

Fertilizer Diffusion in Container Medium

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ADDITIONAL INDEX WORDS. bromide, leachate, hygroscopic, osmotic, modeling, protected diffusion zone, nursery

ABSTRACT. The process of fertilizer diffusion was examined using KBr and NaBr salts placed at the top of columns filled with a container medium at an initial water content of 4.0, 2.5, or 1.0 g·g⁻¹ (mass of water/mass of medium). Columns were sealed to create a protected diffusion zone (PDZ) shielding the system from water infiltration and evaporation. Bromide and water distributions were determined after 5, 10, 25, and 120 days. Using a Fickian diffusion model, effective diffusion coefficients calculated for Br⁻ in the medium at 2.5 g·g⁻¹ ranged from 2.7 to 4.6 × 10⁻⁶ cm²·s⁻¹, which is 3 to 9 times less than the diffusion coefficient in water alone. Diffusion rates increased with increasing medium water content. Differences in the hygroscopicity and solubility of KBr and NaBr affected the distribution of water and diffusion rates in the columns. Redistribution of water was driven to a significant degree by vapor-phase transport, caused by large gradients in osmotic potential, and was most apparent at low water content. At high water content, water redistribution was affected by solution density gradients in the system. This significantly complicates the mathematical modeling of the system, which renders a simple Fickian diffusion model of limited predictive value in high and low water content media.

In intensive nursery crop production systems, fertilizer often moves with applied water through and beyond the plant root zone and has the potential to contaminate ground and surface waters. The importance of protecting water quality is recognized in the horticulture industry, and many solutions have been proposed and implemented. Current practices include optimizing the timing of fertilizer application (Biernbaum, 1992; Cox, 1993; Hershey and Paul, 1982), collecting and treating runoff (Alexander, 1993), and reducing water and fertilizer use through drip irrigation and scheduling (Kabashima, 1993; Ticknor and Green, 1987). The closed insulated pallet system (CIPS) and the conserver are two systems currently being investigated to minimize leaching losses by placing fertilizer in a protected diffusion zone (PDZ) in the medium to shield it from gravitational flow of surface-applied water (Green, 1995; Green and Schneckenburger, 1992; Green et al., 1993a, 1993b; Rost, 1995). Understanding the process of diffusion from these concentrated fertilizer sources is important for incorporating a PDZ into commercial production systems to minimize environmental impact yet sustain economical plant growth.

Agricultural research on diffusion has focused on diffusion of nutrients toward plant roots in natural soils (Barber et al., 1963; Bhadoria et al., 1991; Nye and Tinker, 1977; Olsen and Kemper, 1968). Although many researchers have reported increased diffusion rates with an increase in water content (Graham-Bryce, 1963; Klute and Letey, 1958; Mehta, et al., 1995; Patil, et al., 1963; Schaff and Skogley, 1982), predicting effective diffusion rates based on soil type and water content is usually carried out experimentally or predicted using previously published literature. Most soil ion diffusion literature is concerned with dilute soil solutions diffusing through a nearly saturated medium. Some studies reported diffusion rates using solutions as high as 1.0 M (Graham-Bryce, 1963; Palmer and Blanchar, 1980).

The process of fertilizer salt dissolution and the subsequent

diffusion into relatively dry soils is a complex process that has received limited attention. Wheeting (1925) observed that, in

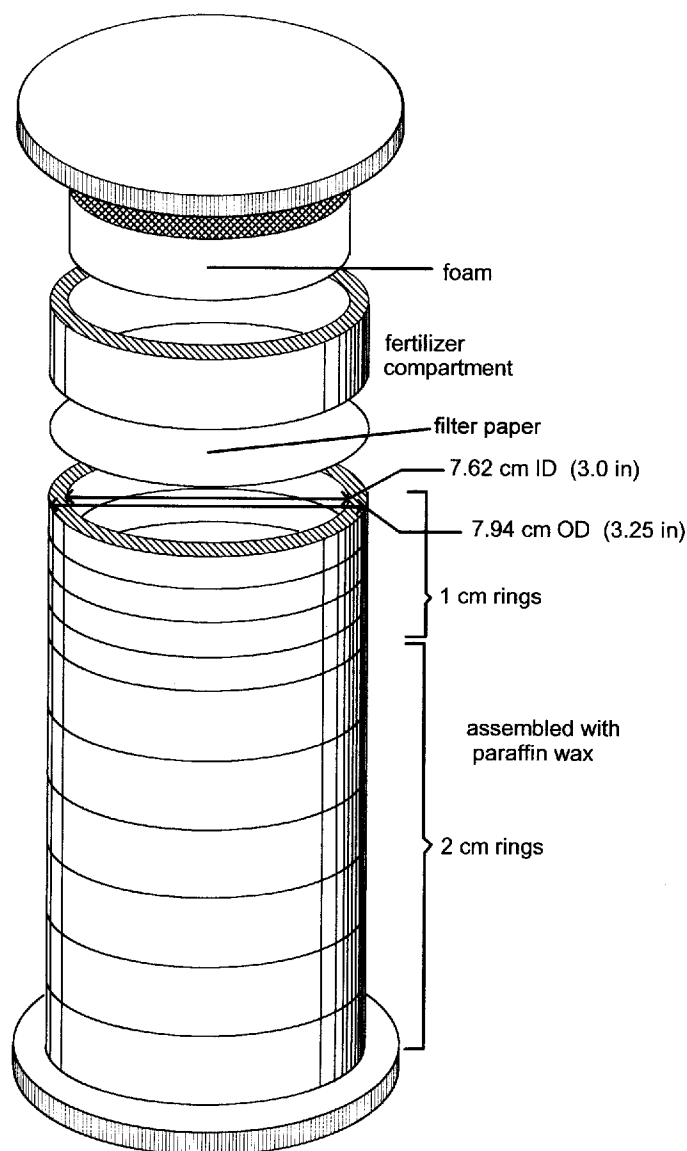


Fig. 1. Construction of soil columns for diffusion experiments.

Received for publication 5 Mar. 1996. Accepted for publication 17 Sept. 1996. We acknowledge the Agricultural Research Foundation and Brigg's Nursery, Inc., Olympia, Wash., for financial support of this research. Use of trade names does not imply endorsement of the products named or criticism of similar ones not named. The cost of publishing this paper was defrayed in part by the payment of page charges. Under postal regulations, this paper therefore must be hereby marked advertisement solely to indicate this fact.

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Table 1. Residual Br^- in the fertilizer compartment for the horizontal columns after 5, 10, 25, and 120 d diffusion time for KBr and NaBr in container medium at initial water contents of 1.0, 2.5, and 4.0 $\text{g}\cdot\text{g}^{-1}$. Values are in grams of Br^- and standard deviations of three replicates are shown in parenthesis.

Salt	Day	Initial water content					
		1.0 $\text{g}\cdot\text{g}^{-1}$		2.5 $\text{g}\cdot\text{g}^{-1}$		4.0 $\text{g}\cdot\text{g}^{-1}$	
		Mean	(SD)	Mean	(SD)	Mean	(SD)
KBr	Initial	33.5	---	33.5	---	33.5	---
	5 d	28.9	(0.2)	24.9	(1.0)	12.7	(1.0)
	10 d	27.2	(0.9)	20.5	(0.3)	4.1	(2.9)
	25 d	20.6	(0.7)	11.1	(0.8)	0.1	(0.2)
	120 d	3.0	(0.8)	0.0	(0.0)	0.0	(0.0)
NaBr	Initial	39.0	---	39.0	---	39.0	---
	5 d	36.6	(0.2)	24.7	(0.4)	11.8	(1.8)
	10 d	34.9	(0.8)	16.1	(0.2)	0.0	(0.0)
	25 d	19.1	(0.2)	0.4	(0.5)	0.0	(0.1)
	120 d	0.0	(0.0)	0.0	(0.0)	0.0	(0.0)

relatively dry soils, dissolution was accompanied by an accumulation of water in the soil immediately surrounding the fertilizer salts and a drying out of the soil farther away from the fertilizer. Several researchers have attributed this phenomenon to water vapor movement toward the salt due to the vapor pressure gradient induced by the salt (Kolaian and Ohlrogge, 1959; Lawton and Vomocil, 1954; Scotter and Raats, 1970). Parlange (1973) presented a theory to describe this process that was subsequently shown to have limited success in predicting the water redistribution in the wetter region adjacent to the salt (Scotter, 1974a). Scotter (1974b) found that the dissolution of salt into a relatively dry soil depended very strongly on the solubility and saturated solution vapor pressure of the salt used and the initial soil water content.

The foundation of the conceptual model for ion diffusion processes are Fick's first and second laws of diffusion (Tyrrell and Harris, 1984). Fick's first law describes the observed instantaneous and irreversible flow or flux of ions from regions of high ion concentration to areas of low ion concentration. Mathematically, Fick's first law in one dimension states that ion flux (J , mass/length²/time) across a plane perpendicular to the direction of flow in the x direction, is directly proportional to the mobile-phase concentration (C , mass/length³) gradient:

$$J = -D(\partial C/\partial x) \quad [1]$$

The proportionality constant (D , length²/time) is called the diffusion coefficient.

Because fertilizer ions diffuse more slowly through a wet, porous medium than in pure aqueous solutions, the diffusion coefficient is modified by factors that account for porosity, pore geometry, physical properties, and chemical interactions with the porous medium. This modified coefficient is called the effective diffusion coefficient, D_e . In this study, D_e is defined as being equal

to $D\tau_a$, where τ_a is the apparent tortuosity, which includes all factors tending to reduce the rate of diffusion in the medium, except for the volumetric water content, θ , since θ is measured easily and independently from other factors. Thus, Fick's first law as modified for a porous medium can be stated as

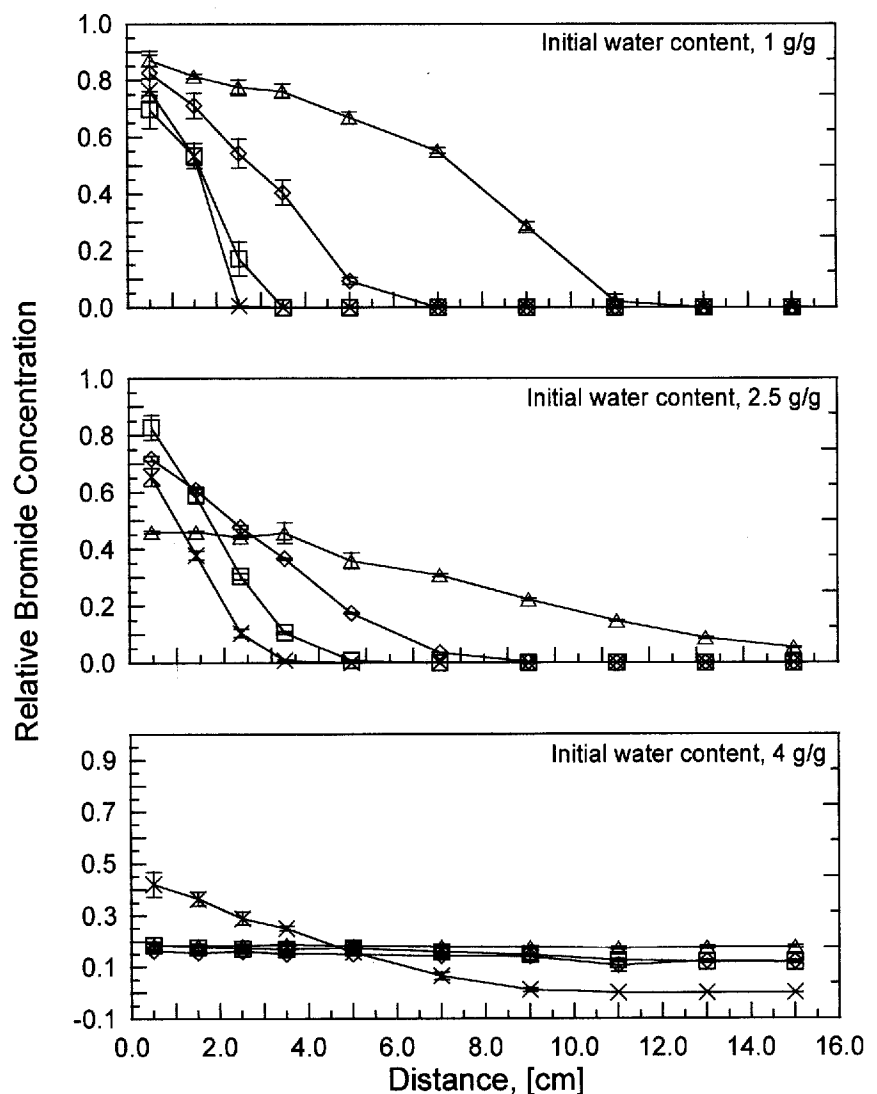


Fig. 2. Average (three replicates) relative Br^- concentrations in sections along horizontal columns with KBr as the diffusing salt after 5 (x), 10 (□), 25 (◇) and 120 d (Δ) at initial water content of 1, 2.5, and 4 $\text{g}\cdot\text{g}^{-1}$. Error bars represent ± 1 standard deviation.

$$J = -D_e \theta (\partial C / \partial x) \quad [2]$$

where C is defined as the concentration of the solute in the liquid phase.

Fick's first law is useful for analyzing steady fluxes, but, for solving time-dependent problems, Fick's second law (derived from the first law and conservation of mass principles) is more useful. Fick's second law as modified for a porous medium in one dimension is

$$(\partial C / \partial t) = D_e (\partial^2 C / \partial x^2) \quad [3]$$

where t is time. Important assumptions made in the derivation of Eq. [3] are 1) there is no interaction of the medium with the ions; 2) θ is constant at all points in space and time; and 3) the medium is sufficiently homogeneous that D_e is constant throughout the column at all points in space and time. Experiments conducted by establishing initial values and boundary conditions applicable to solutions of Fick's second law Eq. [3] form the basis of most methods to determine D_e in a porous medium.

A solution to Eq. [3] satisfying the specified boundary conditions, $C(0, t) = C_0$, and the initial conditions, $C(x, 0) = 0$, is found in Crank (1975):

$$C(x, t) = C_0 \operatorname{erfc}(x^2 / 4 D_e t) \quad [4]$$

Effective diffusion coefficients can be calculated from Eq. [4], providing the assumptions made in deriving Eq. [3] are satisfied along with additional constraints.

- 1) The boundary conditions are constant throughout the experiment (i.e., a sufficient supply of salt crystals is always available at the upper surface).
- 2) There is no convective movement of solutes and negligible water vapor movement.
- 3) The solute never reaches the far end of the column.

The objectives of this paper were to 1) identify the processes contributing to ion transport from concentrated salt sources in an unsaturated container medium inside a PDZ; 2) show that the simple Fickian diffusion model has several constraints that severely limit its ability to predict fertilizer salt diffusion; and 3) demonstrate that initial water content and properties of the diffusing salt affect the resulting solute and water distributions in a PDZ. The experimental approach was to establish specific boundary and initial conditions using KBr and NaBr in columns filled with a standard container medium at a series of initial water contents. The resulting Br⁻ diffusion was observed in vertical and horizontal columns.

Materials and Methods

A standard 1 peat : 1 vermiculite (by volume) container medium was prepared for use in all experiments. Air-dried Canadian sphagnum peat was sieved through a 9.42-mm screen to remove large particles. Vermiculite and the sieved peat were mixed and brought to water contents of 1.0, 2.5, and 4.0 g·g⁻¹ (0.17, 0.42, and 0.68 m³ of water/m³ of medium) by adding the appropriate amount of tap water and allowing equilibrium in sealed containers for a minimum of 1 week before they were used to fill the columns.

Sixteen-centimeter-long columns were constructed from four 1-cm-high and six 2-cm-high rings cut from clear acrylic plastic tubing, 7.62 cm ID × 7.94 cm OD (3-inch ID × 3.25-inch OD). The column segments were assembled with hot paraffin wax as shown in Fig. 1. The bottom of each column was capped with a flat acrylic base. The columns were filled with the container medium and packed by loosely filling the column with wet medium in 2-cm increments and tamping the medium between increments, resulting in an average dry bulk density of 0.17 g·cm⁻³ (sd = 0.013). The columns were filled to 4 cm above the top of the column and then leveled off to 16 cm. To form a fertilizer compartment on each column, a piece of filter paper (Whatman glass microfibre filter, 934-AH, 9 cm) was placed on top of the medium to separate the salt from the medium. An additional 2-cm ring was placed at the top of the column, and 50 g of oven-dried salt was placed in this ring. To ensure contact was maintained between the salt-filter-medium interface, a plastic-wrapped 2-cm-thick foam disc was fitted inside

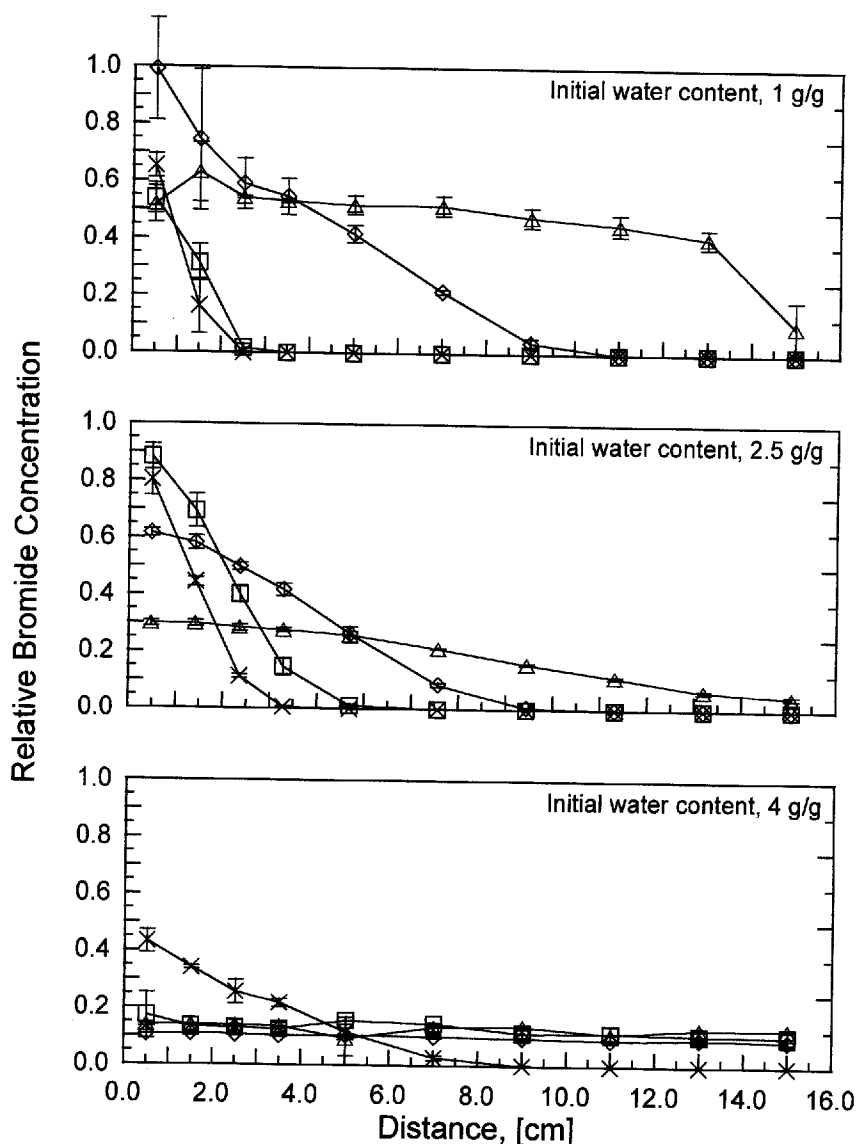


Fig. 3. Average (three replicates) relative Br⁻ concentrations in sections along horizontal columns with NaBr as the diffusing salt after 5 (x), 10 (□), 25 (◇), and 120 d (Δ) at initial water content of 1, 2.5, and 4 g·g⁻¹. Error bars represent ±1 standard deviation.

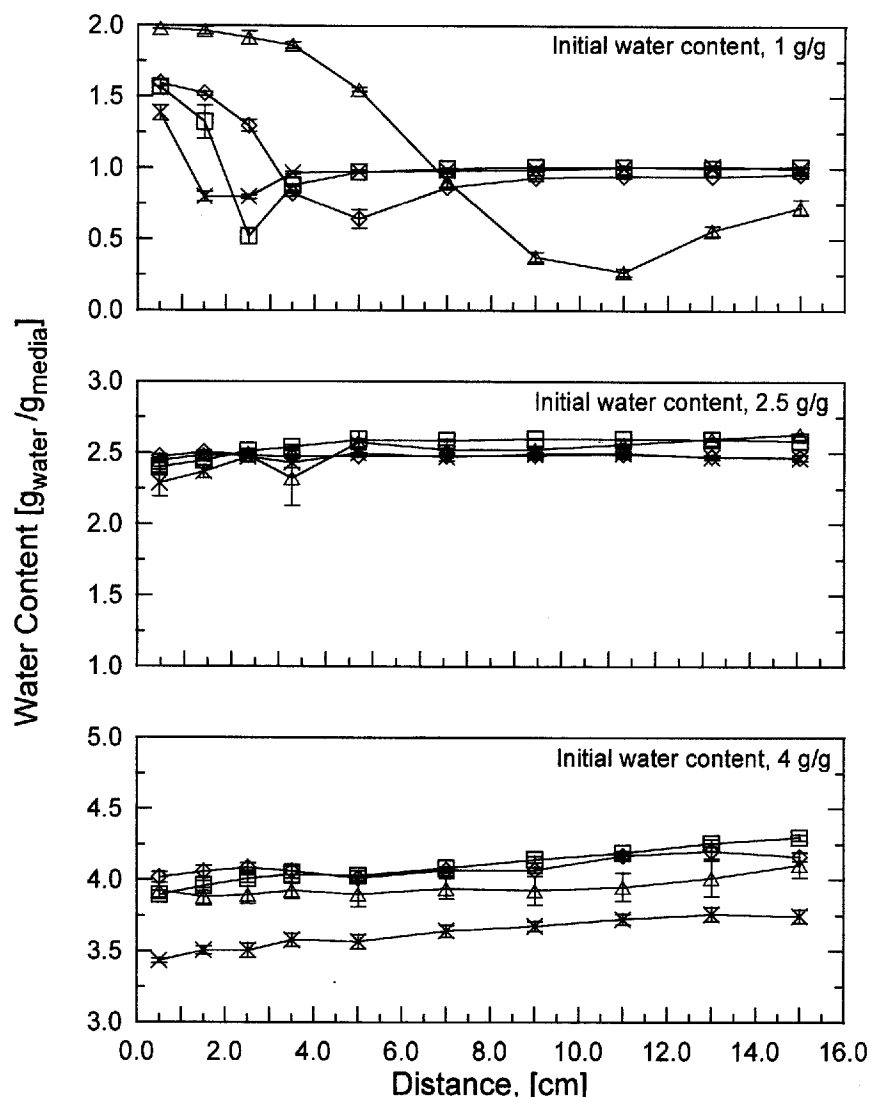


Fig. 4. Final water contents in horizontal columns with KBr as the diffusing salt after 5 (x), 10 (□), 25 (◇), and 120 d (Δ) at initial water content of 1, 2.5, and 4 g·g⁻¹. Error bars represent ±1 standard deviation.

the additional ring on top of the salt. A flat acrylic cap was placed on top of the column, and the entire assembly was sealed with hot paraffin wax and clamped together.

Horizontally oriented column experiments were carried out using columns filled with container medium at three water contents. Either KBr or NaBr, (solubility of Br⁻ at 20 °C is 394 g·L⁻¹ for KBr and 475 g·L⁻¹ for NaBr, Freier, 1976) was applied to each column, providing Br⁻ concentrations equal to the respective solubility at the salt-medium boundary. The prepared columns were laid horizontally and maintained at room temperature (≈20 °C) to be analyzed at intervals of 5, 10, 25, and 120 d. Three replications of each salt and water content were prepared for a total of 72 columns. In vertically oriented column experiments, KBr was brought into contact with the top or the bottom of the container medium at three water contents and Br⁻ was allowed to diffuse either upward or downward. Three replications of each water content and diffusion direction were prepared and analyzed after 10 d for a total of 18 columns.

After the salts had diffused into the medium for the specified time, the columns were disassembled using a wide blade to retain

the medium in individual sections and weighed immediately and set aside. Residual salt in the fertilizer compartment was recovered and oven dried at 100 °C to determine final salt weights (Table 1). Soluble Br⁻ in each section was extracted by adding 15 mL of acetone (as a wetting agent) and 250 mL of distilled water to the medium in a 500-mL Erlenmeyer flask that was shaken for a minimum of 30 min. An additional 200 mL of distilled water was added to this solution and filtered (VWR grade 415 qualitative filter paper, 20.5 cm in diameter) using a vacuum flask. Distilled water was added to obtain a total solution extract of 500 mL, of which 25 mL was saved for later Br⁻ concentration determination using a bromide ion selective electrode (Orion no. 9435). Relative Br⁻ concentrations for each section were calculated from the concentration of Br⁻ in the medium solution normalized by the maximum solubility of the salt (the pure salt boundary condition). The filtered medium was oven dried at 60 °C for 24 h to determine gravimetric water content at the time each section was disassembled. After the Br⁻ concentration in the medium was determined, the gravimetric water contents were corrected for the weight of the salt in the wet medium. Volumetric water contents were calculated from gravimetric water contents, and average bulk densities were calculated from dried medium weights.

Results and Discussion

The Fickian diffusion model was used to calculate D_e . To determine which treatments to include in the analysis, the model assumptions and boundary conditions were checked by inspecting the residual salt weights (Table 1), the Br⁻ distributions (Figs. 2 and 3), and the water content distributions (Figs. 4 and 5) for each column treatment. Only days 5, 10, and 25 for the column treatments with 2.5 g·g⁻¹ initial water content (both salts) satisfied all specified boundary conditions and most nearly the assumption of a constant water content. Effective diffusion coefficients were calculated from this subset of column treatments using a minimum residual sum of squares criterion to select the optimum value of D_e to fit Eq. [4] to the resulting Br⁻ distributions (Fig. 6). An analysis of variance of this subset showed that the differences among D_e were highly significant ($P < 0.01$) (Table 2). Further analysis showed that D_e for NaBr was significantly greater than KBr after 5 ($P < 0.05$) and 10 d ($P < 0.01$) at 2.5 g·g⁻¹. A comparison of Br⁻ distributions in Figs. 2 and 3 showed similar differences between KBr and NaBr at other water contents. For instance, in the column treatments at 1.0 g·g⁻¹ after 25 and 120 d, NaBr had moved further down the column than KBr. Inspection of the residual Br⁻ in the fertilizer compartment (Table 1) revealed that at least 50% more Br⁻ had moved into the medium with NaBr than KBr in all comparable treatments except at 1.0 g·g⁻¹ after 5 and 10 d.

These observed differences in diffusion rates may be attributed to the greater solubility of NaBr versus KBr and the effect of the different accompanying cation of each salt. For diffusion to proceed in the columns, the cation-anion pairs must move in the same direction at the same speed to maintain electroneutrality. This type of diffusion is termed salt or mutual diffusion, and the resulting diffu-

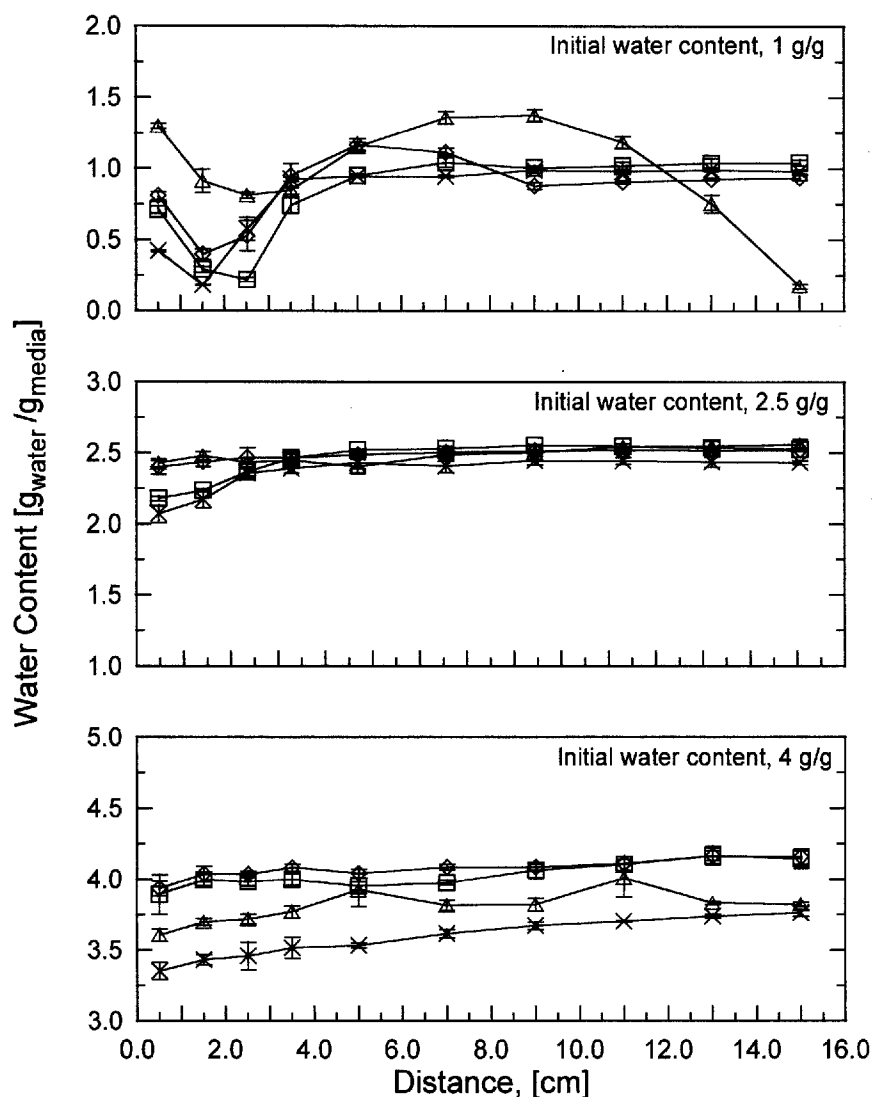


Fig. 5. Final water contents in horizontal columns with NaBr as the diffusing salt after 5 (x), 10 (□), 25 (◇) and 120 d (Δ) at initial water content of 1, 2.5, and 4 g·g⁻¹. Error bars represent ±1 standard deviation.

sion coefficient consists of a harmonically averaged ion mobility for each ion (Robinson and Stokes, 1959). The diffusion coefficient at infinite dilution for KBr ($2.0 \times 10^{-5} \text{ cm}^2 \cdot \text{s}^{-1}$) is greater than that for NaBr ($1.6 \times 10^{-5} \text{ cm}^2 \cdot \text{s}^{-1}$) in an aqueous solution but, surprisingly, the calculated D_e for NaBr is greater than that for KBr in the container medium at 2.5 g·g⁻¹ (Table 2).

Diffusion coefficients in an aqueous solution are not constant with varying solution concentrations, but range from 1.9 to 2.4 and 1.5 to $1.7 \times 10^{-5} \text{ cm}^2 \cdot \text{s}^{-1}$ for KBr and NaBr, respectively (Robinson and Stokes, 1959). The Fickian diffusion model used to calculate the effective diffusion coefficients in the columns requires that the diffusion coefficient be independent of solution concentration, or that solution concentration gradients be sufficiently small so that the diffusion coefficient can be considered to be independent of concentration. The solution concentration in the diffusion columns varies across the whole range of molalities from saturation at the

salt interface to an infinitely dilute solution at the far end. This implies that D_e in the medium will be concentration dependent and that the simple Fickian diffusion model will not sufficiently predict the Br⁻ movement in the columns. Using this reasoning, a complete model to predict ion movement in a porous medium in a PDZ would need to include a non-Fickian diffusion component with a concentration-dependent diffusion coefficient for all diffusing ions. It must be noted, however, that at all concentrations the value of D_e for KBr exceeds that of NaBr; thus, this effect cannot explain fully the observed reversal in magnitudes of D_e for NaBr and KBr in the container medium.

The horizontal experiments showed slight evidence that the effective diffusion coefficient of Br⁻ depends on the diffusion time. The mean effective diffusion coefficient for NaBr in a medium at 2.5 g·g⁻¹ increased significantly ($P < 0.05$) from 5 to 10 d (Table 2). There was no significant increase in mean D_e for KBr with time of diffusion.

There was no difference in Br⁻ distribution after 10 d in either the vertical columns with the salts placed on the top or bottom or the horizontal columns at water contents of 1.0 and 2.5 g·g⁻¹. Plots of Br⁻ distribution for all orientations were similar to those in Fig. 2 for horizontal columns after 10 d. Additionally, there was no statistical difference between D_e calculated after 10 d for KBr in medium at 2.5 g·g⁻¹ water content for the horizontal and vertical columns with salt placed at the top or bottom. Gravitational water distribution occurred in the columns at 4.0 g·g⁻¹ water content (Fig. 7). Bromide movement in vertical columns oriented with KBr at the top and bottom were similar after 10 d, whereas Br⁻ movement was significantly greater in the horizontal column. Faster movement in the horizontal column at 4.0 g·g⁻¹ water content occurred in response to changes in solution density in the medium, which have a negligible effect at a low water content. As KBr dissolves at the end of the column, an unstable density gradient develops convective flows

and the heavier, saturated solution on the top of the column flows down and toward the end of the column. It moves over the equally dense saturated solution on the bottom of the column and displaces the water, gradually moving toward the end of the column. A convective-diffusive flow is developed in this manner, causing the salt to move down the column faster than by diffusion processes alone. This process was evident during column analysis, when horizontal columns were disassembled and some of the sections were laid flat and allowed to dry, revealing higher salt concentrations in the bottom half of the section. This drop-out effect has been reported previously in the literature (Burns and Dean, 1964), but was not observed in any of our vertical columns.

Rate of movement and distribution of Br⁻ depends on the water content in the medium (Figs. 2 and 3). As expected, diffusion rates of KBr and NaBr increased with increasing moisture contents. At 4.0 g·g⁻¹ initial water content, >50% of the cross-sectional area was filled with water pathways in which diffusion can occur freely; after 10 d almost all 50 g of KBr and all the NaBr moved into the medium (Table 1). At a lower water content, much less of the cross-sectional area is available for diffusion of solutes, and

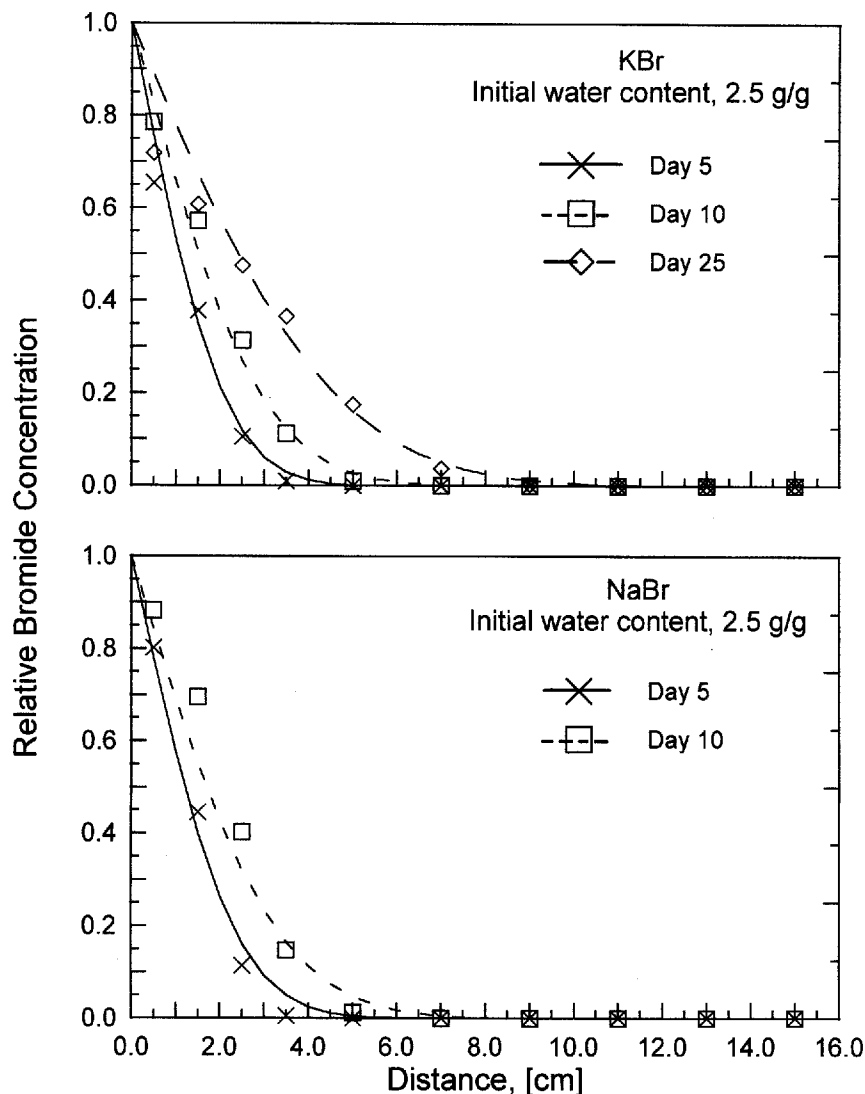


Fig. 6. Relationship between the fitted effective diffusion coefficient using the Fickian diffusion model, Eq. [4], and the selected column data.

correspondingly less salt moves into the medium.

At $1.0 \text{ g} \cdot \text{g}^{-1}$ water content, movement of water toward the salt source at the end of the medium columns is striking (Figs. 4 and 5). The same movement was evident to a lesser extent in columns with $2.5 \text{ g} \cdot \text{g}^{-1}$ water content. More water moved toward the salt compartment from the medium with NaBr than KBr in response to its hygroscopic properties. Sodium bromide is characterized as hygroscopic and forms a hydrated molecule ($\text{NaBr} \cdot 2\text{H}_2\text{O}$), whereas KBr is characterized as slightly hygroscopic and does not have a hydrated form (Weast, 1987). This hygroscopic movement of water in the NaBr column created a zone of drier medium about 2

cm from the salt compartment, forming a barrier of low water content and significantly slowing the rate of Br^- movement for NaBr in the first 10 d compared to KBr. The presence of concentrated solutions in the medium caused the osmotic potential of the solution to increase, resulting in a localized decrease in vapor pressure. Vapor pressure gradients cause water transport through the medium as it is vaporized in areas of low solute concentration and condensed in areas of high solute concentration. At $1.0 \text{ g} \cdot \text{g}^{-1}$, gradients persist in the medium because the liquid-phase transport in the dry medium is less than the vapor transport. This process has been reported to occur only in relatively dry soils (Scotter and Raats, 1970; Wheating, 1925). Although not as evident at higher moisture contents, similar osmotic pressure gradients exist, and it follows that this vapor transport process would occur in an unsaturated medium of higher water content as long as connected gas-filled paths existed in the medium. This process may be obscured at higher water contents in which liquid-phase transport is sufficient to allow redistribution of the water transported through the vapor phase.

Summary

The simple Fickian diffusion model was useful for analyzing selected columns in this study but was inadequate to predict the fertilizer ion movement in a PDZ, where it becomes necessary to model the water movement as well as the ion movement. The results of this research have important implications in predicting ion diffusion from concentrated salt sources in an unsaturated porous medium.

1) The initial quantity of salt applied and the solubility and hygroscopic properties of the salt affect the movement of water near the salt and the salt

diffusion rate.

- 2) There is no simple relationship between D_e in an unsaturated medium and the pure water diffusion coefficient for specific salts.
- 3) As expected, the ionic diffusion in the container medium increases with increasing water content.
- 4) At high water content ($4.0 \text{ g} \cdot \text{g}^{-1}$), water redistribution and solution density gradients increase gravitational flows of solution in the medium.
- 5) Significant water redistribution occurs in the medium in response to an osmotic potential established by the concentrated solution in the medium.

These features significantly complicate the mathematical modeling of the system, rendering the simple Fickian diffusion model of limited predictive value.

Table 2. Effective diffusion coefficients ($\text{cm}^2 \cdot \text{s}^{-1} \times 10^{-6}$) for Br^- in 1 peat : 1 vermiculite (by volume) medium in columns with $2.5 \text{ g} \cdot \text{g}^{-1}$ initial water content. The sample standard error (SE) of the mean effective diffusion coefficients is $0.281 \times 10^{-6} \text{ cm}^2 \cdot \text{s}^{-1}$.

Day	Salt						
	NaBr		KBr				
	Horizontal		Horizontal			Vertical (salt at top)	Vertical (salt at bottom)
5	5	10	5	10	25	10	10
Mean	3.62	4.65	2.74	3.38	2.79	3.33	3.26

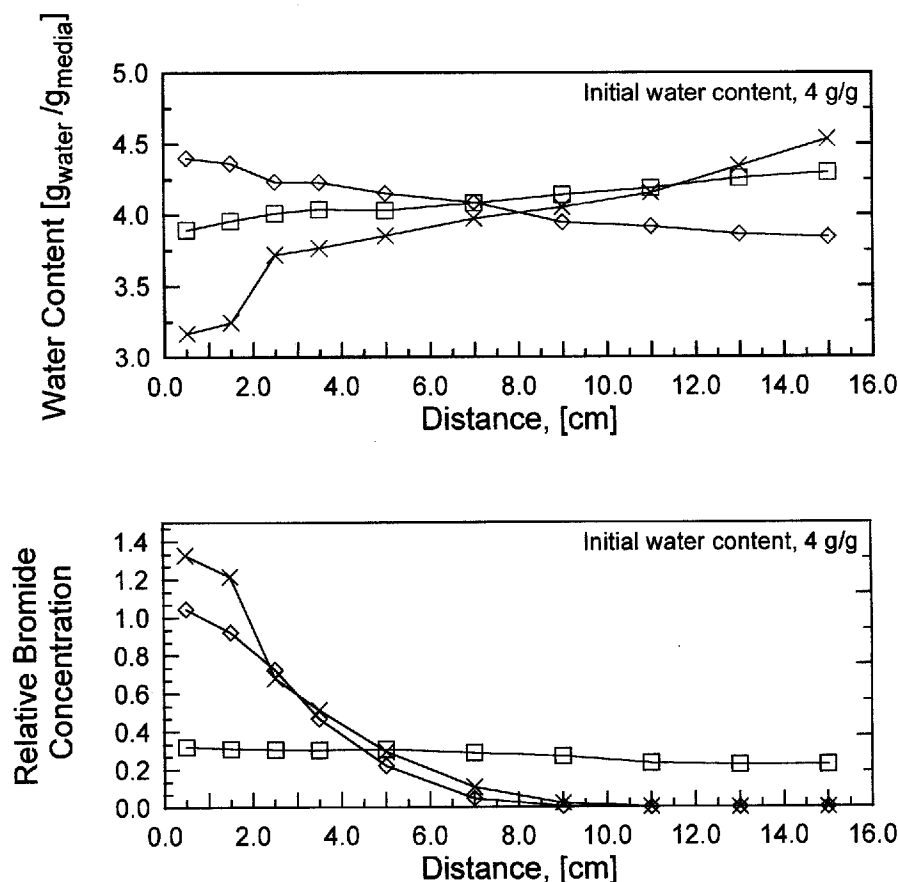


Fig. 7. Average (three replicates) relative Br^- concentrations and final water contents in vertical columns with KBr as the diffusing salt after 10 d with diffusive flux oriented up (\diamond), down (\times), and horizontally (\square).

Even though modeling the processes that control diffusion is complicated, the results of this research show that there is potential to minimize leaching losses in nursery production systems by placing fertilizer in a PDZ. The observed ion diffusion rates in the medium were always less than the predicted maximum diffusion rates expected in pure water and significantly less than would be expected by leaching. For typical moisture contents of a container medium in a conserver and CIPS, a PDZ 15 cm long is sufficient to maintain negligible fertilizer losses, because the maximum diffusion rate of Br^- salts is not expected to be greater than the maximum diffusion rate of fertilizers providing NO_3^- . Although we addressed these issues in the context of these two particular horticultural applications, these results have broader implications for other applications such as fertilizer diffusion under plastic mulches and ion movement in saline soils with limited infiltration. Application and retention of fertilizer ions within the plant root zone is an important step toward conserving water and fertilizer, protecting ground and surface water quality, and sustaining economic plant growth.

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