Delta K$ ) during the experiments for comparison purposes

Depth (m)	Initial $K$ ( $\times 10^{-5} \text{ m s}^{-1}$ )	$\Delta K$ * (%)	Porosity			$\rho_b$ ( $\text{kg m}^{-3}$ )
			Active ( $n_a$ )	Closed ( $n_c$ )	$n_a/n_c$	
0.20	1.4	48	0.37	0.50	0.74	119
0.35	0.8	45	0.24	0.58	0.41	174
0.50	0.1	-4	0.16	0.62	0.26	236
0.62	3.3	-700	0.12	0.63	0.19	240

\* A positive  $\Delta K$  indicates an increase in hydraulic conductivity, while a negative  $\Delta K$  indicates a decrease.

calculated by varying the value of  $D_l$  until a good fit to the breakthrough data was obtained.

#### 4. Results

Peat cores from the bog showed a marked change in humification and colour with depth. The upper 0.20 m of the core consisted of living and dead Sphagnum. Below 0.40 m, the peat became quite dark and relatively well-decomposed, with a more compact matrix and few preserved roots and grasses present. This depth approximates the change from acrotelm to catotelm. Analysis of the thin sections supported this distinct trend in

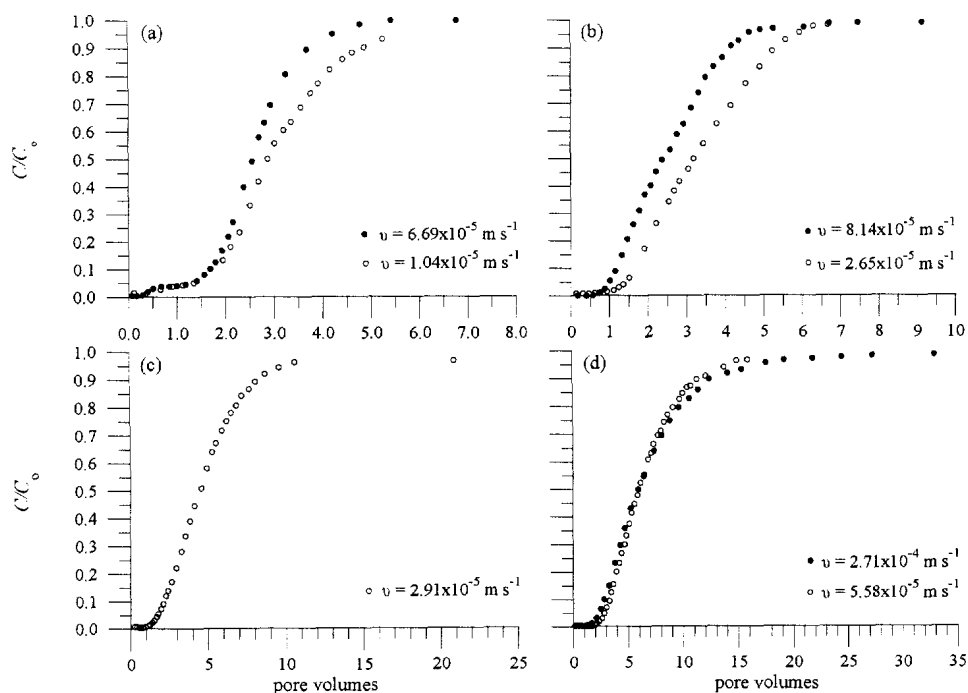


Fig. 4. Breakthrough curves for depths of (a) 0.20 m, (b) 0.35 m, (c) 0.50 m, and (d) 0.62 m. Note change in scale for pore volumes between depths.



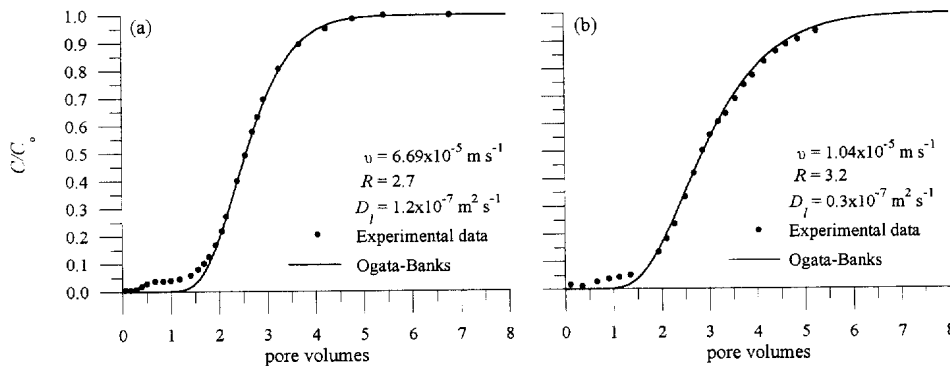


Fig. 5. Solution to Ogata–Banks for a depth of 0.20 m for (a) high velocity and (b) low velocity trials.

the peat structure, which is visible in Fig. 1. At shallower depths, the open structure of hyaline cells in Sphagnum stems and branches was evident, while at greater depths the plant remains were less distinct. The peat matrix was more compact at depth, with the plant remains tightly packed together and the open pores much smaller than at shallower depths. No preserved wood or small-scale heterogeneities were evident in the peat cores, nor were any recurrence intervals (see Chason and Siegel, 1986). In general, the active porosity of the peat decreased with depth (Fig. 3), with a corresponding increase in the proportion of plant cell remains and closed pores. This is reflected in the increased ratio of closed pores to active pores with depth (Table 1). Near the surface, the active porosity was almost 0.60, while at a depth of 0.60 m it was 0.12. Bulk density increased significantly with depth, indicating greater decomposition (Table 1). There was also a general trend of decreasing hydraulic conductivity (calculated from Darcy’s law) with depth, although the trend was not as distinct as that noted by Hoag and Price (1995), where field measurements showed a 6 to 7 order of magnitude decrease in hydraulic conductivity over the upper 0.5 m of peat. The laboratory values were generally within an order of magnitude of the field values, the exception being the column at 0.62 m, which exhibited a higher hydraulic conductivity than the other samples. The reason for this is unknown, but it may be due to natural spatial heterogeneity. Alternatively, tearing

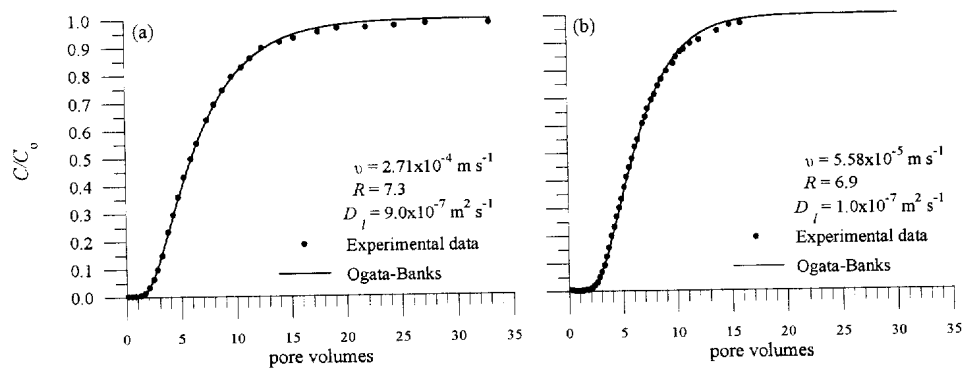


Fig. 6. Solution to Ogata–Banks for a depth of 0.62 m for (a) high velocity and (b) low velocity trials.

Table 2

Change in retardation factor ( $R$ ) and dispersion coefficient ( $D_l$ ) with depth, flow velocity ( $v$ ), dry bulk density ( $\rho_b$ ), and the ratio of actively flowing to closed pores ( $n_a/n_c$ )

Depth (m)	$R$		$D_l (\times 10^{-7} \text{ m}^2 \text{ s}^{-1})$		$\rho_b \text{ (kg m}^{-3}\text{)}$	$n_a/n_c \text{ (-)}$
	high $v$	low $v$	high $v$	low $v$		
0.20	2.7	3.2	1.2	0.3	119	0.74
0.35	2.8	3.8	4.0	1.0	174	0.41
0.50	no data	4.4	no data	0.9	236	0.26
0.62	7.3	6.9	9.0	1.0	240	0.19

along the core boundary when it was thawing may have created larger pores, although this was not evident at the ends of the core, which were inspected after the experiment was finished. Leakage or a faulty apparatus was not evident.

The breakthrough curves had very long tails, with the curves for depths greater than 0.40 m not reaching  $C/C_o = 1.0$ , even at late time (Fig. 4). Breakthrough at  $C/C_o = 0.5$  occurred well after one pore volume for each trial, and occurred at a greater number of pore volumes for greater depths. Initial breakthrough (i.e.  $C/C_o > 0$ ) followed the same trend, occurring at later time for greater depths. The solution to Ogata–Banks closely approximated the data (Figs. 5 and 6). As such, retardation was depth-dependent, with  $R$  increasing from 2.7 at a depth of 0.20 m, to more than 7.3 at a depth of 0.62 m (Table 2). This corresponds to an increase in peat matrix compaction, as illustrated by the ratio of active to closed pores and bulk density (Table 1). In addition, retardation was found to be velocity-dependent, with lower flow velocity resulting in greater retardation. At depths less than 0.40 m, changes in velocity resulted in proportionately greater changes in retardation. It is not certain if  $R$  is asymptotic since the retardation process may not be at steady state in these short columns. Longer columns would be required to verify this dispersion increased with depth in the peat profile (Table 2). The Peclet # ( $vd/D^*$ ), where  $d$  is the average particle diameter (estimated from Fig. 1), was estimated at 10.8 and 18.6 for the 20 and 50 cm depth cores, respectively.

## 5. Discussion

The decrease in the active porosity of the peat with depth is a result of increased compaction at lower levels, caused by the weight of the overlying plant material and water trapped in the closed plant cells (see Romanov, 1968), as well as decomposition of the original structure. The low active porosity at greater depths (Table 1) resulted in higher linear flow velocities from relatively low specific discharges. The trend in hydraulic conductivity followed that found in the field, with the exception of the column at a depth of 0.62 m. The relatively high hydraulic conductivity at this point is uncharacteristic of peat, and may indicate the presence of a macropore created by freezing the core or natural heterogeneity. Some preferential flowpath, as evidenced by the high hydraulic conductivity at 0.62 m, affected solute transport since the breakthrough curves in Fig. 4(d) are nearly identical, in spite of the difference in velocity.

During the initial trial, hydraulic conductivity decreased by about 25% when  $C/C_0 = 0.85$ . The total decrease upon completion of the test was nearly an order of magnitude (Table 1). This change was hysteretic, since it did not completely recover even after flushing the column for several days, reaching about 60% of its initial value. The slight increase in hydraulic conductivity at 0.20 and 0.35 m (Table 1) is consistent with the observations of others (D.I. Siegel, pers. comm.). The decrease in hydraulic conductivity noted at 0.62 m may have resulted from floc and colloid formation, as a consequence of the high  $\text{Na}^+$  concentration (Tarchitzky et al., 1993), blocking the small pores. There is a possibility this was enhanced by freezing of the core, which could rupture cells and promote the formation of flocs. However, since freezing occurs on an annual basis at least in the upper section of the core, freezing of the core in the lab is unlikely to have any added consequence.

The step increase in solute concentration underwent dispersion as evidenced in the “S” shaped elution curves (Fig. 4). Dispersion in the flow direction was dominantly mechanical, based on the Peclet numbers ( $< 10$ ) (Gillham and Cherry, 1982). Retardation by matrix diffusion is evident in the late breakthrough of the centre of mass of solute ( $C/C_0 = 0.5$ ) within all columns. With no retardation, breakthrough at  $C/C_0 = 0.5$  normally occurs at one pore volume (Bouwer, 1991). However, breakthrough at this level of concentration occurred well after one pore volume in each of the trials, indicating retardation. The estimated retardation factors ( $R$ ) show that retardation was depth-dependent.  $R$  increased with depth owing to the larger proportion of closed pores there (Table 1), which resulted in a greater capacity to trap solute. Higher velocity trials had less retardation since there was less time for molecular diffusion of the solute into closed pores, compared to lower velocities. Similarly, initial breakthrough ( $C/C_0 > 0$ ) occurred later for low velocities since there was more time for the solute at the front to diffuse into closed pores.

The data indicate that  $R$  increases as dry bulk density ( $\rho_b$ ) increases, and  $n_a/n_c$  decreases (Table 2). A greater number of observations with  $R$  values determined from longer columns (to attain a steady-state  $R$ ) is required to be certain since the data are insufficient to perform a statistical evaluation. It follows that it may be possible to use  $n_a/n_c$  and dry bulk density to give a rough estimate of  $R$ , or at least the relative magnitude of  $R$ , since the structure of peat and hence its ability to attenuate contaminants will vary from site to site. However, dry bulk density is a relatively easy parameter to determine, while  $n_a/n_c$  is more difficult to assess since it requires the preparation of thin sections to measure  $n_a$ .

The long tails of the breakthrough curves for trials below a depth of 0.40 m were a consequence of the increased proportion of plant cell remains at depth, as evidenced by the decrease of  $n_a/n_c$  with depth (Table 1). At greater depths there is a larger sink for solutes which takes considerable time to reach equilibrium with the actively flowing pores. Furthermore, the structure of the matrix in more decomposed peat may render some dead-end and closed pores less accessible to solute flowing in active pores. Consequently, some ability to attenuate contaminants remained even after dozens of pore volumes.

The results have shown that retardation delays the passage of the centre of mass of solute ( $C/C_0 = 0.5$ ). The distribution of solute concentration (dispersion) with time is

also enhanced at depth (Table 2) because of the larger sink for solute there. Since more solute was being removed from actively flowing pores, more spreading of the solute and greater dispersion occurred. The column at 0.62 m was the exception to this trend. Further tests are required to determine if this is typical behavior for solute at greater depth in peat. Density-dependent flow may have occurred within the column owing to the high source density (Schincariol and Schwartz, 1990). Gravitational instabilities possibly affected the breakthrough curves, and will be considered in future model development.

## 6. Conclusions

Peat effectively attenuated the flux of a NaCl solution through matrix diffusion into closed and dead-end pores. Compaction and humification of the deeper layers of peat produced a greater proportion of closed and dead-end pores. Consequently, both retardation and dispersion increased with depth since there was a larger sink for solute, and the breakthrough curves exhibited longer tails owing to the greater time required to attain equilibrium in solute concentration between actively flowing and closed pores. The degree to which solute was retarded and dispersed depended on flow velocity, with lower flow velocity causing greater retardation and more dispersion, since there was more time for solute to be abstracted by diffusion from actively flowing pores. The column tests indicated that there may be a relationship between dry bulk density and the retardation factor. Although the retardation factors reported here are high for a non-sorbing solute, the heterogeneity in peat properties means that its effectiveness in attenuating contaminants will depend on the depth of release, as well as the degree of decomposition and compaction.

## Acknowledgements

The authors gratefully acknowledge the support of the Natural Sciences and Engineering Research Council of Canada (JSP). David Lapen provided able assistance, and Dr. B.H. Kueper and Dr. D.I. Siegel offered useful insights into the project.

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