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# Bromide Transport through Soil Columns in the Presence of Pumice

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**ABSTRACT:** *Solute transport parameters, similar to soil's physical and chemical properties, can be affected by the presence of organic and mineral soil conditioners. In this study, the effect of different levels (0, 3, and 6 weight percent) of the inexpensive and easily accessible pumice conditioner on the parameters of bromide transport in sandy loam soil columns (diameter and height of 10 cm) was investigated. The transport parameters were estimated based on the BreakThrough Curves (BTCs) by the inverse modeling of the Convection-Dispersion Equation (CDE) and the mobile-immobile model (MIM) using the CXTFIT software. The BTCs showed that bromide transport in the sandy loam soil columns, regardless of the presence of pumice was mainly equilibrium, and the CDE was more efficient than the MIM, which is based on non-Fickian and non-equilibrium transport. The peak of the BTCs (maximum relative concentration) was lower in the treatments containing pumice and belonged to more pore volume than the controlled treatment did. This indicates a lack of preferential flow and thus, a reduction in the amount of bromide consumed in the treatments containing pumice. The increase in pumice content did not have a significant effect on the parameters of mobile water fraction ($\beta$) and mass transfer coefficient ($\omega$) in the MIM, confirming the equilibrium transport of bromide. A 3% increase in the pumice content in the soil caused an increase and a decrease of 47% in dispersivity ($\lambda$) and Peclet number (Pe), respectively. In general, it can be concluded that the use of pumice in field conditions can prevent water loss and nutrients and reduce groundwater contamination by reducing preferential paths.*

**KEYWORDS:** *Convection-dispersion equation, Dispersivity, Equilibrium transport, Mobile-immobile model, Peclet number.*

## INTRODUCTION

Considering the quantitative and qualitative limitations of water and soil resources, one of the practical methods in modern agriculture is the use of inexpensive organic and mineral conditioners to increase the soil water storage

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capacity, prevent moisture loss, increase irrigation efficiency, reduce soil erosion, improve soil structure and provide a suitable condition for seed germination. Pumice is a mineral conditioner with a non-crystalline chemical compound of aluminum silicate with a high moisture adsorption capacity. This conditioner is a volcanic rock that consists of highly vesicular rough-textured volcanic glass. Pumice can widely be used especially in coarse-textured soils like sandy soils. This rocky material is less expensive than similar conditioners like pearlite and vermiculite [1-2]. Pumice makes the soil lighter, facilitates plowing, and improves soil aeration and water storage. This conditioner, combined with the soil, improves hydraulic conductivity and reduces the negative effects of soil crusting, cracking, and waterlogging. It can be useful for long-term applications due to its physical and chemical durability [3]. After sieving, pumice can have a high water storage capacity and a low bulk density compared to soil [4].

Solute transport parameters are important properties in transport processes in soil and, similar to soil's physical and chemical properties, can be affected by several factors like soil conditioners and organic materials. Inverse modeling is one of the methods to estimate transport parameters. In this method, in addition to observed data, a mathematical model indicating the governing equations for the phenomenon as well as an optimization algorithm to minimize the objective function is needed. The proper mathematical model is fitted to the measured data based on the BreakThrough Curve (BTC). BTCs are obtained by plotting the ratio of the effluent solute concentration to the influent solute concentration versus the effluent water volume, the time, or the number of pore volumes [5-7].

In recent years, several studies have reported the effect of natural organic soil conditioners [8-11] and synthetic or mineral soil conditioners [2-4, 12-15] on hydraulic and solute transport parameters in sandy soils and other types of soils [16,17]. *Asghari et al.* [18] investigated the effects of the four organic conditioners polyacrylamide (PAM) (0.25 and 0.5 g/kg of air-dried soil), cattle manure (12.5 and 25 g/kg of air-dried soil), vermicompost (2.5 and 5 g/kg of air-dried soil) and biological sludge (1.7 and 3.4 g/kg of air-dried soil) on hydraulic and transport parameters in sandy loam soil. They concluded that the van Genuchten parameter $\alpha$ decreased significantly ($p<0.05$) at both the levels of PAM and the second levels of cow manure and vermicompost. All the conditioners reduced the dispersivity parameter ($\lambda$) in the CDE. PAM and cow manure were more effective in improving the soil's physical condition than the other conditioners at all the levels. *Lamy et al.* [19] investigated the transport of colloid and solute through sandy and porous gravel media under both saturated and unsaturated conditions in and without the presence of geotextile. They described the flow pattern using the MIM model and the HYDRUS-1D code. The results showed that geotextiles could increase flow homogeneity and decrease contaminant transport. They also shifted the BTC to the right side and lowered the peak, indicating a late and less breakthrough. The breakthrough occurred faster in the gravel column than in the sandy soil column. The HYDRUS-1D model could satisfactorily simulate the transport phenomena with a regression coefficient of close to 1. *Zhen et al.* [20] assessed the effects of Pisha sandstone on $Br^-$ and $Na^+$ transport in sandy soil. They obtained the transport parameters using the CXTFIT code. The results showed that the incorporation of Pisha sandstone into sandy soil prevented the early breakthrough by decreasing the saturated hydraulic conductivity compared to the controlled column. The MIM fitted the Br- BTCs more accurately and better than the CDE did. *Moosavi et al.* [21] evaluated the effects of cattle manure and palm residue biochars produced at different pyrolysis temperatures on saturated hydraulic conductivity and transport coefficients of chloride in sandy loam soil. Application of 0.5, 1 and 2% cattle manure and palm residue biochars increased the diffusion-dispersion parameter by 89%, 80%, 44% and 95%, 45%, 45%, respectively. Also, it increased the saturated hydraulic conductivity by 24%, 20%, and 18%, respectively. In general, the relative immobile water content and the mass exchange coefficient increased. *Yaghoubi et al.* [22] investigated the effect of the biochar layer in porous media on the breakthrough curve and the distribution profile of nitrate using the HYDRUS-1D model. The increase of the isotherm adsorption of the biochar shifted the BTC to the right side and decreased the nitrate effluent concentration, while the increase of the longitudinal dispersion coefficient shifted the BTC to the left side. The presence of an 11-cm-thick layer of biochar decreased the distributed nitrate at the bottom of the soil column by about 61%.

To our knowledge, despite numerous studies on the effect of various conditioners on soil physical and chemical properties and solute transport parameters, there is no research available on the effect of pumice on solute transport in sandy soils. The present study aims to investigate the effect of various levels of the inexpensive and easily accessible pumice conditioner on the parameters of bromide transport in a sandy loam soil column based on the BTCs obtained by the inverse modeling of the CDE and the MIM using the CXTFIT code.

## EXPERIMENTAL SECTION

### *Samples, treatments, and soil incubation*

The soils samples were collected from the research station of the Faculty of Agriculture and Natural Resources, the Islamic Azad University, Tabriz Branch with a longitude of 38° 02' 77.88" N and latitude of 46° 43' 97.07" E from the depths 0-20 cm. After being air-dried, part of the soil was passed through a 2-mm sieve to determine the physical and chemical properties, and a greater part was sieved with a 4.76-mm sieve for incubation with pumice. Also, pumice was sieved with a 2-mm sieve, and at the weight percent of 0, 3, and 6 was mixed with soil. The mixtures with a bulk density of 1.4 g/cm$^3$ were uniformly poured into pans with a height of 12 cm and a diameter of 36 cm. The samples, subjected to wetting-drying cycles between the Field Capacity (FC) and 0.5FC, were stored for 4 months at 20-25 °C (the application of wetting-drying cycles was to simulate the natural condition for the samples). Table 1 gives some physical and chemical properties of the soil and the pumice.

***Table 1: The physical and chemical properties of the soil and the pumice***

| Property | Sandy loam soil | Pumice |
|---|---|---|
| Sand (%) | 61.99 | - |
| Silt (%) | 18.19 | - |
| Clay (%) | 19.88 | - |
| Bulk density (g/cm$^3$) | 1.45 | 1.1 |
| Porosity | 0.406 | - |
| Mean weight diameter (mm) | 0.32 | - |
| pH | 7.71 | 9.25 |
| EC (ds/m) | 0.157 | 0.135 |
| Cation exchange capacity (%) | 2.43 | - |
| Calcium carbonate equivalent (%) | 14.1 | 0.138 |
| Organic carbon (%) | 0.30 | - |
| Nitrogen (N) (%) | - | 0.06 |

### *$CaBr_2$ injection into soil columns*

In order to obtain the BTCs, the PVC soil columns with a height and a diameter of 10 cm were prepared with three different pumice contents: 1) no pumice (the controlled treatment), 2) 3% pumice, and 3) 6% pumice. *Kamra* and *Lennartz* [23] stated that a laboratory soil column with a height of about 10 cm could satisfactorily simulate field conditions. The bottoms of the soil columns were fixed inside a plastic funnel on a sponge and a wire mesh to support the weight of the soil column and prevent the soil particles from being washed from the bottom. The pores of the sponge and the mesh were coarse enough not to restrict the water flow in the soil columns. To prevent preferential flow along the sidewalls of the columns, paraffin was applied to the inner walls. The breakthrough experiment was carried out in saturated conditions and under a water head of 3 cm. The soil columns were saturated from the bottom with a $Ca(NO_3)_2$ solution of 0.01 molar as the background solution. A Mariotte bottle was used to establish a constant water head (Fig. 1). After $CaBr_2$ injection started, the effluents with a volume of 0.1 pore volume were collected at different times, and their bromide concentrations were determined using a pH-meter (Crison, Spain) equipped with a bromide selector electrode. After the complete injection of $CaBr_2$, the steady-state saturated flow of the background solution was re-established. The Darcy flux was measured by recording and weighing the effluent samples [24]. The experiment continued until the bromide concentration in the effluent was almost zero. The measured concentrations (C), by dividing by the initial concentration (C0), were converted to relative concentrations (C/C0). Then the BTCS was plotted as C/C0 versus time (t) or the number of pore volumes (dimensionless Pv). Each pore volume (Vp) is defined as the ratio of the total pore volume occupied with the fluid to the total volume of the soil and is calculated as follows:

$$V_p = \theta_s \times V_t \quad , \quad Pv = V_e / V_p \tag{1}$$

Where, $V_t$ is the total volume of the soil, $\theta_s$ is the saturated volumetric water content (cm$^3$/cm$^3$), and $V_e$ is the cumulative volume of effluent (cm$^3$) from the moment of $CaBr_2$ injection.

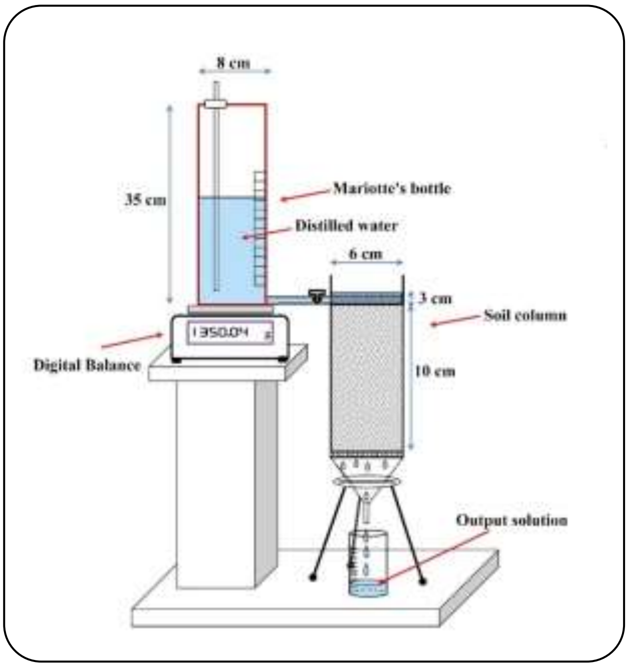

*Fig. 1: The soil column with a constant water head*

The parabolic CDE for the one-dimensional steady-state transport of an inert or non-reactive solute is described as follows [25]:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial t^2} - V\frac{\partial C}{\partial x} \tag{2}$$

Where C is the solute concentration (M/L$^3$), D is the hydrodynamic dispersion coefficient (L$^2$/T), and V is the pore water velocity (L/T). x (L) and t (T) are distance and time, respectively.

The MIM consists of the CDE to describe the transport in the mobile region and an equation to describe the solute transfer between the two mobile and immobile regions. The latter defines solute transport as proportional to the concentration difference between the two regions. The one-dimensional MIM for the steady-state transport of an inert and non-reactive solute is as follows [26- 27]:

$$\theta_m\frac{\partial C_m}{\partial t} + \theta_{im}\frac{\partial C_{im}}{\partial t} = \theta_m D_m\frac{\partial^2 C_m}{\partial x^2} - \theta_m V_m\frac{\partial C_m}{\partial x} \tag{3a}$$

$$\theta_{im}\frac{\partial C_{im}}{\partial t} = \omega\left(C_m - C_{im}\right) \tag{3b}$$

Where, $\theta_m$ (-) and $\theta_{im}$ (-) are the volumetric water content in the mobile and immobile regions, respectively. $\theta_m + \theta_{im} = \theta$ is the total volumetric water content and is equal to the porosity in a saturated medium. $C_m$ (M/L$^3$) and $C_{im}$ (M/L$^3$) is the solute concentration in the mobile and immobile regions, respectively. $D_m$ (L$^2$/T) is the hydrodynamic dispersion coefficient in the mobile region, $V_m$ (L/T) is the pore water velocity in the mobile region, and $\theta_m V_m$ (L/T) is the flow velocity or Darcy flux in the mobile region. ω (1/T) is the mass transfer coefficient between the two regions.

### *Estimation of the transport parameters*

An inverse modeling method was used to determine the transport parameters. For this purpose, in addition to the observed data, a mathematical model indicating the governing equations and an optimization algorithm to minimize the objective function needed. The CXTFIT code is capable of inverse modeling by using the Levenberg-Marquardt optimization method and is based on the analytical solution of the CDE and MIM [28]. This code was used to estimate the parameters D and V of the CDE, and the parameters Vm, Dm, $\beta = \theta_m/\theta$ and, ω of the MIM. The objective functions for the CDE and the MIM are described as Eqs. (4) and (5), respectively [27]:

$$\chi^2(V, D) = \sum_{i=1}^{n}\left[C_i - C_e(x, t_i; V, D)\right]^2 \tag{4}$$

$$\chi^2(V_m, D_m, \beta, \omega) = \sum_{i=1}^{n}\left[C_i - C_e(x, t_i; V_m, D_m \beta, \omega)\right]^2 \tag{5}$$

Where, $C_i$ is the observed concentration. $C_e(x, t_i; V, D)$ and $C_e(x, t_i; V_m, D_m, \beta, \omega)$ are the estimated concentrations at the distance *x* and the time *t* in the CDE and the MIM, respectively, and *n* is the total number of observations.

The performance of the models was evaluated by two frequently used statistical indices of determination coefficient ($R^2$), and Root Mean Square Error (RMSE) [27]:

$$R^2 = 1 - \frac{\sum_{i=1}^{n}\left(C_i - C_e\right)^2}{\sum_{i=1}^{n}\left(C_i - \bar{C}_i\right)^2} \tag{6}$$

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(C_i - C_e\right)^2} \tag{7}$$

Where $Ce$ is the estimated concentration, $Ci$ is the observed concentration, $\bar{C_i}$ represent the mean values of $Ci$, and *n* is the total number of observations.

## RESULTS AND DISCUSSION

### *The observed and fitted bromide BTCs*

The observed and fitted bromide BTCs obtained by the CDE and the MIM for the three sandy loam columns with no pumice (the controlled treatment), 3% pumice, and 6% pumice are shown in Fig. 2. All the BTCs are an average of three replications. According to Fig. 2, the peak of the BTCs is higher, and the area under the curve is larger in the controlled treatment than in the others. This could be because of the presence of preferential flow, more bromide consumption, and more bromide transport through deep pores and cracks in the soil with no pumice. Li and Ghodrati [29] have attributed the lower peak in the soils containing pumice to the absence of preferential flow. *Asghari et al.* [18], and *Ali* and *Abdul-Hussein* [30] have also reported similar results.

The number of pore volumes in the peaks and the tails of the BTCs have increased with increasing the pumice content. This could be attributed to the dispersion from the fine pores (inter-granular pores) to the coarse pores, leading to a late breakthrough and a long tail [18, 31]. Shahmohammadi-*Kalalagh* and *Taran* [32] have reached a similar conclusion. In the treatments containing pumice, compared to the controlled treatment (no pumice), the breakthrough time increased, the preferential flow decreased and, as a result, the bromide consumption decreased. Therefore, it is expected that in field conditions, the use of pumice can prevent water loss and nutrients (such as nitrate) and reduce groundwater contamination by reducing the preferential paths. However, further research on field conditions is needed in order to make a more accurate statement. As shown in Fig. 2, the CDE better fitted the observed data than the MIM in all three soil types. This indicates that the CDE was more effective than the MIM, suggesting that the bromide transport in the sandy loam soil columns was mainly in equilibrium. The statistical criteria of the coefficient of determination ($R^2$) and root mean square error (RSME) in Table 2 confirm this. In general, it is believed that the CDE is valid when the solute transport in soil follows Fick's law. According to Fick's law, the dispersion rate is linearly proportional to the concentration gradient. It appears that this condition is met in sandy soils and the equilibrium model is more practical [5, 32].

### *Estimation of the transport parameters*

The bromide transport parameters in the soil columns with no pumice, 3% pumice, and 6% pumice estimated with the CDE and the MIM are given in Table 2. The parameters ω and β can be considered as the indicators of equilibrium or non-equilibrium transport. The constraints of $0 < \beta < 1$ and $0 < \omega \leq 100$, are implicitly defined in CXTFIT. When $\beta \rightarrow 0$, there is some fraction of immobile water, and therefore a non-equilibrium condition exists. High ω values correspond to fast sorption/diffusion, and low ω values correspond to rate-limited sorption/diffusion. When $\omega \rightarrow 100$, transport is regarded to be at the equilibrium condition [28; 32-33]. The values of $\omega$ and $\beta$ in Table 2 show the equilibrium transport in the columns with and without pumice. Therefore, the CDE (developed based on equilibrium transport) is expected to have higher efficiency than the MIM (non-equilibrium). Fig. 1, along with the values of $R^2$ and RMSE in Table 2 confirm this. According to Table 2, the increase in pumice did not have a significant effect on ω and β in the MIM. It can be concluded that immobile water did not have an important role in the transport of bromide, and preferential flow was probably the main process. *Asghari et al.* [18] have reported a similar result. However, the use of 1.5% petroleum in gypsum soil by *Ali* and *Abdul-Hussein* [30], and Pisha sandstone in sandy soil by *Zhen et al.* [20] resulted in significant changes in ω and β values, indicating the non-equilibrium physical transport.

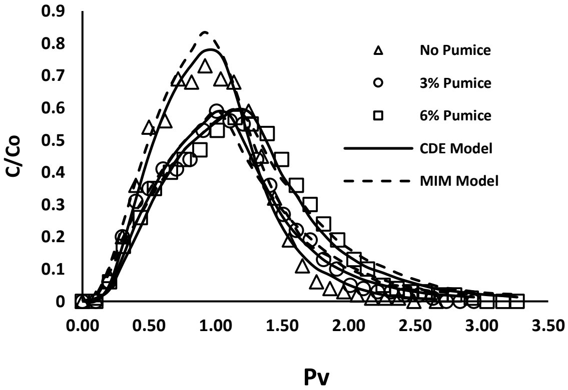

***Fig. 2: The observed and fitted bromide BTCs in the soil columns with and without pumice obtained by the CDE and MIM models.***

Dispersivity (λ=D/V) is an empirical constant and depends on various factors such as the soil texture, the degree of soil heterogeneity, the water flux in soil, the soil moisture content, the scale of the porous medium, etc. [5-6] The value of λ is often less in coarse-textured and homogenous soils than fine-textured and heterogeneous soils [18, 20, 34].

In the present study, the increase of pumice increased the value of λ, which is consistent with the results of *Asghari et al.* [13], *Asghari et al.* [18], and *Zhen et al.* [20]. Probably, the increase in pumice content disturbed the soil structure and caused heterogeneity in the soil, resulting in increased dispersivity and hydrodynamic dispersion. On the other hand, according to *Jury* and *Horton* [35] and *Asghari et al.* [18], the less the BTC is extended and the higher the peak is, the smaller the hydrodynamic dispersion and the dispersivity will be. Therefore, one of the possible reasons for the low value of λ in the controlled treatment is the high peak of the bromide BTC compared to those in the treatments containing pumice (Fig. 2). In other words, the increase of λ in the treatments containing pumice can be attributed to the low peaks and the short tails of the BTCs. Similar results have been reported by *Ali* and *Abdul-Hussein* [30] and *Asghari et al.* [18].

A direct and power relation was obtained between the pumice content and the dispersivity ($\lambda_{CDE} = 3.31P^{0.041}$, $R^2 = 0.9981$, P: Pumice, $R^2$: determination coefficient), so an increase of 3% in the pumice content increased the dispersivity by 46.9% while the twofold increase (6%) in the pumice content increased the dispersivity only by about 3% (from 46.9 to 50%). The effect of pumice content on $\lambda_{MIM}$ was almost similar to its effect on $\lambda_{CDE}$ similar to the results of *Asghari et al.* [14] and *Zhen et al.* [20] (Eq. (8)):

$$\lambda_{MIM} = 14.257\lambda_{CDE} - 23.63\ ,\ R^2 = 0.9896 \tag{8}$$

Peclet number ($Pe = VL/D$), defined as the ratio of convective transport to hydrodynamic dispersion (the sum of mechanical dispersion and molecular diffusion) (Table 2), is one of the important factors influencing the BTC. This number shows asymmetry and skewness or kurtosis of the BTC. The low value of the Peclet number indicates more dispersion and more asymmetry. For large values of Peclet number, the BTC becomes more symmetrical and sigmoid [5, 32]. In the present study, the decrease in the values of Peclet number with increasing the pumice content (Table 2) as well as the kurtosis of the BTCs of the treatments containing pumice compared to that of the controlled treatment confirmed this issue. Similar to the dispersivity (λ), the change in the values of the Peclet number with the pumice content is power, but the inverse relation ($Pe_{CDE} = 2.98P^{-0.045}$, $R^2 = 0.9939$, P: Pumice). Accordingly, an increase of 3% in the pumice content has increased the Peclet number by 47% while an increase of 6% in the pumice content has increased the Peclet number only by 51%. In other words, the increase in pumice content has reduced mixing and thus, the displacement rate. As a result, the predominant process in bromide transport has shifted from a convective phenomenon in the controlled column to a dispersive phenomenon in the columns containing pumice. In fact, convective transport had an important role in bromide transport in the controlled column while with the increase of pumice content, the effect of dispersivity became more important. As already mentioned, this could be attributed to the soil heterogeneity due to the presence of pumice, and hence, the increase of dispersion coefficient (D) and the decrease of Peclet number. *Zhen et al.* [20] have achieved a similar result. To further investigate the relationship between the values obtained by fitting the CDE and MIM models, a linear regression analysis was carried out. According to the results, there was a positive correlation between the values obtained using the two models (Eq. (9)).

$$Pe_{MIM} = 1.08Pe_{CDE} - 2.64\ \ ,\ \ R^2 = 0.9904 \tag{9}$$

## CONCLUSIONS

The effect of different levels of pumice conditioner on the bromide transport parameters in sandy loam soil columns was investigated. In order to achieve the BTCs, incubated soil was mixed with 0, 3, and 6 weight percent of pumice. Then, the $CaBr_2$ solution was injected into the soil columns in three replications. The transport parameters were estimated based on the BTCs and by the inverse modeling of the CDE and the MIM. The pumice content had a direct relation with dispersivity and an inverse relation with the Peclet number in both the CDE and the MIM. As the pumice content increased, the displacement rate decreased, and the dominant process in the transport shifted from convection in the column with no pumice to dispersion in the columns with pumice. The pumice treatment had an insignificant effect on the parameters β and ω in the MIM, indicating an equilibrium transport. The use of pumice in field conditions can prevent water loss and nutrients and reduce groundwater contamination by reducing the preferential paths. In the present study, only the effect of the application of three low levels of pumice on the hydraulic parameters of the inert and non-reactive solute (bromide) has been evaluated

*Table 2: The estimated bromide transport parameters in the soil columns with and without pumice obtained by the CDE and MIM.*

| Treatment (soil type) | CDE | | | | MIM | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | $\lambda$ (cm) | Pe (-) | $R^2$ | RMSE | $\lambda$ (cm) | Pe (-) | $\beta$ (-) | $\omega$ (1/min) | $R^2$ | RMSE |
| No pumice (control) | 1.81 | 5.53 | 0.9771 | 0.033 | 3 | 3.33 | 0.99 | 100 | 0.9514 | 0.068 |
| 3% pumice | 3.41 | 2.93 | 0.9761 | 0.040 | 26.43 | 0.38 | 0.88 | 100 | 0.9572 | 0.051 |
| 6% pumice | 3.62 | 2.76 | 0.9687 | 0.048 | 26.67 | 0.36 | 0.85 | 100 | 0.9552 | 0.054 |

on a laboratory scale. However, more research in field conditions with various levels of pumice content, reactive solute, and soil texture are needed to make a clearer statement on this issue.

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