

AMMONIUM TRANSPORT EXPERIMENTS IN SOIL COLUMNS: ESTIMATING TRANSPORT PARAMETERS

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Received: 14 September 2016; Accepted for publication: 7 April 2017

ABSTRACT

Soluble contaminant transport into groundwater aquifers is one of the current practical problems occurring not only in Vietnam but also in many countries around the world. In this paper, according to the main objective of estimating the ammonium (NH_4^+) transport parameters in soil and groundwater aquifer of Con Son Island - District Con Dao, experiments with conservative tracers (sodium chlorides) and ammonium chlorides solution had been carried out on the Con Son soil columns. The transport parameters are estimated under supporting of software Hydrus 1D using the inverse parameter estimation method. Results of the experiments showed that dispersion coefficient, distribution coefficient and mass transfer coefficient characterized the ammonium transport process in the Con Son soil and aquifers are compatible with mechanical and physical properties of Con Son soil matrix and porous media.

Keywords: ammonium transport, groundwater pollution, Hydrus 1D.

1. INTRODUCTION

Inorganic nitrogen in the form of ammonia (NH_3) and the ammonium ion (NH_4^+) is recognized as one of the most common groundwater contaminants arising from waste disposal activities, fertilizer use and contaminated land [1]. As a relatively mobile ion, NH_4^+ accordingly would have the ability to spread rapidly and widely in the aquifers and contaminate groundwater when it reaches into groundwater aquifers, especially under supporting of rain and the favorable hydro-geological conditions such as the sandy aquifers, shallow aquifers... The impacts of NH_4^+ on groundwater resources in particular or water resources in general are causing significant consequences to environment and ecosystems. In drinking water supplies, NH_4^+ can reduce disinfection efficiency, lead to nitrite formation, and cause taste and odour problems [2]. NH_4^+ itself is not directly toxic to humans and animals, but when taking place in a living organism, its conversion to nitrite NO_2^- can damage cells and also morph into molecules that cause cancer and various diseases such as methemoglobinemia (or baby blue syndrome). Therefore, NH_4^+ is

considered as one of the important criteria used to assess the level of groundwater pollution, especially the aquifers pollution beneath the landfills.

Being a distant islands with limited surface water due to the distance from the mainland, groundwater exploitation and utilization is the best choice of residents in Con Son Island, Con Dao District, Ba Ria - Vung Tau Province. However, the increasing waste from domestic and tourist activities, especially from the waste-landfills have led to an urgent alarm in term of the extremely high probability of transporting pollution from the surface to Pleistocen aquifer, the major contain of groundwater for exploitation and supplying in Con Dao Island. Considering as a special landfill, cemeteries may potentially cause pollution of groundwater due to the contaminants derived from human corpses found as dissolved and gaseous organic compounds and dissolved nitrogenous forms (particularly ammoniacal nitrogen) may migrate into the underlying aquifers [3]. The degradation of human corpses normally takes 10-12 years and it is estimated that over half of the contaminant load leaches within the first years and continue to infiltrate through the soil to the groundwater aquifers in the next years. The time taken to flush out contaminants from the burial to groundwater aquifers is directly related to the soil conditions, the effective rainfall and burial practice (e.g. depth of burial and coffin construction) [4].

Many models have been developed to simulate and predict the fate and transport of contaminants such as ammonium transport. The most famous models are based on convection-dispersion equations and consider such mechanisms as convection, dispersion, sorption, and degradation. The transport of solutes has been widely studied under field and laboratory conditions in soil columns. The sorption and degradation of ammonium are included in the studies [5, 6].

The main objectives of this study were to simulate ammonium transport in Con Son soil columns with the HYDRUS-1D model under different aqueous ammonium concentrations and flow rates, to evaluate ammonium transport parameters in soil columns, including the dispersion coefficient, distribution coefficient and mass transfer coefficient for ammonium. The data used in the case study were collected from soil columns.

2. THEORETICAL CONSIDERATIONS

Generally solute transport in soil was performed with three mechanisms: mass flow, molecular diffusion and hydrodynamic dispersion. Considering the effect of these three processes in a non-steady state condition, the vertical 1D transport of a conservative tracer (nonvolatile and non-degradable) in a porous media with constant volumetric water content is described by the convection–dispersion equation as [7]:

$$R \frac{\partial C(z,t)}{\partial t} = \frac{-v}{\theta} \times \frac{\partial C(z,t)}{\partial z} + D \times \frac{\partial^2 C(z,t)}{\partial z^2} \quad (1)$$

where R is the retardation factor, $C(z,t)$ is the solute concentration in the liquid phase (g cm^{-3}), z is depth (cm), t is time (s), v is the Darcy's velocity (cm s^{-1}), θ is the volumetric water content ($\text{cm}^3 \text{cm}^{-3}$) and D is the hydrodynamic dispersion coefficient ($\text{cm}^2 \text{s}^{-1}$). The dispersion coefficient D is given by:

$$D = \frac{v}{\theta} \lambda$$

where λ is the longitudinal dispersivity (cm).

In order to better simulate solutes transport through porous media, it's necessary to take into account physical and/or chemical non-equilibrium processes [8]. The physical non-equilibrium model assumes that the adsorption always occurs in equilibrium, but the soil water is divided into two parts, mobile and an immobile. The chemical non-equilibrium model (CNEM) corresponds to a kinetic behaviour, because the adsorption process takes time to reach equilibrium between the concentration in the liquid and solid phases. Two types of adsorption sites are assumed to co-exist: type I sites are assumed to be in instantaneous equilibrium with the concentration in the liquid phase, while type II sites have time-dependent kinetic adsorption [9], [10].

Assuming that the solute degradation is negligible, the transport equation for the two sites non-equilibrium model becomes:

$$R \frac{\partial C(z,t)}{\partial t} = \frac{-v}{\theta} \frac{\partial C(z,t)}{\partial z} + D \frac{\partial^2 C(z,t)}{\partial z^2} - \frac{\rho}{\theta} \alpha [(1 - F)K_d C] \quad (2)$$

where F is the fraction of all sites, K_d is the linear adsorption coefficient or distribution coefficient ($\text{cm}^3 \text{g}^{-1}$) and α is the first-order rate coefficient or mass transfer coefficient (s^{-1}).

In this study, Hydrus 1D code was used to solve the equation (2) for ammonium transport in Con Son soil columns. Hydrus 1D code has been developed by Simunek et al. [11] in U.S. Salinity Laboratory and involves the numerical solution of Richard's equation for water flow, the convection–dispersion equation for evaluating solute transport and heat movement through the soil. Equations are solved by finite element method. This model can simulate solute transport in saturated and unsaturated conditions and estimates soil parameters using inverse solution.

3. MATERIAL AND METHODS

3.1. Soil characterization

The soil samples were collected in the Con Son Valley, about 1.8 km far from Con Dao cemetery. Con Son soil samples were taken from depths of up to 75.3 m in the vicinity of Quang Trung Lake in August 2016 (Fig. 1).

Con Son soil characteristics were analyzed and released by Southern Institute of Water Resources Research, Vietnam in August 2016. The soil is sandy with medium diameters: 11.7 % of $1 \div 0.5$ mm, 43.5 % of $0.5 \div 0.25$ mm and with small diameters: 44.3 % of $0.25 \div 0.1$ mm, 0.5 % of $0.1 \div 0.05$ mm. The total organic carbon (TOC) is low 0.36 % which indicated that this soil is poor in organic matter. The initial ammonium concentration of soil 0.15 mg / 100 g.



Figure 1. Soil sample location.

3.2. Column experiments

The laboratory experiments are carried out in Con Son soil columns. The columns which has 9 cm diameter and 70 cm height were operated in the upflow mode (Fig. 2). Soil filled in these columns were compacted homogeneously until reaching a soil bulk density of 1.5 g cm^{-3} , which is equal to the field density. The soil mass used for each single column was about 7.3 kg. At the boundaries of the column, 5 cm of gravel was set in order to ensure the water drainage. Each column was run from the lower boundary using pump AP3500 Lifetech with tap water, conservative tracer and the influent containing ammonium.

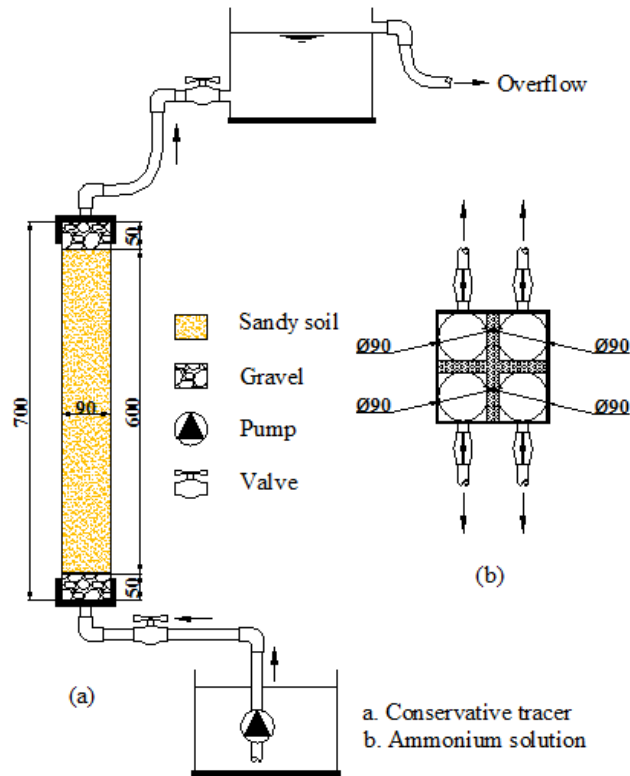


Figure 2. The experimental setup. (a) Setup Profile; (b) Setup Plan.

At the beginning of the experiments, the tap water was injected at a very low flow rate for 24 h in order to have a fully saturated porous media. Afterwards, conservative tracer, which is the Chlorides anions (Cl^-) was supplied at a concentration of 5.8 g L^{-1} of sodium chloride (NaCl) in order to determine the longitudinal dispersivity (λ) and the hydrodynamic dispersion coefficient (D) of Con Son soil matrix. These tracer assays were stopped when the measured Cl^- concentrations at the outlet of the column became equal to the injected one. Cl^- concentrations were determined by two methods: (1) using a calibrated specific electrode (Windaus LF538) and (2) by the titration solution AgNO_3 0.0141 N.

At the end of each conservative tracer experiment, the soil in the column was leached with tap water in order to remove the residual salinity caused by the tracer. This step was followed by the ammonium adsorption experiments consisting in outlet ammonium concentrations progress measure through the analysis of 100 mL aqueous solutions sampled at different times by the colorimetric method. The experiments were ended when the outlet ammonium concentration became equal to the inlet one indicating that all adsorption sites were saturated. During all the

experiments, the pH and temperature were maintained constant at 7.8 and 30 ± 2 °C, respectively.

3.3. Ammonium experimental conditions

Condition 1: The change of aqueous ammonium concentrations

Six separate columns with respectively 4.84, 9.65, 19.56, 36.54, 79.82 and 102.88 mg L⁻¹ of ammonium concentrations were used to assess the influent ammonium concentration effect on Con Son soil adsorption capacity. These values are inside the ammonium concentration range of urban wastewater discharge by Rach Ba -Vung Tau wastewater treatment plant [15] and Ho Chi Minh city wastewater treatment plants such as Go Cat, Phuoc Hiep, Dong Thanh [16]. A flow rate of 20 mL min⁻¹ was maintained for each experiment, corresponding to a Darcy's velocity of 4.53 m day⁻¹ and a contact time of 10 hours.

The ammonium adsorption isotherm onto Con Son soil was then determined by fitting three common models (Langmuir, Freundlich and linear) to the curves relating the ammonium adsorbed mass to the inlet aqueous concentration. The adsorption process is best described by the isotherm with the highest correlation coefficient [12].

Condition 2: The change of flow rates

Three separate columns were used to assess the effect of the influent flow rate on Con Son soil adsorption capacity. The used flow rates were fixed to 10, 20 and 35 mL min⁻¹, corresponding to Darcy's velocities of 2.26, 4.53 and 6.80 m day⁻¹, respectively. This range was chosen based on the results of determination of Darcy's velocities at different locations in Con Son Valley [17]. The inlet ammonium concentration was maintained constant at 1002.88 mg L⁻¹.

3.4. Calculation of ammonium adsorbed mass

The adsorbed ammonium mass (M_s) was calculated through the integration of the relative concentration versus the relative volume collected at the outlet of the column. This integration has been approximated using the trapeze method [5]:

$$M_s = C_0 \int_0^{V_{tot}} \frac{C}{C_0} dv = \frac{C_0}{2} \sum_{i=0}^{i=n} (C_i + C_{i+1})(V_{i+1} - V_i) \quad (3)$$

where V_{tot} is the total volume measured at the outlet of the column when the initial concentration is reached at the outlet ($C/C_0 = 1$), C_0 is the initial ammonium concentration, C_i , C_{i+1} are the ammonium concentrations at the moment i and $i+1$, respectively.

4. RESULTS AND DISCUSSION

4.1. Conservative tracer experiment

After 150 min of contact time, the effluent chlorides concentration of all four columns became equal to the influent (data not shown). The chlorides breakthrough curve (BTC) was almost symmetrical up to a value of $C_{tr}/C_{0,tr}$ equal to about 0.87 (Fig. 3), showing that immobile water is not important for transport under the used hydraulic conditions. Above this value, the BTC shows a non-ideal flow pattern, which is likely due to the two layers of gravel located at the boundaries of the column. However, this behaviour should have only a minor effect on the

calculated dispersion coefficient. The parameters v (Darcy's flow velocity) and λ (longitudinal dispersivity) of the convection–dispersion model were optimized using Hydrus-1D inverse method [11].

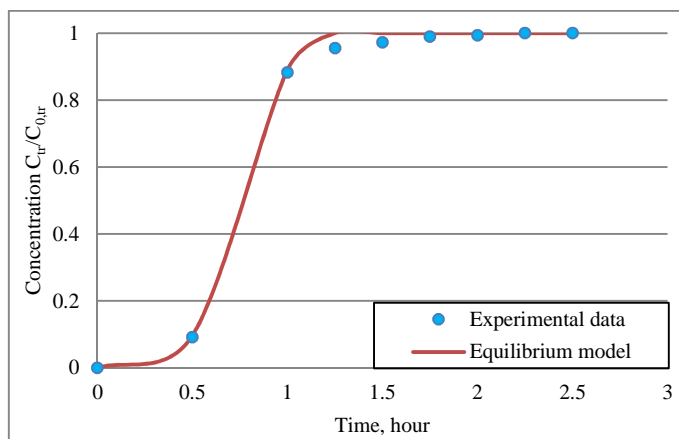


Figure 3. Fitted and observed breakthrough curve of the conservative tracer (chlorides) experiment ($C_{Cl^-} = 5.8 \text{ g L}^{-1}$; $T = 30 \pm 2^\circ\text{C}$, $q = 20 \text{ ml min}^{-1}$).

The longitudinal dispersivity λ and the hydrodynamic dispersion coefficient D of Con Son soil were estimated to 0.2 cm and $8.40 \text{ cm}^2 \text{ h}^{-1}$, respectively. The small values found, indicates that the used soil is a relatively homogeneous porous media. The comparison between the experimental and predicted relative concentrations showed good concordance especially for the values of $C_{tr}/C_{0,tr}$ lower than 0.87 with a correlation coefficient of 0.97 (Fig. 3).

4.2. Ammonium experiments

The total ammonium amount sorbed on Con Son soil matrix has been determined using the equation (3). It varies from almost 1.37 mg/kg to about 24.32 mg/kg for respectively an aqueous ammonium concentration of 4.84 to 102.88 mg/l (Table 1).

Table 1. Ammonium sorbed concentration (C_s) evolution versus the aqueous ammonium concentration.

C_0 , mg/l	4.84	9.65	19.56	36.54	79.82	102.88
C_s , mg/kg	1.37	2.75	6.47	10.41	18.74	24.32

These results show that the ammonium sorption increases proportionally to its aqueous concentration. The linear model was found to be the most appropriate to fit experimental data and then to describe the ammonium adsorption process onto Con Son soil (Fig. 4). Freundlich and Langmuir models failed to fit the experimental data and resulted in low correlation coefficients. Taking into account the overall curve, the distribution coefficient K_d (or C/C_0 rate) value was estimated to be about 0.23 mL g^{-1} with a correlation coefficient of 0.9925. However, this model seem to overestimate and underestimate just a little of the adsorbed ammonium masses for the high aqueous concentrations (36.54 ; 79.82 and 102.88 mg L^{-1}) and the low aqueous concentrations (4.84 ; 9.65 and 19.56 mg L^{-1}) respectively. In order to calibrate this issue, two linear portions were distinguished. The first one corresponded to the relatively low

concentrations and the second to the higher ones. The distribution coefficients and correlations coefficients were determined to be 0.35 mL g^{-1} , $R^2 = 0.9962$ and 0.21 mL g^{-1} , $R^2 = 0.9963$ (Fig. 4).

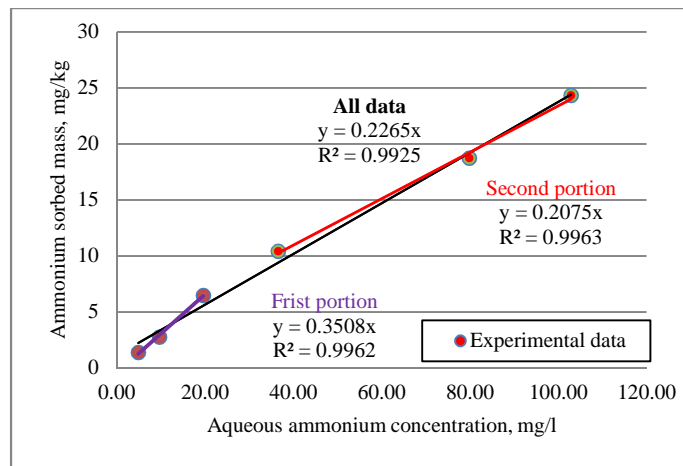


Figure 4. Equilibrium adsorption isotherm of ammonium onto Con Son soil ($T = 30 \pm 2 \text{ }^\circ\text{C}$, $q = 20 \text{ ml min}^{-1}$).

The relatively low distribution coefficient obtained confirms that Con Son soil matrix corresponds to a sandy soil lacking clay particles and organic matter which are the most responsible for ammonium adsorption. As stated by [13], K_d increase with the soil clay fraction. The results of transport parameters estimated by Hydrus 1D are represented in Table 2.

Table 2. Predicted transport parameters for ammonium using the equilibrium and 1 site chemical non-equilibrium models in Hydrus-1D for different initial aqueous ammonium concentrations.

C_{0,NH_4^+} (mg/l)	Equilibrium model			1 site chemical non-equilibrium model			
	λ (cm)	K_d (1/kg)	R^2 (%)	λ (cm)	K_d (1/kg)	α (1/h)	R^2 (%)
4.84	0.2	0.35	93.81	0.2	0.35	1.00 (0.35)	99.17
9.65	0.2	0.35	89.28	0.2	0.27	2.82 (0.17)	99.44
19.56	0.2	0.35	91.53	0.2	0.35	5.20 (0.68)	98.27
36.54	0.2	0.21	93.89	0.2	0.21	5.43 (0.70)	98.90
79.82	0.2	0.21	82.61	0.2	0.21	6.77 (0.70)	99.17
102.88	0.2	0.21	76.61	0.2	0.21	7.19 (0.70)	98.85

The experimental breakthrough curves (BTCs), the fitted equilibrium (Eq. (1)) and the chemical non-equilibrium (Eq. (2)) results are presented in Fig. 5a–f. All the BTCs simulated by the equilibrium model appear after the experimental ones. In addition they have very steep slopes analogous to the tracer's, which differ enormously from the experimental BTCs. So, the equilibrium model could not simulate well the ammonium adsorption process, predicting a faster ammonium transport. Using the two-site chemical non-equilibrium model shows that the ammonium BTCs fit well with the experiment data. To better simulate the experimental

ammonium BTCs, the two-site chemical non-equilibrium model requires two additional parameters, the mass transfer coefficient (α) and the fraction (F) of sorption sites in equilibrium with the liquid-phase concentration [14]. The results (data not shown) returned very small value of F (around 0.01). For this reason, the 1 site chemical non-equilibrium model ($F = 0$) was used in order to simulate the ammonium BTCs for different aqueous ammonium concentrations and the 2 site chemical non-equilibrium model was used for different flow rates.

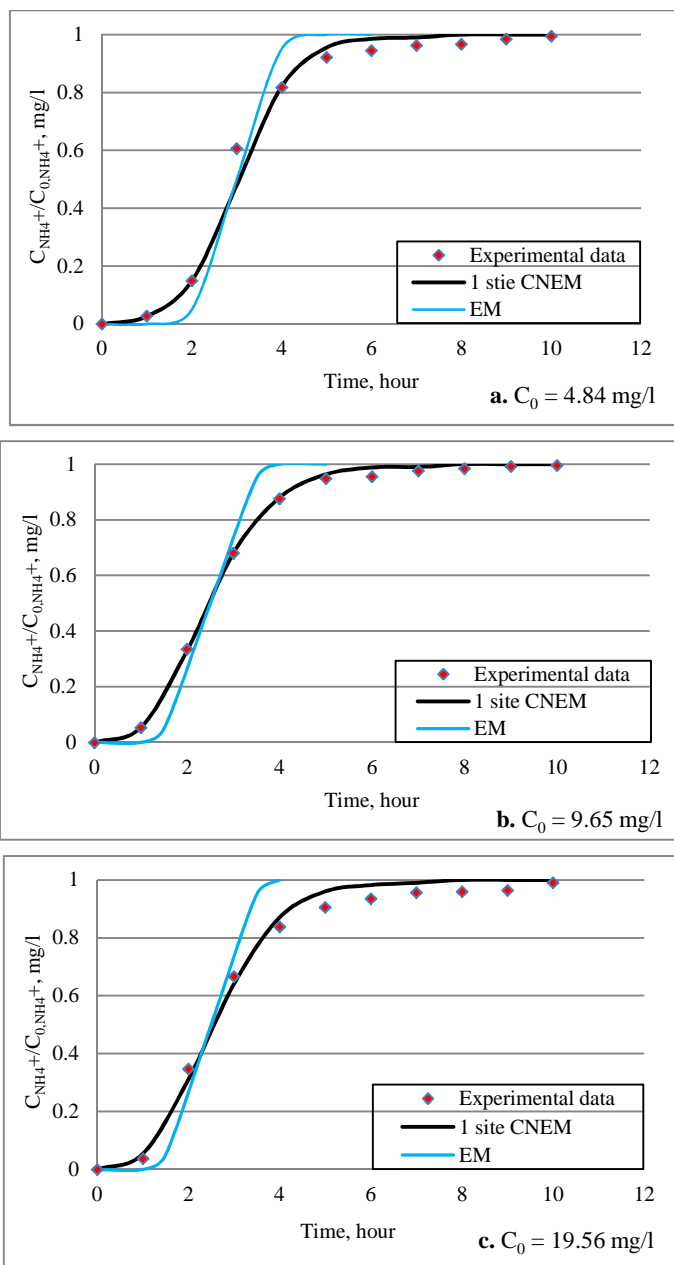


Figure 5a, b, c. Fitted and observed breakthrough curves of ammonium transport experiments ($T = 30 \pm 2 \text{ }^\circ\text{C}$; $q = 20 \text{ ml min}^{-1}$).

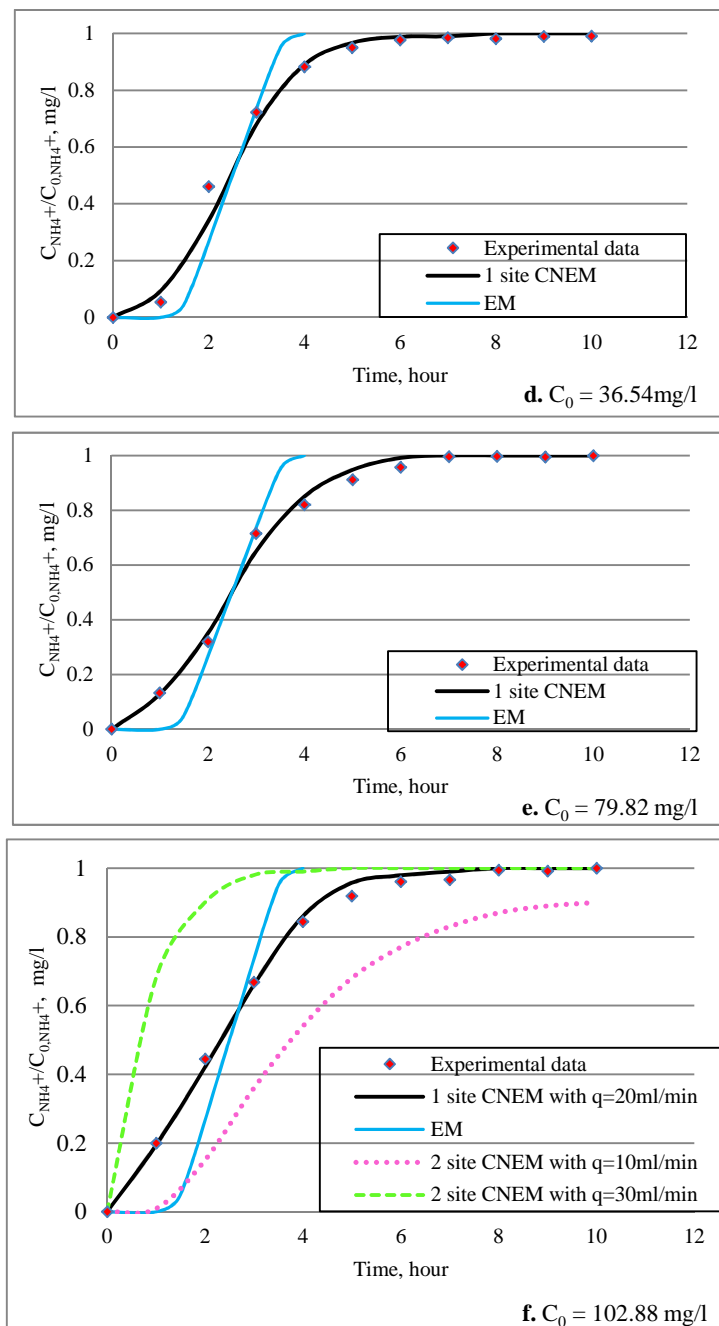


Figure 5d, e, f. Fitted and observed breakthrough curves of ammonium transport experiments ($T = 30 \pm 2 \text{ }^\circ\text{C}$, $q = 20 \text{ ml min}^{-1}$).

BTCs simulated by one site chemical non-equilibrium model fit very well with the experimental ones (Fig. 5), indicating that the ammonium adsorption is chemical kinetic process [9]. Compared with the fraction (F) of sorption sites, the mass transfer coefficient (α) characterizes the adsorption kinetic process. High values of the mass transfer coefficients leads to relatively fast equilibrium between ammonium in the liquid and onto the solid phases, while

low values lead to less adsorption [6]. It is obvious that the non-equilibrium model predicts well the experimental BTCs without needing to fit the experimental distribution coefficients K_d .

The mass transfer rate coefficient (α) varies from 1.00 to 7.19 h^{-1} for aqueous ammonium concentrations of 4.84 and 36.4 mg L^{-1} respectively, indicating a limited mass transfer of the ammonium from the aqueous solution to the soil matrix for low concentrations.

The BTCs of ammonium obtained for the three applied flow rates (10, 20 and 30 mL min^{-1}) are given by Fig. 6. Figure 6 showed an increase of ammonium adsorption onto Con Son soil matrix with a decrease in flow rate and hence an increase in the contact time (see Table 3). For the three experiments, the correlation coefficient was higher than 0.97 (see Table 4). The K_d values were proportional to contact time between ammonium and Con Son soil matrix. For ammonium, a significant decrease in the mass transfer coefficient was observed as velocity decreased.

Table 3. Ammonium sorbed concentration (C_s) evolution versus flow rates (q) and contact time (t).

q ≠ const			
q, ml/min	10	20	30
Cs, mg/kg	29.12	19.55	14.94
t ≠ const and q = 10ml/min			
t, min	60	120	180
Cs, mg/kg	0.11	3.80	11.70

Table 4. Predicted transport parameters for ammonium using the 2 site chemical non-equilibrium model in Hydrus-1D. for different flow rates and a constant initial aqueous ammonium concentration.

Flow rate, ml/min	2 site chemical non-equilibrium model				
	λ (cm)	K_d (1/kg)	α (1/h)	F (-)	R^2 (%)
10	0.2	0.29	1.02 (0.01)	0.09	97.51
20	0.2	0.19	2.34 (1.04)	0.01	99.27
30	0.2	0.15	2.89 (1.86)	0.00	98.23

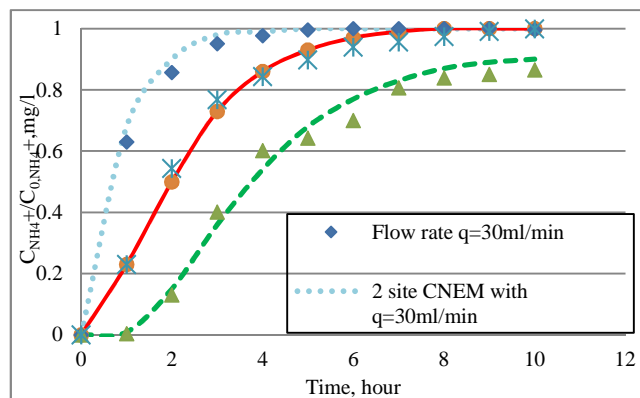


Figure 6. Fitted and observed breakthrough curves of ammonium transport experiments for different flow rates ($T = 30 \pm 2 \text{ }^\circ\text{C}$, $C_{0,\text{NH}_4^+} = 100 \pm 2 \text{ mg/l}$).

5. CONCLUSIONS

Based on the conservative tracer experiments (CI), the longitudinal dispersivity λ and the hydrodynamic dispersion coefficient D of Con Son soil were estimated to 0.2 cm and 8.40 cm² h⁻¹, respectively. The small values found, indicates that the used soil is a relatively homogeneous porous media. The ammonium BTCs for six aqueous concentrations varying between 4.84 and 102.88 mg L⁻¹ showed that the one site chemical non-equilibrium processes played a major role in the ammonium transport through Con Son soil matrix. The two distribution coefficients K_{d1} and K_{d2} were determined to be 0.35 mL g⁻¹ with correlations coefficients $R^2 = 0.9962$ and 0.21 mL g⁻¹ with $R^2 = 0.9963$. The mass transfer coefficient α was found to be dependent on the ammonium aqueous concentration, it varied between 1.00 and 7.19 h⁻¹ for an ammonium concentration of 4.84 and 102.88 mg L⁻¹ respectively. The flow rate through Con Son soil column has an important role on ammonium adsorption. The BTCs of ammonium obtained for the three applied flow rates (10, 20 and 30 mL min⁻¹) showed that the amount of adsorbed ammonium increases significantly and the mass transfer coefficient decreased when the contact time in the column increased.

Based on the experimental and simulation results obtained under the conditions of fine sand with low TOC contents, it can be concluded that the ammonium attenuation in the groundwater by Con Son soil matrix adsorption is very weak. Hence it is completely possible to applying the in-situ treatment measures, comprising ammonium nitrification and denitrification processes in unsaturated/saturated zones in order to prevent and reduce ammonium transport from Con Dao cemetery to the underlying groundwater.

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