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Transport of selenium and its modeling through one dimensional saturated soil columns

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The selenium, an essential nutrient and potentially toxic element to human and animal health, is widely used in agriculture and medicine. Soil is the transitional zone through which selenium entered into groundwater. Therefore, the transport of selenium through porous soil is of great environment concern, which is not well understood. The transport of selenite in loessial soil columns was studied using a miscible displacement technique under various flow rates and influent concentrations in this paper. A mathematical model based on one-dimensional convection-dispersion equation was formulated to describe the selenium transport. The breakthrough curves with and without tracers were fitted by using the computer program "studio of analytical models". The results showed that the method without tracer was more accurate than the method with the tracer. Studio of analytical models is a very flexible tool for approximate analysis of one dimensional solute transport problems in soil. The method of parameter estimation using convection-dispersion equation accurately defined the dispersion coefficient and retardation factor from the breakthrough curves and modeled selenite transport through soil. These results suggest that deterministic equilibrium convection-dispersion equation can accurately simulate the breakthrough curves of Se transport in saturated soil and the selection of tracer ions should be cautious when studying the transport of different chemicals.

Key words: Loessial soil selenite transport, flow rate, influent concentration, convection-dispersion equation, studio of analytical models.

INTRODUCTION

Solute transport in porous media is a complex physical and chemical process and has been studied extensively for 70 to 80 years (Laryean et al., 1982; Kool et al, 1989 and Davidson, 1989; Mansell et al., 1990). The soil is an important porous media through which solute transport into groundwater. In certain environmental conditions, contaminants in soils will be transported along with water, which affects the quality of surface water and groundwater and cause severe environmental problems. Therefore, the transport of solute through soils has attracted most attractions from scientist at different fields. The trace element selenium (Se) is both an essential

nutrient and potentially toxic element to human and animal health. The range of healthy Se intake is rather narrow (Navarro-Alarcon and Lopez-Martinez, 2000) and high Se in environment can cause toxic problems to biota (Rodriguez et al., 2005). The Se has been widely used in agriculture and medicine. The Se in soils determines its concentration in grains and drinking water (Wang and Chen, 2003). Additionally, electronic waste and other waste containing Se increase the risk of soil and water to Se pollutant.

The Se is most likely to be present as selenate or selenite in soils (Adriano, 2001). Agricultural irrigation can cause the transport of Se and make Se concentrated at locations where drainage water is disposed (Tanji et al., 1992; Zhao et al., 2007) and rain is also an important reason of Se transport, which poses potential contaminations to soil and groundwater. And the studies

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Table 1. Conditions of the soil column displacement experiment.

Column (mm × mm)	Inlet SeO_3^{2-} concentration (mg/L)	Waterhead height (cm)	Bulk density (g/cm³)	Average pore-water velocity (cm/h)
100 × 40	0.5	5	1.31	4.36
	1	5		4.37
	2	5		4.88
	4	5		4.66
	1	2		3.59
	1	4		4.23
	1	6		4.53
	1	8		5.63

on Se fate and its behavior in soil water systems have been carried out since Se contamination in soil and water causes serious damage to wildlife (Frankenberger and Benson, 1994; Frankenberger and Engberg, 1998). The transformations and transport of Se were investigated under a number of different conditions by laboratory and field experiment (Deverel and Fujii, 1988; Fujii et al., 1988; Deverel and Milliard, 1988; Mirbagheri et al., 2008). The transport modeling of Se has been reported by several researches (Fio et al., 1990; Alemi et al., 1991). In these models, the effort was to study the transport of Se in one dimension steady-state conditions with small residence times. Tayfur et al. (2010) used a validated 2-D finite element model for water and salts transport in a saturated-unsaturated zone to consider Se transformations and transport. However, convection dispersion equation (CDE) (Nielsen and Biggar, 1960), which was widely used in simulating solute transport (Unold et al., 2009; Fonseca et al., 2009; Koestel et al., 2009; Kumar et al., 2010; Mei et al., 2010; Jomeh and Hoorfar, 2010), was rarely used to simulate Se transport in soils.

So in this paper, selenite (SeO_3^{2-}) transport behaviors in loessial soil columns under different experimental conditions were analyzed by using CDE to see if this model could be use to fit Se transport data. Many analytical solutions have been derived for solute transport equations and these analytical solutions are now widely used for analyzing solute transport under steady-state conditions (Simunek, 2007). Most of studies on solute transport used CXTFIT2.1 (Toride et al., 1999), which are DOS-based codes developed by the U. S. Salinity Laboratory (USSL) for data processing. The studio of analytical models package (STANMOD) (Simunek et al., 2004) is a software suite that integrates a number of widely used analytical solutions developed by the USSL under a common, consistent, easy-to-use Windows graphical user interface (Feinstein and Guo, 2004). It is a very flexible tool for approximate analysis of one- two-, or multidimensional solute transport problems in soil and groundwater (Simunek et al., 2008). So STANMOD was used instead of CXTFIT to estimate the transport parameters of Se in soil.

The objectives of this study were to model Se transport in saturated soil columns by CDE. And the fitting methods with and without tracer were conducted to compare the accuracy of parameter estimation.

MATERIALS AND METHODS

Site and sampling

The soils used in this study were collected from Liudaogou watershed (110°21′, 38°47′), which locates in the wind-water erosion region of the northern Loess Plateau, China. The soil is a sandy loessial soil, corresponding to Loessi-Orthic primosols according to Chinese and FAO/ISRIC/ISSS Soil Taxonomy. The vegetation is dominated by *Artemisia desertorum*. Sampling plots were randomly chosen and surface (0-20 cm) soil samples (about 1 kg) were collected at each point in a typical slope. Soil samples were mixed thoroughly, air dried and sifted through a 2 mm plastic sieve. Basic properties of soil samples were: pH 8.19, O.M. (organic matter) 8.95 g/kg, CEC (cation exchange capacity) 26.68 cmol/kg, clay 1.96%, silt 46.71% and sand 51.34%.

Soil column transport experiments

A miscible displacement technique was used to examine the transport of Na_2SeO_3. Soil samples were loaded into polymethyl methacrylate (PMMA) pipe by the stratified filling method and the bottom of the PMMA pipe was fixed with a perforated PMMA plate. The soil column was immersed in distilled water for 24 h to saturate the entire soil column. A Mariotte bottle was used to control flow rate and the first solution in Matiotte bottle was distilled water to stabilize the soil. When the average pore-water velocity reached steady state, flow was switched to a prepared Na_2SeO_3 solution (with tracer NaCl solution). At the same time, we collected the effluent in 20 ml volumetric flasks and marked the corresponding elution time. The experiments were stopped when the concentration of the effluent was the same as the initial influent solution (indicating saturation of soil with the test solution). Silver nitrate titration was used to measure chloride ion content and selenite ion content was measured by AFS930 atomic fluorescence spectrometer. The different experimental conditions are listed in Table 1.

Application model

The software STANMOD was used to obtain the parameters of

Table 2. Fitted parameters of BTC of chloride in loessial soil with equilibrium CDE.

Fixed data	v(cm/h)	D(cm^2/h)	R	Determination coefficient	RSS
R = 1.000	4.451	3.764	-	0.9982	0.0126
V = 4.460	-	3.781	1.004	0.9982	0.0126

v, Average pore-water velocity; D, dispersion coefficient; R, retardation factor; RSS, residual sum of squares.
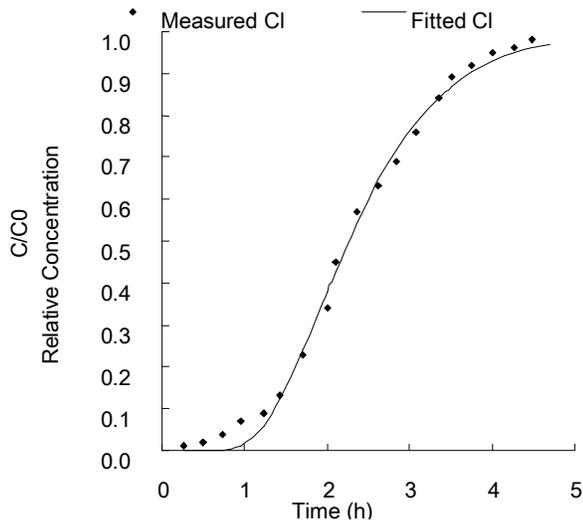

Figure 1. The breakthrough curves (BTCs) of chloride in loessial soil. The x-axis was relative time, and the y-axis was relative concentration. Smooth line was obtained by fitted data and splattering was obtained by measured data.

SeO_3^{2-} transport through soil columns. A modified and updated version of the CXTFIT code of Toride et al. (1999) that is included in STANMOD was used to solve the inverse problem by fitting experimental results to mathematical solutions of theoretical transport models based on the one dimensional convection-dispersion equation (CDE). The program may also be used to solve the direct or forward problem to determine concentrations as a function of time and position. The software is based on the Levenberg-Marquardt algorithm. First, the break-through curve (BTC) is obtained using the miscible displacement technique. The retardation factor (R) is estimated by solving the inverse problem of the convection-dispersion equation (CDE). The convection-dispersion equation is a mathematical expression to study the migration and transformation of pollutants in soil and quantitative relationship between environmental factors (Toride et al., 1999). The convection-dispersion equation for one-dimensional transport of reactive solutes subject to adsorption, first-order degradation, and zero-order production, in a homogeneous soil and the dimensionless mathematical expression is written as:

$$R\frac{\partial C_r}{\partial T} = \frac{1}{P}\frac{\partial^2 C_r}{\partial Z^2} - \frac{\partial C_r}{\partial Z} - \mu^E C_r + \gamma^E(Z) \tag{1}$$

Where C_r is the reduced volume-averaged solute concentration, P is the Peclet number, μ^E is a first-order decay coefficient (= $L\mu/v$), γ^E is a zero-order production coefficient for equilibrium transport (= $L\gamma/vc_0$), Z is and T are the dimensionless space and time variables, respectively (T = vt/L, Z = x/L), R is retardation factor (= $1 + \rho k/\theta$): Θ

is the volumetric water content (-), ρ is the soil bulk density (kg/L or g/cm^3), k is the partitioning coefficient (L/kg). Equation 2 is the initial value and boundary value condition of Equation 1:

$$\begin{cases} c = 0 \ , \ s = 0 \ 0 \le x \le L \ , \ t = 0 \\ vc - D\partial c/\partial x = vc_0 \ , \ x = 0 \ , \ t \le t_1 \\ vc - D\partial c/\partial x = 0 \ , \ x = 0, t > t_1 \\ \partial c/\partial x = 0 \ , \ x = L \ , \ t > 0 \end{cases} \tag{2}$$

Where c is the volume-averaged or resident concentration of the liquid phase (M/L^3), D is dispersion coefficient (L^2/T), v is average pore-water velocity (cm/h), x is distance (L) and T is time (T).

RESULTS

Tracer breakthrough curve fitting

The chloride transport parameters, based on deterministic equilibrium (CDE), were estimated with STANMOD software (Table 2). Chloride was used as the tracer because it is non-reactive. First, D and v were estimated assuming that R = 1, then D and R were estimated when v (measured value) was fixed using the Mariotte bottle. The data obtained using the two methods were almost equal but the fitted data were slightly better when v was fixed than when R was fixed. Residual sum of squares (RSS) was approximately 0 and the determination coefficient was approximately 1. The fitting results were more accurate when RSS was smaller. The determination coefficient, meaning the degree of proximity between fitted BTCs and measured BTCs, was approximately 1, indicating that the fitting results were acceptable. So the results could be used to fit BTCs of selenite (and determine the transport parameters). The BTC of chloride transport in loessial soil was performed to analyze whether chloride is a suitable tracer (Figure 1).

Results showed that the measured curve and fitted curve overlapped, and that the relative concentration of the peak was approximately 1, underscoring the suitability of chloride.

Selenite breakthrough curve fitting

The methods with and without tracer were used to fit the BTCs of selenite. The value of R was estimated by STANMOD software when the v of selenite and the D of the tracer were fixed (the method with the tracer). The R

Table 3. Fitted parameters of selenite with different influent concentrations.

C (mg/L)	V(cm/h)	D(cm^2/h)	R	Determination coefficient	RSS
0.5		3.764[*]	4.744	0.904	0.230
		0.624	4.886	0.996	0.011
1.0		3.764[*]	4.473	0.910	0.307
		0.769	4.685	0.998	0.006
	4.49[*]				
2.0		3.764[*]	4.287	0.962	0.127
		1.122	4.432	0.997	0.009
4.0		3.764[*]	4.192	0.969	0.078
		1.475	4.271	0.987	0.038

C, Concentration of Na$_2$SeO$_3$; v, average pore-water velocity; D, dispersion coefficient; R, retardation factor; RSS, residual sum of squares; *, Fixed data of the method with the tracer.

Table 4. Fitted parameters of Selenite with different average pore-water velocity.

C (mg/L)	V (cm/h)	D(cm^2/h)	R	Determination coefficient	RSS
	3.59[*]	3.764[*]	4.268	0.864	0.393
		0.522	4.514	0.995	0.016
	4.23[*]	3.764[*]	4.184	0.896	0.246
		0.634	4.398	0.990	0.023
1.0[*]	4.53[*]	3.764[*]	4.119	0.904	0.348
		0.533	4.356	0.996	0.012
	5.02[*]	3.764[*]	4.028	0.920	0.217
		0.884	4.236	0.977	0.053

C, Concentration of Na$_2$SeO$_3$; v, average pore-water velocity; D, dispersion coefficient; R, retardation factor; RSS, residual sum of squares; *, fixed data of the method with the tracer.

and D values were estimated under the condition of fixed v (the method without tracer). Parameters of selenite transport were estimated using both methods with different influent concentrations (Table 3). Results showed that D and RSS by the method with the tracer were higher than those obtained by the method without tracer, but the R and determination coefficient were contrary. The D values as determined by the method without tracer increased along with influent concentrations, while R decreased whether the tracer was considered or not when fitting the BTCs. Overall, fitting results by the method without tracer was more reliable than the method with the tracer. The parameters of selenite transport were estimated using both methods with different average pore-water velocities (Table 4). Results showed that the values of D and RSS by the method with the tracer were higher than those calculated by the method without tracer, but again the R and determination coefficient was contrary.

The value of D by the method without tracer increased along with v except when v was 4.53 cm/h and R decreased by both two methods as v increased, with coefficients of variation of 2.447 and 2.623%.

The BTCs of selenite

We investigated the effects of different initial selenite concentrations on BTCs in loessial soil columns (Figure 2). Results showed that the maximum relative concentration for all BTCs was approximately 1, but the times when BTCs of selenite reached maximum C/Co were later than those of the tracer. The curve clearly shifted to the left (shorter delay) as the concentration increased and the curve inflection point (the time when the solute began to penetrate the soils column) occurred earlier when the concentration was higher. The time to the final penetration also decreased with increased
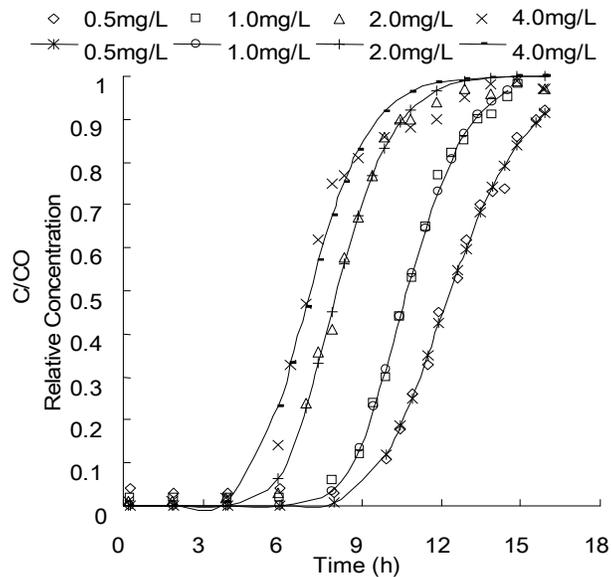
Figure 2. The Breakthrough Curves (BTCs) of selenite with different influent concentrations in loessial soil. The x-axis was relative time, and the y-axis was relative concentration. The curves are obtained in different influent concentrations. Smooth line was obtained by fitted data and splattering was obtained by measured data.
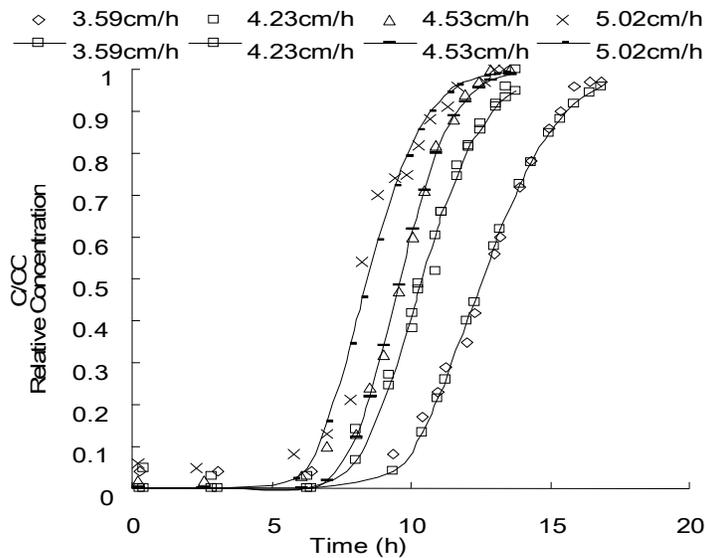

Figure 3. The breakthrough curves (BTCs) of selenite in different average pore-water velocity in loessial soil. The x-axis was relative time, and the y-axis was relative concentration. Curves are obtained with different average pore-water velocity. Smooth line was obtained by fitted data and splattering was obtained by measured data.

concentration. The value of D increased with longer delays, while R values decreased with longer delays. The BTCs of selenite under different average pore-water velocities in loessial soil columns were also determined (Figure 3). The curves shifted to the left and the time to the final penetration decreased with increasing average

pore-water velocity. The duration to the inflection point also decreased with velocity (5.02< 4.23< 4.53< 3.59 cm/h), with corresponding Ds of 0.884, 0.634, 0.553 and 0.522, respectively.

The exception to this trend was that time required for the curve inflection point was shorter when v was 4.23 than that when v was 4.53. When v was 4.23, the curve was also more flat. With the rightward shifts in the curve (longer time to inflection), the corresponding R value decreased.

DISCUSSION

An important method for estimating the retardation factor (R) is to solve the inverse problem of the convection-dispersion equation (CDE) using the miscible displacement technique. According to the formula $R = 1 + \rho k/\theta$, R can also be obtained by batch equilibrium experiments that yield the adsorption coefficient K of adsorption isotherms. Many studies obtained the R by batch equilibrium experiments and then the R was used to estimate other parameters by the miscible displacement technique (column experiment) Yolcubal and Akyol, 2008; Li, 2006; Lu et al., 2010). There are also many studies obtain transport parameters only by the miscible displacement technique (Dong et al., 2005; Huang et al., 2009; Szenknect et al. 2008; Owabor et al., 2009). Hu et al. (2005) found that the adsorption coefficient K calculated by batch equilibrium experiment was unable to fully reflect the interaction between solute and soil particles, the true leaching process, and the vertical migration process. Fonseca et al. (2009) fond batch tests tended to underestimate transport parameters. So in this paper the miscible displacement technique was used to obtain the transport parameters. Khan et al. (2010) found the model fit was much better for Freundlich isotherm because of low adsorptive capacity of the soil. The retardation factor (R) of solute in the same soil was a relatively fixed value according to formula $R = 1 + \rho k/\theta$.

Although the retardation factor using the miscible displacement technique changed when average pore-water velocity and influent concentrations changed, the coefficient of variation (5.37%) was relatively small. Thus R can be accurately estimated by repetitive trials as long as variations in pore-water velocity and influent concentrations are minimized. In sum, our results demonstrated that the method of estimating retardation factors using miscible displacement technique was accurate and applicable. Liu and Xu (2005) investigated the retarding factors of copper transport in soils of different textures. They found that the estimations by using the batch equilibrium technique was more accurate than using the miscible displacement technique. However, they investigated retardation factor from simulations of heavy metal transport in sandy loamy soil,

and found that the flow equilibration method (miscible displacement technique) was equally precise (Liu and Xu, 2005). Miscible displacement technique (column experiment) was widely used in studying the transport of all kinds of pollutants in recent years (Traugott et al., 2007; (Scheytt et al., 2007; Palagyi et al., 2009; Persson et al., 2008). Wall et al. (2008) investigated transport and attenuation of microbial tracers and effluent microorganisms in saturated pumice sand aquifer material using batch study and column study, but batch study only was used to compare the partitioning coefficient (k) of different microorganisms. Therefore, it is generally feasible to estimate R by miscible displacement technique.

Non-reactive ions are usually used as tracers (chloride or bromide). The parameters of tracers transport are used to estimate the basic conditions porous media through which other reactive ions transported (Guo et al., 2009; Zhang et al., 2008; Traugott et al., 2007; Scheytt et al., 2007; Wall et al., 2008; Persson et al., 2008; De Wilde et al. 2010). However, many studies on solute transport have been performed without tracers (Deng et al., 2005; Wang et al., 2007; Han and Zhang, 2009; Palagyi et al. 2010; Khan et al. 2010). In this study, the methods with and without tracer were compared for estimating transport parameters of selenite. The results showed that fitting data without tracer was more accurate. This might because the analytical solutions for the forward one-dimensional CDE problem require prior knowledge of the tracer D value to model the BTC. Using the inverse problem method, however, R and D are estimated by the BTC so no tracer is feasible and accurate. Another reason maybe that selenite ion is anion. Chon et al. (2007) investigated the transport and removal of chromate without tracer, but Yolcubal and Akyol (2008) used Cl⁻ as tracer to study the adsorption and transport of arsenate in carbonate-rich soils. Therefore, the selection of tracer ions should be cautious when studying the transport of different chemicals.

The BTCs can reveal the parameters of solute transport in soil columns. The value of R was smaller when the curve was closer to the y-axis and when the curve was steeper. As the time to the curves inflection point became shorter, D increased. Chen et al. (2006) found that the experimental leaching curve which R was larger was a little bit sharper than the simulating curve. Liu and Xu (2005) investigated the R of copper movement in soils, and they found that average pore-water velocity was the main factor affecting the time to the curves inflection point. The average pore-water velocity was an important factor affecting the dispersion coefficient (D). The results were consistent with results reported by Kim et al. (2006) and chen et al. (2007). The influent concentration was another important factor influencing the dispersion coefficient (D). The results were consistent with findings by Cui et al. (2009). But Yin et al. (2010) fond decreases in ionic strength enhanced

the release of both colloids and Pb in the soils.

Deng et al. (2005) found that different concentrations of nitrate had no obvious influence on the breakthrough curves of nitrate vertical transport. The reason may be that influent concentrations had little influence on the BTCs, but the influence increased when estimating parameters by BTCs.

Conclusions

In this study, deterministic equilibrium CDE was extended to simulate Se transport in saturated soil columns. The model can accurately simulate the BTCs of Se transport in saturated soil. The parameters estimated by both the method with the tracer and the method without tracer are all reliable, but it is worth noticed that the results by the method without tracer are more accurate. So we can obtain parameters of Se by solving the inverse problem and the parameters can be used to predict the instantaneous concentration of Se in different depth of soil column. It also needs to be pointed out that the STANMOD is a very flexible tool for approximate analysis of one dimensional solute transport problems in soil. However it is worth noting that the results obtained by deterministic equilibrium CDE are laboratory simulation test, which can not completely represent the real situation of field.

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