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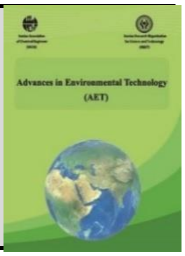

Experimental and mathematical investigation of time-dependence of contaminant dispersivity in soil

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ABSTRACT

Laboratory and field experiments have shown that dispersivity is one of the key parameters in contaminant transport in porous media and varies with elapsed time. This time-dependence can be shown using a time-variable dispersivity function. The advantage of this function as opposed to constant dispersivity is that it has at least two coefficients that increase the accuracy of the dispersivity prediction. In this study, longitudinal dispersivity values were obtained for the conservative NaCl solute transport in a laboratory porous medium saturated with tap water. The results showed that the longitudinal dispersivity initially increased with time (pre-asymptotic stage) and eventually reached a constant value (asymptotic stage). Four functions were used to investigate the time variations of dispersivity: linear, power, exponential and logarithmic. In general, because of the linear increase of dispersivity during a long time of transport, the linear function with R^2=0.97 showed better time variations than the other three functions; the logarithmic function, having an asymptotic nature, predicted the asymptotic stage successfully (R^2=0.95). The ratio of the longitudinal dispersivity to the medium length was not constant during the transport process and varied from 0.01 to 0.05 cm with elapsed time.

1. Introduction

Nowadays, groundwater is a valuable resource that is used for many purposes: irrigation, drinking, domestic activities, etc. Unfortunately, this resource is exposed to contamination from agricultural chemicals (herbicides, pesticides and fertilizers), industrial wastes, leachate from landfills, and various human activities. In order to implement proper contaminant disposal methods, the prediction of the material distribution in the soil subsurface is necessary. Such a prediction needs methods that qualitatively and quantitatively describe the phenomena affecting the contaminant transport. The transport of dissolved contaminants in the subsurface is controlled by various mechanisms and is usually described using the advection-dispersion equation (ADE). In this equation, it is assumed that the movement of the dissolved contaminants includes advective and dispersive fluxes with constant dispersion and velocity [1]. The advective flux describes the solute movement with water flow, and the dispersive flux is caused by variations in the pore-water velocity due to the porous medium heterogeneity [2]. However, one of the disadvantages of the ADE is that it does not take into consideration dispersion variability. So, many studies solve this equation based on the assumption of time- and scale-constancy of dispersion [3]. However, laboratory and field experiments show that the dispersivity and, thus, the pattern of solute distribution are time-dependent. So, the dispersivity increases linearly with elapsed time (pre-asymptotic stage) and finally reaches a constant asymptotic value (asymptotic stage) [4]. This can affect the mechanism of solute transport. However, the time-variability of dispersivity in the heterogeneous porous media is a fully accepted issue by researchers. Field studies show that the variance of the mean travel distance increases non-linearly with elapsed time. This non-linear relationship is attributed to the heterogeneity of the porous media. To describe the solute transport in such medium, it is necessary to use time-dependent dispersivity [5]. Glimm et al. [6] recommended using a time-variable dispersivity function in the ADE to

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consider this non-linear relationship. Levy and Berkowitz [7] pointed out the early arrival times and long late time tails based on tracer test measurements, which is in contrast to the breakthrough curves predicted by the ADE. Such anomalous behavior is often attributed to the heterogeneity of the porous medium and the pre-asymptotic stage [8]. When the dispersivity is considered as a constant parameter in transport processes, it means that the porous medium is a homogeneous system [9]. However, the ADE is still known as the main equation to describe the solute transport in porous media. On this basis, one of the methods to solve the problem of an inaccurate prediction of transport by the ADE is to define the dispersivity by considering its time-dependence. Thus, instead of using equations other than the ADE, dispersivity is introduced in the ADE as a time-variable function. This increases the applicability of the ADE in porous media. Moreover, in most experiments of limited duration, it is probable that the asymptotic stage is not reached at all. In such cases, the appropriate analysis of the experiments should be based on the time-variability of dispersivity [10-12]. Several studies have been conducted in the field of time-dependence of dispersivity. Basha and El-Habel [13] as well as Mustafa and Liao [14], assuming the time-dependence of the dispersion coefficient, presented an analytical solution for the one- and two-dimensional ADE, respectively. They used the linear, power and asymptotic functions to describe the time-variability of the dispersion coefficient. They concluded that a time-variable dispersion coefficient could be used to model the solute transport in heterogeneous hydrological systems. Srivastava et al. [15] and Sharma and Srivastava [16] presented analytical and numerical (finite difference) solutions for the one-dimensional ADE, respectively. They defined dispersivity by a time-variable exponential function for describing the time-variability of hydraulic conductivity. The results showed that their solutions could successfully predict the effect of heterogeneity on the solute transport. However, they only investigated the increase of dispersivity with time, neglecting the asymptotic stage. Zhou [9] used linear and non-linear fractal functions to describe the time-dependence of dispersivity. He concluded that the fractal dimension has a significant effect on the breakthrough curve (BTC). So that the more its value is, the longer the BTC's tailing will be [9]. Su et al. [3] analytically solved the ADE under three different boundary conditions. They defined the dispersion coefficient as a time- and scale-variable. For this purpose, they introduced a product of the power function of time, distance and velocity in the ADE, instead of a constant dispersion coefficient. The BTCs of the analytical solutions showed a good agreement with the results of the laboratory and field experiments. Kumar et al. [5], using a non-linear function, evaluated the time-dependence of dispersivity of conservative solutes with a constant continuous source for a system of parallel fractures. Their results were consistent with the experimental study of Neretnieks et al. [10] and Moreno et al. [11]. Sharma et al. [17], assuming time- and scale-dependent dispersion coefficients, used a finite volume method to solve the conservative solute transport equation. They assumed that dispersivity was linearly proportional to the mean travel distance (the mean travel distance is equal to the product of pore-velocity and time). Their results showed that this numerical model better simulated the radionuclide concentration profile than did the constant dispersion model. Natarajan [18] investigated the effect of time-dependent dispersion on non-linearly sorbed multi-species contaminants in porous media. He defined the dispersivity as a multiple of the mean travel distance. He concluded that the time-dependent dispersivity led to a higher concentration when compared to that obtained with the constant dispersivity. This occurred because the time-dependent dispersivity provided a significant dispersion coefficient, due to which the concentration was distributed in the medium in a much higher amount. However, since dispersion is one of the important phenomena in contaminant transport in porous media, the dispersivity variations with time, despite its certainty, is still open to study. According to our knowledge, there are no studies in the field of contaminant transport in porous media in which the various time-variable dispersivity functions are thoroughly compared with each other. Moreover, in most of the studies regarding the time-dependence of dispersion, hypothetical data, rather than experimental observations, have been used to verify the accuracy of analytical and numerical models. In this study, the conservative NaCl solution was injected in a laboratory porous medium; then, the dispersivity values of NaCl were determined at different times of the experiment. The linear, power, exponential and logarithmic functions were fitted to the dispersivity-time curve to identify which one could better predict the time variations of dispersivity.

2. Materials and methods

2.1. Experimental setup

The transport experiment was conducted in a cube-shaped metal box with the following dimensions: 200 cm length, 100 cm width and 25 cm height. A rainfall simulator was installed on top of the box at a 60 cm height. There were two reservoirs to supply water and solution; each of them had its own pump. This box (main box) had two small boxes on both sides with the dimensions of 20 cm in length, 100 cm in width and 40 cm in height; one box was for the solute injection and another for discharging the solute and keeping the porous medium saturated. Each of these small boxes had a hole in the center at the height of 20 cm in contact with the main box. The holes were covered by metal meshes to prevent particles entering from the main box into the small boxes. Two vertical pipes with adjustable heights were installed inside each of the small boxes to establish constant hydraulic head. The pipes of the injection and discharge boxes were adjusted at the heights of 40 and 30 cm, respectively. Therefore, a 20 cm constant hydraulic head of NaCl solution was established in the injection box. Similarly, the constant hydraulic head of the discharge box was 10 cm. The concentration of the NaCl solution was 5 g/L. In order to get samples from the solute, 9 piezometers were installed at 20 cm horizontal intervals in the bottom of the main box. This box was filled with gravels that were 2-4 mm in diameter as the porous medium. The porous medium was completely saturated with tap water using a rainfall simulator. The concentration of the tap water (0.225 g/L) was considered negligible in the data analysis. Figure 1 shows a schematic of the experimental setup, and Table 1 gives the hydraulic conditions of the experiment.

Table 1. Hydraulic conditions of the transport experiment in the porous medium.

i (-)	k_s (cm/min)	q (cm/min)	n (%)	v (cm/min)
0.05	1.9	0.095	22.12	0.43

(i) hydraulic gradient, (k_s) saturated hydraulic conductivity, (q) flow rate, (n) porosity, (v) pore-water velocity

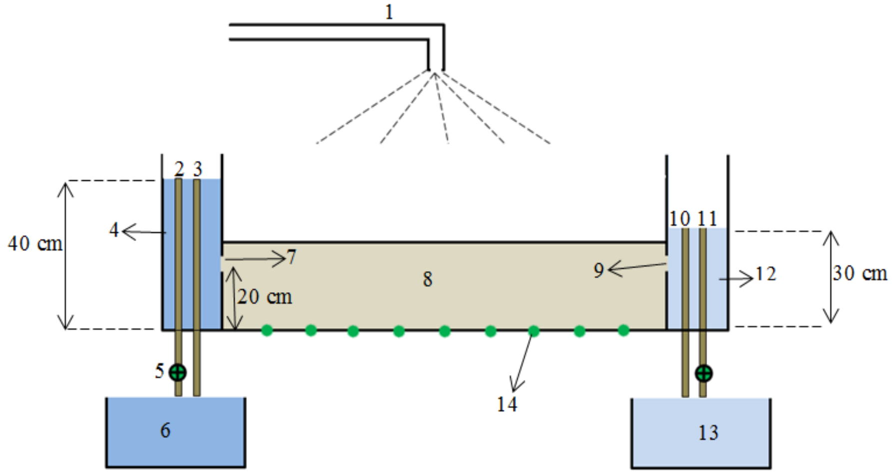

Fig. 1. Schematic of the transport experiment in the porous medium.

(1) rainfall simulator, (2) solute inlet pipe in injection reservoir, (3) solute outlet pipe in injection reservoir (to establish constant hydraulic head), (4) solute injection reservoir, (5) pump, (6) solute supply reservoir, (7) inlet hole in the interface of injection reservoir and porous medium, (8) porous medium, (9) outlet hole in the interface of porous medium and discharge reservoir, (10) water/solute outlet pipe in discharge reservoir (to establish constant hydraulic head), (11) tap water inlet pipe in discharge reservoir, (12) discharge reservoir, (13) tap water supply reservoir, (14) piezometers

2.2. Transport experiment and determination of dispersivity at different times

After establishing the hydraulic gradient between the solute inlet and outlet holes, the transport experiment began. The samples were taken from the piezometers at 15 min intervals using an EC-meter until the concentration of solute in all the piezometers become equal to that of the injected solute (5 g/L). Then, the values of the dispersion coefficient were estimated using the non-linear least-squares parameter optimization code CXTFIT2.1 of Toride et al. [19]. This code can inversely estimate the dispersion coefficients from the observed concentrations in the different locations and times through solving the one-dimensional ADE. The one-dimensional ADE for steady water flow in a rigid and saturated porous medium for a conservative solute is generally described as follows:

$$\frac{\partial c}{\partial t} = \frac{\partial}{\partial x}\left(D_L \frac{\partial c}{\partial x}\right) - v_L \frac{\partial c}{\partial x} \qquad (1)$$

where c is the solute concentration (ML^{-3}), D_L is the longitudinal hydrodynamic dispersion coefficient (L^2/T), v_L=q/n is the (constant) mean pore-water velocity (L/T) in which q is the flow rate (L/T) and n is the porosity (-), t is time (T), and x is distance from the injection source (L). The hydrodynamic dispersion includes the mechanical dispersion (due to the medium heterogeneity and the pore-water velocity variations) and molecular diffusion (caused by the random collision of the molecules). However, the molecular diffusion is negligible compared to the mechanical dispersion [20] and thus, the hydrodynamic dispersion (also briefly called dispersion coefficient) is linearly proportional to the pore-water velocity. The factor of this proportionality, being considered as a medium constant [21] and a property of heterogeneity [22] is known as dispersivity (a).

$$D = av \qquad (2)$$

In the CXTFIT2.1, for properly fitting the observed concentrations and estimating the dispersion coefficients, an objective function (the sum of squared residuals, SSQ) is minimized:

$$SSQ = \sum_{i=1}^{N} [c(x_i) - c'(x_i, [v_L, D_L])]^2 \qquad (3)$$

where $c(x_i)$ and $c'(x_i,[v_L,D_L]$ are the observed and estimated concentrations at the distance of x_i from the injection source, respectively. The CXTFIT2.1 assesses the accuracy of the fitting by using the coefficient of determination (R^2).

The linear, power, exponential and logarithmic functions were chosen to describe the time-variability of the longitudinal dispersivity as follows:

$$a_L(t) = \alpha t + \beta \qquad \text{Linear}$$

$$a_L(t) = \alpha t^{\beta} \qquad \text{Power}$$

$$a_L(t) = \alpha e^{\beta t} \qquad \text{Exponential}$$

$$a_L(t) = \alpha \ln(t) + \beta \qquad \text{Logarithmic}$$

where t is time, e is the Neper number (≈2.72) and ln is the natural logarithm (the logarithm to the base of e). α and β are the constants of these functions.

2.3. Statistical criteria

It is not efficient to use only the criteria based on the correlation between the observed and predicted data in analyzing the accuracy of the models, and thus, other criteria must also be used [23]. In this study, to assess the accuracy of the four time-variable functions in predicting the dispersivity at different times, the root mean square error (RMSE) was used in addition to the coefficient of determination (R^2):

$$R^2=\frac{\left[\sum_{i=1}^{n}(O_i-\bar{O})(P_i-\bar{P})\right]^2}{\sum_{i=1}^{n}(O_i-\bar{O})^2\sum_{i=1}^{n}(P_i-\bar{P})^2} \tag{4}$$

$$RMSE=\sqrt{\sum_{i=1}^{n}\frac{(O_i-P_i)^2}{n}} \tag{5}$$

where O_i and P_i are the observed and predicted values and $\bar{O}$ and $\bar{P}$ are their mean values, respectively. n is the number of data pairs. If the precision of the model is high, then the value of R^2 will be close to 1. The closeness of RMSE to 0 indicates the low average error in the model, however, in the same unit as the variable.

Table 2. Dispersivity values at different times.

t (min)	D_L (cm^2/min)	R^2 (-)	a_L (cm)	t (min)	D_L (cm^2/min)	R^2 (-)	a_L (cm)
15	0.26	0.9826	0.61	360	2.39	0.9993	5.56
30	0.27	0.9827	0.63	375	2.59	0.9979	6.02
45	0.46	0.9932	1.08	390	2.72	0.9971	6.32
60	0.48	0.9950	1.12	405	2.87	0.9973	6.68
75	0.52	0.9966	1.21	420	3.01	0.9973	7.00
90	0.57	0.9973	1.34	435	3.15	0.9973	7.33
105	0.64	0.9980	1.48	450	3.29	0.9973	7.66
120	0.71	0.9985	1.64	465	3.37	0.9942	7.87
135	0.78	0.9987	1.82	480	3.70	0.9831	8.89
150	0.87	0.9991	2.03	495	3.90	0.9788	9.07
165	0.96	0.9991	2.24	510	4.06	0.9647	9.44
180	1.05	0.9993	2.44	525	4.10	0.9647	9.54
195	1.15	0.9991	2.68	540	4.15	0.9377	9.66
210	1.25	0.9991	2.90	555	4.17	0.9377	9.70
225	1.35	0.9991	3.13	570	4.20	0.9377	9.77
240	1.46	0.9993	3.41	585	4.22	0.9377	9.81
255	1.61	0.9988	3.75	600	4.24	0.9377	9.85
270	1.71	0.9989	3.97	615	4.25	0.9377	9.89
285	1.84	0.9986	4.28	630	4.25	0.9377	9.89
300	1.96	0.9989	4.56	645	4.27	0.9377	9.92
315	2.07	0.9987	4.81	660	4.28	0.9377	9.96
330	2.13	0.9981	4.95	675	4.28	0.9377	9.96
345	2.30	0.9979	5.34	690	4.30	0.9377	10.00

3. Results and discussion

Table 2 gives the fitted dispersion coefficients by the CXTFIT2.1 at different times along with corresponding dispersivity values. The R^2 in this table reflects the goodness-of-fit. According to Equation (2), the dispersion coefficient was divided by the constant pore-water velocity to calculate the dispersivity(a) at different times.

Figure 2 shows the four functions fitted on the dispersivity-time curve. The α and β coefficients for the four functions were obtained using this figure. Therefore, the form of these functions will be as follows:

$$a_L(t) = 0.0165t - 0.1619 \qquad \text{Linear}$$

$$a_L(t) = 0.0262t^{0.9141} \qquad \text{Power}$$

$$a_L(t) = 1.1045e^{0.0039t} \qquad \text{Exponential}$$

$$a_L(t) = 3.3920\ln(t) \qquad \text{Logarithmic}$$

In Figure 2, it is observed that the dispersivity initially increases from the time zero to about the minute 525 (from 0 to about 9.54 cm) until it reaches an approximately constant value. From this time until the end of the transport experiment, no significant increase occurs in its value (from 9.54 to about 10.00 cm). This is exactly in accordance with the results of many studies [5,14,22]. In general, it has been concluded by several researchers in their experimental and theoretical studies that the dispersion phenomenon includes two important time stages. These stages include: (1) pre-asymptotic stage: the early times of the dispersion phenomenon in which the dispersivity grows linearly with time, and (2) asymptotic stage: the late times of the dispersion phenomenon when the dispersivity becomes constant. Now that the coefficients of the four functions are available, the dispersivity values can be estimated at different times. Figure 3 shows the dispersivity-time curves derived from the four functions along with the observed one. According to this figure and the values of R^2 and RMSE in Table 3, in general, the dispersivities predicted by the linear function have the best fit to the observations with the highest R^2 (0.97) and the least RMSE (0.52 cm).

Table 3. Performance criteria of the four functions.

	linear	power	exponential	Logarithmic
R^2	0.97	0.95	0.90	0.78
RMSE	0.52	0.67	1.80	1.53

After this function, the performance of the power function is satisfactory and better than that of the exponential and logarithmic functions. The logarithmic function fails to predict the dispersivity with an acceptable accuracy. The reason behind a good prediction by the linear function is that approximately 525 minutes of the transport process belong to the pre-asymptotic stage (linear growth of dispersivity), while the asymptotic stage extends only from the minute 525 to the minute 690 (the end of the transport experiment). This means that more than three-quarters of the transport duration belong to the pre-asymptotic stage. However, the disadvantage of the linear function is that it does not show the asymptotic stage. In other words, the dispersivity never reaches a constant value in the linear function and increases linearly. This means that from the minute 525 to the end of the experiment, the dispersivity varies from 8.50 to 11.22 cm, which is 1.22 cm more than the last observed dispersivity (10.00 cm). In the power function, since the time variable (t) has a power close to 1 (0.9141) and a small factor (0.0262), the slope of the dispersivity growth is smoother than that of the linear function. As a result, the dispersivity value at the end of the transport experiment can be predicted with good accuracy (10.31 cm), which only has a 0.31 cm difference with the observed value. However, the power function fails to show the asymptotic stage appropriately because the dispersivity increases with the time, like the linear function. Despite having a curve of asymptotic nature, the exponential function is not suitable for predicting the asymptotic stage of dispersivity because it has a positive exponent that makes its asymptotic segment vertical while the asymptotic segment of the dispersivity-time curve is horizontal. As a result, the exponential function estimates the final dispersivity with a large difference in respect to the observed one (16.29 cm).

The logarithmic function, having a curve of asymptotic nature with horizontal tailing, is suitable to predict the asymptotic stage of dispersivity. However, this function with an R^2=0.78 has not properly shown the general trend of dispersivity. Since the accuracy of the four functions in predicting the pre-asymptotic and asymptotic stages are contradictory, separate analysis of these two stages can better describe the performance of the aforementioned functions. In the general analysis, the linear function is more accurate than the other three functions. Now, we investigate the dispersivity only at the asymptotic stage (i.e., from the minute 525 to the end). Figure 4 shows the four functions fitted on this stage. It is observed that the linear function is no longer the best fit; instead, the logarithmic function has a better fit than the other three. As previously mentioned, this occurs because of the asymptotic nature and the horizontal tailing of the logarithmic function. Basha and El-Habel [13] believed that an asymptotic dispersion function is more important and practical. So, they restricted their analysis regarding the solute transport to this function. However, they stated that to describe the solute transport, it is necessary to use a time-dependent dispersion coefficient at the onset of the transport in which dispersivity increases. In general, it seems that the use of the linear function is more suitable. Especially that this function has good predictions in most times of the dispersion phenomenon (the pre-asymptotic stage) because the dispersivity increases linearly with time before it reaches a constant value. In fact, all the mass initially moves with the mobile water. With time, more and more solute diffuses into the immobile water in the very small pores. This mass diffusion is gradual, and the time needed to achieve a constant-asymptotic dispersivity is relatively lengthy.

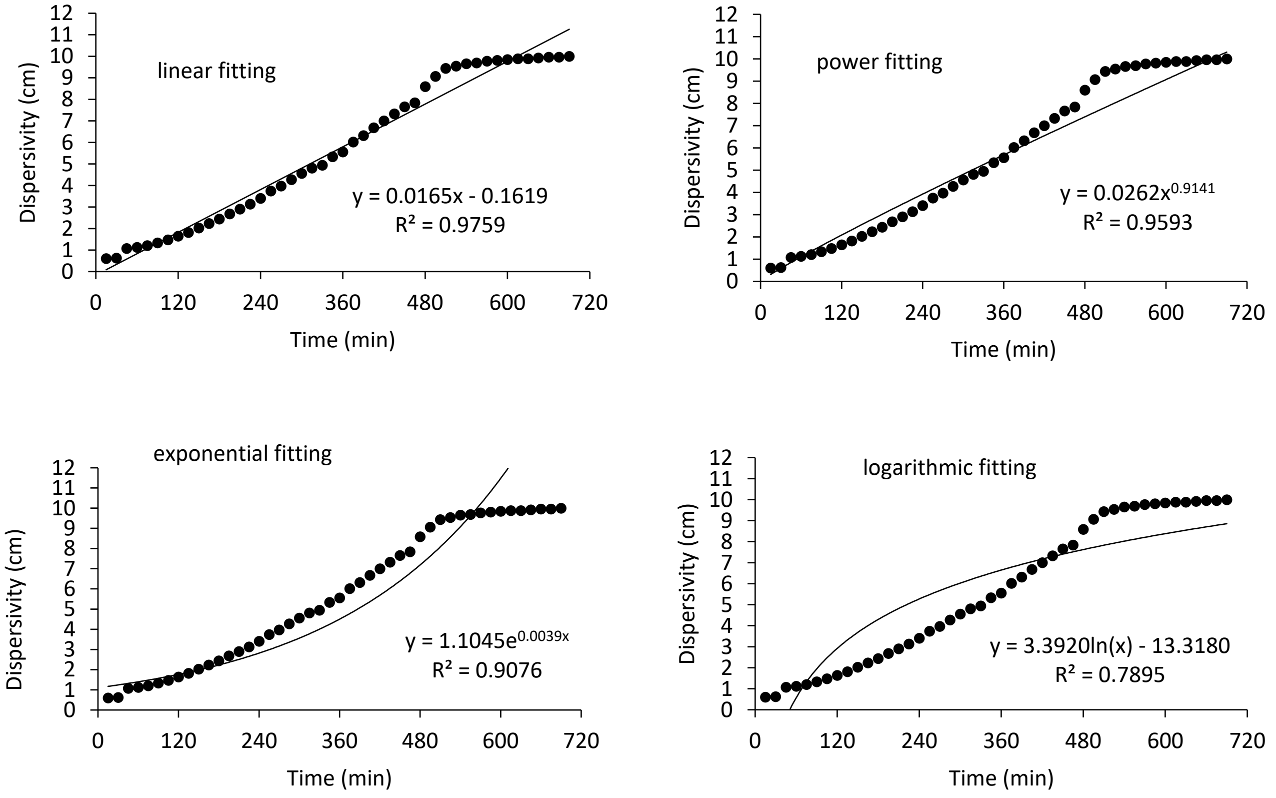

Fig. 2. The four time-variable functions fitted on the dispersivity-time curve.

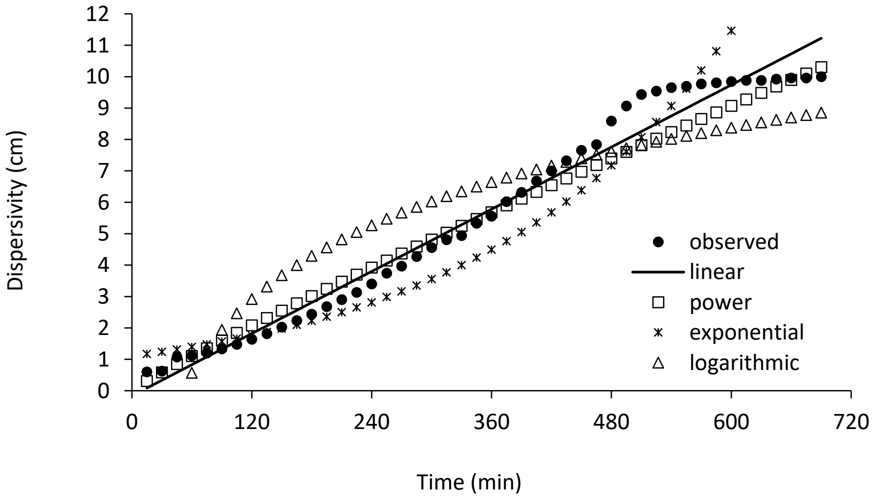

Fig. 3. Observed and predicted dispersivity-time curves.

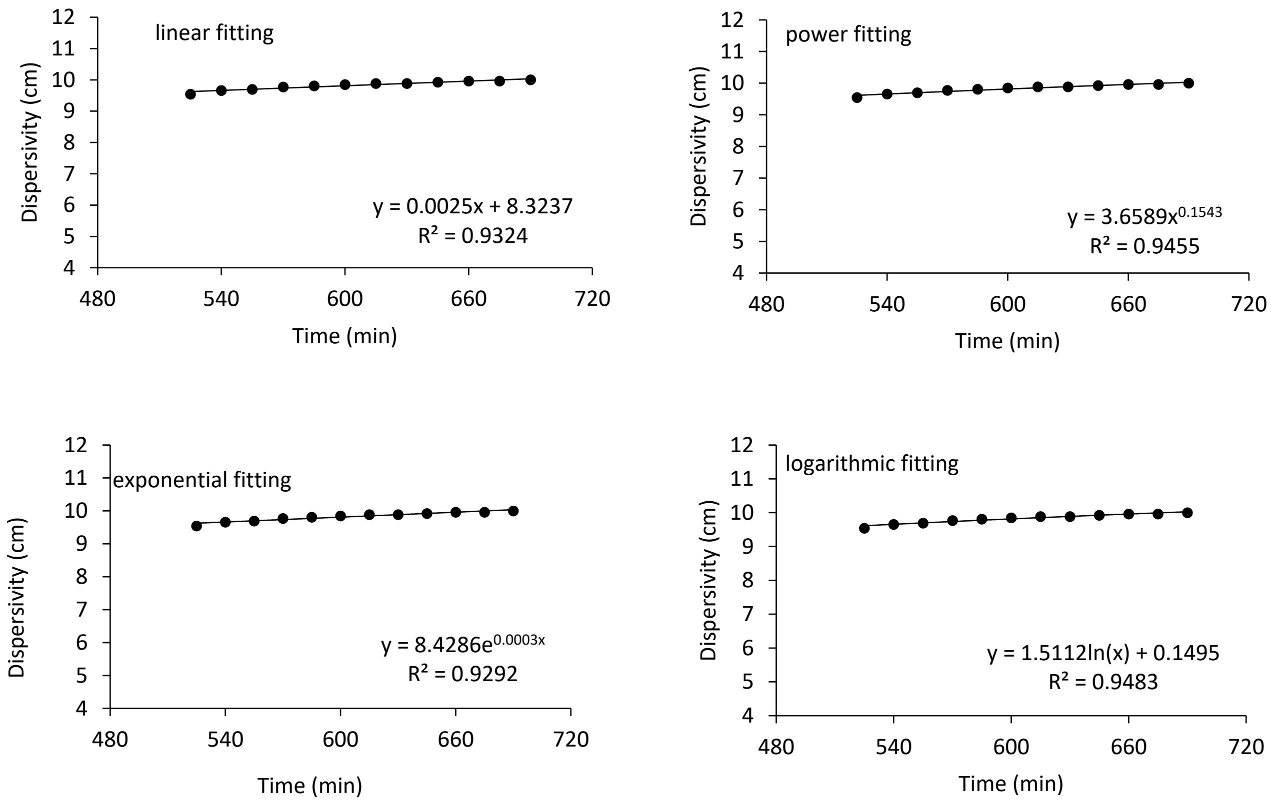

Fig. 4. The four time-variable functions fitting on the asymptotic stage of the dispersivity-time curve.

In several studies [24-27], it was assumed that the longitudinal dispersivity was approximately equal 0.1 to the medium length. However, in the previous sections it was found that the longitudinal dispersivity changed significantly with time. Therefore, the ratio of the longitudinal dispersivity to the medium length (a_L/L) could not be considered constant in all the transport duration. Figure 5 shows this ratio at different times of the transport experiment. Note that the length of the porous medium (L) was 200 cm. According to this figure, the a_L/L changed between 0.00 and 0.05 but did not reach 0.1. This occurred because the lab-scale dispersivity was several orders of magnitude smaller than the field-scale values for the same material. This difference was a reflection of the more heterogeneity in the field-scale that produced more irregular flow patterns [24]. The a_L/L-time curve had a stepwise shape. Gelhar et al. [24] critically reviewed the dispersion data in aquifers and found that the longitudinal dispersivity ranged from 10^{-2} to 10^{4} m for scales ranging from 10^{-1} to 10^{5} m. Hence, it was not appropriate to assume a single value for the longitudinal dispersivity in a certain transport area.

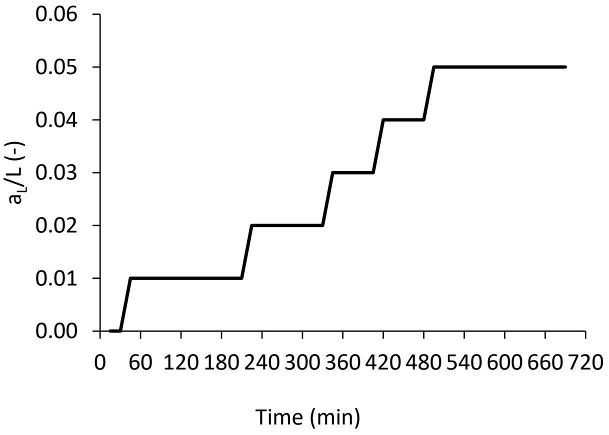

Fig. 5. The ratio of the longitudinal dispersivity to the medium length at different times.

4. Conclusions

Dispersion is caused by the heterogeneity of the porous media. Previous studies have shown that this phenomenon is time-dependent. This dependence can be shown using a time-variable dispersivity function. The advantage of using such a function instead of a constant dispersivity is that it has at least two fitting coefficients that increase the accuracy of description of dispersivity. The results of this study show that, in general, the linear function can better determine the time-variability of dispersivity than the power, exponential and logarithmic functions; this occurs because most of the transport duration is allocated to the pre-asymptotic stage (the linear increase of the dispersivity). However, in the asymptotic stage (the constant dispersivity), the accuracy of the logarithmic function is mostly due to its asymptotic nature with a horizontal tailing. Also, the ratio of the longitudinal dispersivity to the medium increases with time. The results of this study can be useful for future research in the field of two- and three-dimensional transport, non-conservative solutes, etc. However, to determine the overall time-dependence of dispersivity, various long-term experiments should be conducted on a large scale.

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