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Extraction of Model Parameters for Reactive Solute Transport

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ABSTRACT

Rock material is common in the construction of hydraulic structures. In the present study, to the aim is to examine the reactive solute relationships for transport and degradation processes through the rockfill media. By applying the analytical solution of reactive transport, the 1st to 3rd theoretical temporal moments have been extracted, consequently by applying two methods of curve fitting and temporal moment matching, the coefficients of dispersion and degradation have been exploited. Two rock diameter, two operating discharges and five instantly injection mass have been used as the variables of experiments. The EC sensors with operation software were installed inside the rockfill media and then the experimental breakthrough curves with intervals of 4 seconds have been extracted. It is concluded that both methods are suitable for application of transport and degradation processes inside the media. It was found that by increasing inflow discharges, pore velocity, and media diameters the dispersion coefficient decreases and with a decrease in media diameter or with increase in injection mass the decay rate decreases. The sensitivity analysis on the derived moment equation and also skewness coefficient equation indicated that the velocity and degradation are the most and less effective parameters on the moment equations respectively.

1. Introduction

Transport of pollutants through the different type aquatic ecosystems have

been inspected by researchers since many years ago. By assuming complete mixing in the cross section, the one-dimensional

transport model of Taylor [1], can be applied. This model is somewhat simple and do not suffer diversity of model parameter and the transport process can be interpreted by only one parameter which is called Dispersion coefficient [1]. The velocity of the mentioned model is cross sectional average velocity. The dispersion coefficient have been computed firstly by routing method which were presented by Fischer [2]. It is believed by previous researchers that after initial mixing of the tracer over the cross section, the Gaussian distribution is dominant and the variance increases linearly by increasing of the time. However, due to the complexity of the dispersion process and geometrical irregularity of the rivers or other flow fields, the explanation of the dispersion process with this model is not accurate [3]. This fact forces the researchers to use other complicated transport models with more model parameters like Transient storage model or aggregated dead zone model [3]. Moreover, different methods exist for extraction of the model parameters. Two applicable and most famous methods are curve fitting and temporal moment methods. Thackston and Schnelle [4] and Fischer et al. [2] tried to catch temporal variation of the concentration in the dead zone and concluded that the dead zone model is applicable for interpretation of the skewness of pollutographs, having long tail in the falling limb. Thackston and

Schnelle [4], Pedersen [5], Nordin and Troutman [6], and Seo and Maxwell [7] confirmed that the transient storage model produce better curve fitting for rebuilding of the long tail of the breakthrough curves. Czernuszenko et al. [8] concluded that there is difficulty in physically interpretation of the dead zone model parameters. Firstly, mathematical closed form of the first to third temporal moments of the dead zone model were extracted by Hays et al. [9] afterwards, the detailed description of them have been investigated by Nordin and Troutman [6] by application of Laplace transform. These relationships have been extracted for instantaneous mass slug over the cross section in the conservative mood for the tracer. After these researchers, the temporal moment relationships for none-conservative mood have been extracted by Schmid [10]. For 1st to 3rd moments and consequently the fourth moment were presented by Seo and Cheong [11].

Generally it can be concluded that theoretically relationships for different types of transport model in various initial and boundary conditions have been propped by Harvey and Gorelick [12], Luo et al. [13] for mass transfer of transport model, Czernuszenko and Rowinski, [8], Schmid, [14] for transient storage model, Goltz and Roberts [15], Cunningham and Roberts [16] for equilibrium and none equilibrium sorption models, Lees et al. [17] for

aggregated dead zone models and Argerich et al. [18] for metabolically active TS model [19].

2. Material and Methods

2.1 Theoretical Approach

If the advection and dispersion of solute is in none-conservative mood like so many solutes such that, the recovered mass from experimental BC¹ curves is not equal with injected mass, it is called reactive solute transport. Putting it differently, in this case, the total mass of contaminants does not recover itself. The one-dimensional mass transport equations by adding degradation term becomes as equation (1).

$$\frac{\partial c}{\partial t} + v \frac{\partial c}{\partial x} = D \frac{\partial^2 c}{\partial x^2} - kc \quad (1)$$

Where D is dispersion coefficient and k is degradation rate.

Equation (1) is extension of classical AD equation by first-order degradation term and $k > 0$ that implies the contaminant degrades in its transportation path. By contemplating instantaneous slug release with mass of M to one dimensional flow, analytical solution of equation (1) becomes as equation (2).

$$C(x, t) = \frac{M}{A\sqrt{4\pi Dt}} \exp\left(-\frac{(x-Vt)^2}{4Dt} - kt\right) \quad (2)$$

Where A is cross sectional area, x is the location of data acquisition, V is the cross

sectional average flow velocity and t is the time matrix [3].

One of the important methods for extraction of dispersion coefficient and degradation rate is curve fitting to the analytical solution (equation 2) with experimental acquired data series.

Since experimental results are normally based on the temporal distribution of concentration (concentration variation versus time at the fixed location), therefore temporal analysis leads to extract characteristics of dispersion and degradation parameters. Applying analytical temporal moments for experimental BC curves, the central tendency of them can be computed. The absolute n th temporal moment of concentration time series $C(t)$ at the distance of x with respect to zero is defined as equation (3):

$$\mu_n = \int_0^{\infty} t^n C(t) dt \quad (3)$$

And additionally n th normalized temporal moment about zero moment can be defined as equation (4):

$$\mu_n^* = \frac{\mu_n}{\mu_0} = \frac{\int_0^{\infty} t^n C(x, t) dt}{\int_0^{\infty} C(x, t) dt} \quad (4)$$

If the moments is computed with respect to the mean, then n th normalized temporal moment about the mean is m_n and is defined as the equations of (5) and (6):

¹) Breakthrough

$$m_n = \frac{1}{\mu_0} \int_0^\infty (t - \mu_1^*)^n C(t) dt \quad (5)$$

$$m_n = \sum_{i=0}^n \binom{n}{i} \mu_{n-i}^* (-\mu_1^*)^i \quad (6)$$

Where: i is an index. The equation (6) is an inverse binomial transform which can be applied to compute normalized central moments of order 1 (mean travel time), 2 (variance) and 3 (skewness) (Equations of 7, 8 and 9)[20].

$$m_1 = \mu_1^* \quad (7)$$

$$m_2 = \mu_2^* - (\mu_1^*)^2 \quad (8)$$

$$m_3 = \mu_3^* - 3\mu_1^*\mu_2^* + 2(\mu_1^*)^3 \quad (9)$$

By contemplating the equation of (2) as the base equation for our research and also doing some mathematical integrations, the zero ,1st ,2nd and 3rd theoretical temporal moments have been computed respectively as equations of (10), (11), (12) and (13). If the classical advection-dispersion equation was the base function then the zero moment was equal to the M/Q and due to the constant rate of flow, the mass of the tracer is product of volumetric flow rate and zero moment.

$$\mu_0 = \frac{M}{A\sqrt{V^2 + 4kD}} \times \exp\left(\frac{xV - x\sqrt{V^2 + 4kD}}{2D}\right) \quad (10)$$

$$\mu_1 = \frac{M(2D + x\sqrt{V^2 + 4kD})}{A(V^2 + 4kD)^{3/2}} \times \exp\left(\frac{xV - x\sqrt{V^2 + 4kD}}{2D}\right) \quad (11)$$

$$\mu_2 = \frac{M \left[\frac{x^2(V^2 + 4kD) + 12D^2}{+6Dx\sqrt{V^2 + 4kD}} \right]}{A(V^2 + 4kD)^{5/2}} \times \exp\left(\frac{xV - x\sqrt{V^2 + 4kD}}{2D}\right) \quad (12)$$

$$\mu_3 = \frac{M \left[\frac{12Dx^2(V^2 + 4kD) + 120D^3}{+60xD^2\sqrt{V^2 + 4kD}} + x^3(V^2 + 4kD)^{3/2} \right]}{A(V^2 + 4kD)^{7/2}} \times \exp\left(\frac{xV - x\sqrt{V^2 + 4kD}}{2D}\right) \quad (13)$$

By manifesting more algebraic operations,

The 0th, 1st, 2nd and 3rd normalized moments with respect to the zero have been computed as equations of (14) to (17). As is illustrated, the normalized zero moment is equal to one.

$$\mu_0^* = 1 \quad (14)$$

$$\mu_1^* = \frac{(2D + x\sqrt{V^2 + 4kD})}{(V^2 + 4kD)} \quad (15)$$

$$\mu_2^* = \frac{\left[\frac{x^2(V^2 + 4kD) + 12D^2}{+6Dx\sqrt{V^2 + 4kD}} \right]}{(V^2 + 4kD)^2} \quad (16)$$

$$\mu_3^* = \frac{\left[\begin{array}{l} 12Dx^2(V^2 + 4kD) + 120D^3 \\ + 60xD^2\sqrt{V^2 + 4kD} + \\ x^3(V^2 + 4kD)^{\frac{3}{2}} \end{array} \right]}{(V^2 + 4kD)^3}$$

Furthermore, with regard of equations of (7) to (9) and (14) to (17) by applying some more algebraic operations, the normalized temporal moments with respect to the mean have been computed as the equations of (18) to (21). These relationships can be applied for calculation of mean travel time, temporal variance and skewness coefficient of experimental BC curves.

$$m_1 = \mu_1^* = \frac{(2D + x\sqrt{V^2 + 4kD})}{(V^2 + 4kD)} \quad (18)$$

$$= t_m$$

$$m_2 = \mu_2^* - (\mu_1^*)^2 \quad (19)$$

$$= \frac{8D^2 + 2Dx\sqrt{V^2 + 4kD}}{(V^2 + 4kD)^2} = \sigma_t^2$$

$$m_3 = \mu_3^* - 3\mu_1^*\mu_2^* + 2(\mu_1^*)^3 \quad (20)$$

$$= \frac{64D^3 + 12D^2x\sqrt{V^2 + 4kD}}{(V^2 + 4kD)^3}$$

$$SC = \frac{m_3}{m_2^{\frac{3}{2}}} \quad (21)$$

$$= \frac{16D + 3x\sqrt{V^2 + 4kD}}{(4D(4D + x\sqrt{V^2 + 4kD}))^{\frac{3}{2}}}$$

Where: t_m is mean travel time of the BC curve, σ_t^2 is temporal variance of BC curve and SC is skewness coefficient of BC curve

The first moment has dimension of time and is from $t=0$ to the center of gravity of

(17) the area under time-concentration curve. Generally, it is called the average travel time of the tracer (equation 22). In many experimental or field conditions, by manifesting more simplification on the second central temporal moment of the classical advection-dispersion equation and neglecting of one ignorable term, the mean residence time of tracer can be approximated as (22):

$$m_1 = \mu_1^* = t_m = \frac{x}{V} \quad (22)$$

Where t_m is average travel time.

The equation of (23) can be computed from second central moment from classical advection-dispersion equation.

$$D = \frac{V^3}{2x} (\mu_2^* - (\mu_1^*)^2) \quad (23)$$

$(\mu_2^* - (\mu_1^*)^2)$ is variance of BC curve. It is clear that time temporal variance changes linearly with distance which gives equation (24) [3].

$$D = \frac{V^3}{2} \frac{d\sigma_t^2}{dx} \quad (24)$$

On that account, it is apparent that if σ_t^2 is plotted against x as straight line, then the longitudinal dispersivity is a constant value independent to the travel distance and the dispersion coefficient can be calculated applying the slope of straight line from (24).

Now by calculation of central temporal moments from experimental data series and regarding analytical solutions of

different moments, one set of two equations (18 to 21) with two unknown parameters can be formed for estimation of dispersion coefficient and degradation rate or by using equation (22) which is operated with large number of previous researchers [19], the dispersion coefficient would be extracted by using one other equation (18 to 21). Hence, the degradation rate would be computed.

2.2 Experimental Data

The experiments have been operated in the rockfill media box with 1.3m length and 0.2m width having erect up and downstream sides. Two rock median diameters of 1.1 and 1.8 cm having porosities of 42 and 47% respectively were imposed inside the media box. Five injection masses of 5, 10, 15, 25 and 50 gr have been introduced instantly at the upstream of media as tracer.

Two completely flowthrough discharges of 0.26 and 0.37 (l/s) have been applied to the media. Pre-calibrated EC sensors were installed at the distances of 0.36 and 1.1m from media entrance, connecting to the data logger with designed software system. Data recording interval was 4 seconds and solution of sodium chloride have been injected instantly at the upstream of the media and simultaneously the data recording has begun. The different parts of the experimental setup has been presented in figure 1.

3. Results and Discussion

All of the experimental BC curves were zeroed with base concentration of the flowing water through the porous media. Consequently, the results of experiments for both 1st and 2nd sensors which were positioned at distances of 0.36m and 1.1m respectively have been displayed in the tables of 1 and 2. The range of longitudinal dispersion coefficient is between 2 to 25 (cm²/s) and the magnitudes of degradation rate is in the range of 0.01 to 0.05 (1/s). Generally the computed dispersion coefficients and degradation rate (extracted from both least square curve fitting and temporal moments method) for first sensor is greater than second one which exhibit the spatial variation of the above-mentioned parameters. Furthermore, it is observed that by increase in pore flow velocity due to increase of media diameter or entrance discharges, the dispersion coefficient and degradation rate of tracer increases. This is due to the enhancing of the mechanical dispersion inside of the large porous media. The average difference between the quantities of for the dispersion coefficients which were computed from two different methods is about 33%, presenting any considerable differences, but the values of the degradation rate show significant differences. Increase in tracer injection mass dose not shows any special trend in the dispersion coefficient but the values of degradation rate which were extracted from curve fitting method has shown a reverse order with injection mass. Furthermore, degradation rates that

were computed from method of moments have displayed somewhat a constant value. Recovery percentage of injected mass from experimental BC curves is in direct relation with injection mass. in other words, with increase in injection mass, the recovered mass through the BC curves increases and inversely

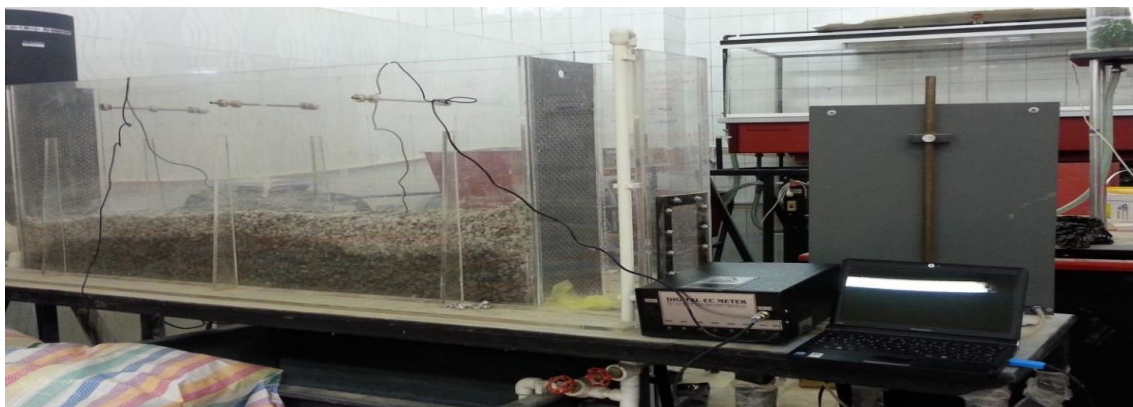
degradation rate decreases. It is also noteworthy to mention that changing of all parameters which would increase pore velocity inside of the large porous media would enhance the mechanical dispersion and consequently the large values of the dispersion coefficients would be exhibited.



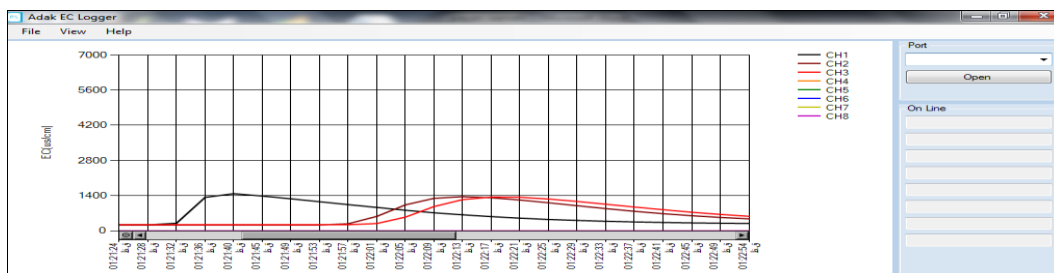
(B)



(A)



(C)



(D)

Fig. 1. Laboratory flume (C), EC meter sensors (A), Data accusation system (B), Designed software (D).

Table 1. Extracted parameters for 1st sensor from curve fitting method to the analytical solution and temporal moments method.

Media diameter (cm)	Discharge (l/s)	Injection mass (mgr)	Recovery * (%)	Dispersion coefficient (cm ² /s)	Dispersivity (cm)	Degradation rate (1/s)	Dispersion coefficient (cm ² /s)	Dispersivity (cm)	Degradation rate (1/s)
				CF ²	CF	CF	MOM ³	MOM	MOM
1.8	0.26	5000	31.42	8.677	5.903	0.0414	9.0452	5.6540	0.0400
1.8	0.26	10000	45.92	5.523	3.757	0.0283	5.3246	3.8262	0.0357
1.8	0.26	15000	51.80	9.231	6.280	0.0213	7.0506	4.7240	0.0378
1.8	0.26	25000	68.11	5.381	3.660	0.0139	5.1856	3.8254	0.0348
1.8	0.26	50000	93.15	5.213	3.546	0.0033	5.1343	3.7793	0.0349
1.8	0.37	5000	30.81	6.486	3.612	0.0533	8.1416	4.3193	0.0480
1.8	0.37	10000	45.05	6.026	3.355	0.0358	7.0977	4.2527	0.0425
1.8	0.37	15000	47.18	10.266	5.716	0.0307	8.4140	4.8258	0.0441
1.8	0.37	25000	56.66	19.851	11.053	0.0198	12.1302	5.9038	0.0512
1.8	0.37	50000	74.03	10.571	5.886	0.0123	8.9427	5.0197	0.0449
1.1	0.26	5000	30.20	2.933	1.968	0.0510	5.5993	4.2459	0.0336
1.1	0.26	10000	43.45	6.124	4.110	0.0308	5.7696	4.1893	0.0351
1.1	0.26	15000	51.06	5.875	3.943	0.0251	5.0205	3.8412	0.0335
1.1	0.26	25000	69.63	5.398	3.623	0.0137	5.2043	3.9860	0.0334
1.1	0.26	50000	92.42	4.430	2.973	0.0039	4.2947	3.6022	0.0307
1.1	0.37	5000	44.20	3.212	2.007	0.0350	3.6364	2.4167	0.0396
1.1	0.37	10000	59.57	6.642	4.151	0.0192	7.0176	4.6978	0.0378
1.1	0.37	15000	61.87	7.260	4.537	0.0176	5.8664	3.9552	0.0380
1.1	0.37	25000	71.72	5.656	3.535	0.0136	4.7959	3.5657	0.0347
1.1	0.37	50000	97.44	5.777	3.611	0.0021	5.6281	4.0493	0.0355

*Recovery percentage is calculated by dividing of the recovered mass by experimental BC curve to the injection mass.

Table 2. Extracted parameters for 2nd sensor from curve fitting method to the analytical solution and temporal moments method.

Media diameter (cm)	Discharge (l/s)	Injection mass (mgr)	Recovery (%)	Dispersion coefficient (cm ² /s)	Dispersivity (cm)	Degradation rate (1/s)	Dispersion coefficient (cm ² /s)	Dispersivity (cm)	Degradation rate (1/s)
				CF	CF	CF	MOM	MOM	MOM
1.8	0.26	5000	34.46	18.696	7.922	0.0215	24.2937	9.6010	0.0215
1.8	0.26	10000	43.38	10.565	4.477	0.0166	10.7374	4.3397	0.0217
1.8	0.26	15000	54.95	16.785	7.112	0.0122	36.4640	16.0166	0.0187
1.8	0.26	25000	57.02	11.869	5.029	0.0111	11.3712	4.7388	0.0210
1.8	0.26	50000	73.45	11.857	5.024	0.0062	12.0859	5.0310	0.0210
1.8	0.37	5000	22.08	20.823	7.545	0.0353	25.4458	8.0742	0.0270
1.8	0.37	10000	35.96	16.076	5.825	0.0233	17.0280	5.5472	0.0267
1.8	0.37	15000	42.04	21.033	7.621	0.0192	19.6074	6.2630	0.0271

²) Curve Fitting

³) Method of moments

1.8	0.37	25000	46.52	32.644	11.828	0.0160	19.6462	5.8572	0.0291
1.8	0.37	50000	60.27	22.631	8.200	0.0110	17.6963	5.7354	0.0268
1.1	0.26	5000	17.53	7.029	3.138	0.0377	14.3434	6.7434	0.0184
1.1	0.26	10000	29.83	10.653	4.756	0.0237	13.7571	5.9007	0.0203
1.1	0.26	15000	35.84	10.303	4.599	0.0200	11.7250	5.1558	0.0199
1.1	0.26	25000	42.05	10.697	4.775	0.0168	11.5724	5.2019	0.0194
1.1	0.26	50000	54.23	8.902	3.974	0.0125	10.9006	5.1183	0.0186
1.1	0.37	5000	21.48	7.645	3.058	0.0353	14.2303	5.5916	0.0222
1.1	0.37	10000	38.90	12.764	5.106	0.0205	24.6519	9.5134	0.0220
1.1	0.37	15000	39.43	12.481	4.992	0.0201	15.2303	5.9076	0.0224
1.1	0.37	25000	43.88	11.285	4.514	0.0184	12.1847	5.0363	0.0211
1.1	0.37	50000	52.10	12.371	4.948	0.0146	13.6492	5.6433	0.0210

The figures of 2(a, b) are indicating some examples of experimental and analytical BC curves which have been conducted at the two above mentioned positions. As is presented, by moving along the large porous media, the dispersion and degradation processes have acted and the downstream curves have indicated a lower magnitude in climax point. In addition to the experimental curves, analytical curves by applying the dispersion and degradation coefficients, which were exploited from both above-mentioned methods, have been depicted. As a conclusion it can be implied that both of the methods is beneficial and can be applied to the tracer tests. Furthermore, the depicted curves indicates asymmetry between two limbs of the BC curves because of transient storage of tracer inside the media and gradually outgoing from that.

The sensitivity analysis for 1st to 3rd temporal moments and skewness coefficient have been arranged. 4 parameters of velocity, degradation rate,

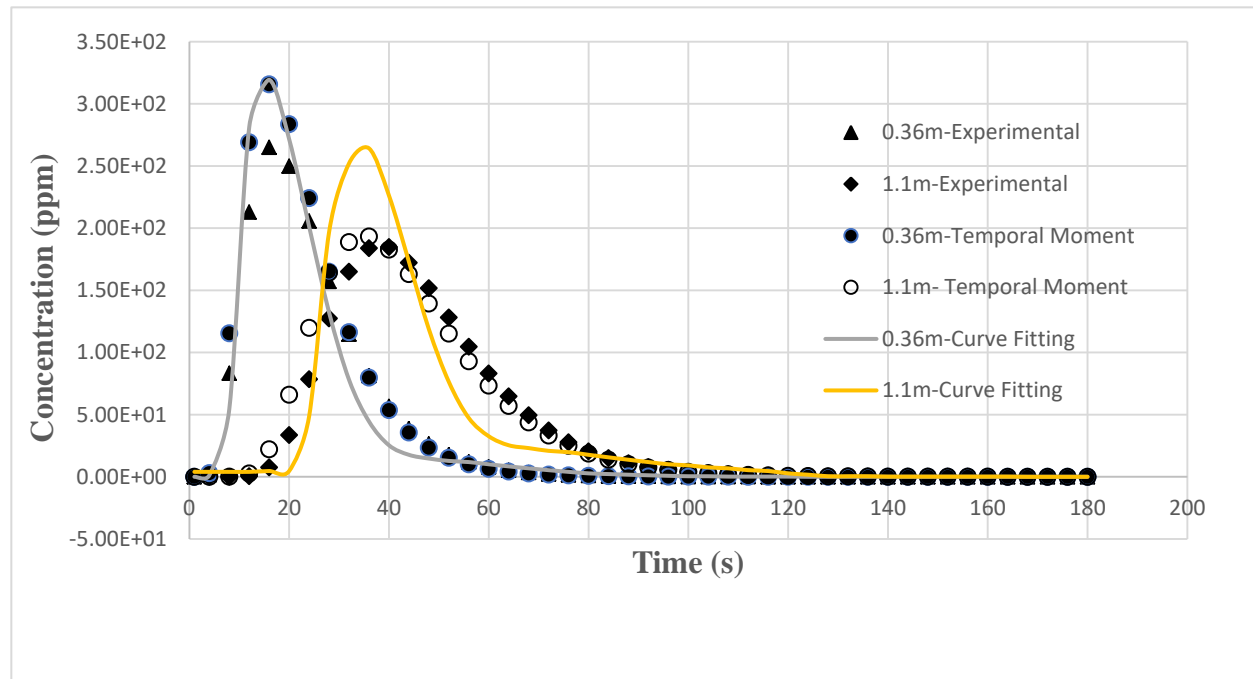
dispersion coefficient and length scale were chosen as variables of the moments. The real magnitudes for mentioned parameters were selected to examine the sensitivity of them in the moment equations. Since all the parameters are in different dimensions, the normalized form of them have been computed in order to arrange them in the limited domain. The normalization process were accomplished applying equation of (25).

$$x_n = \frac{(x_i - \bar{x})}{std(x_i)} \quad (25)$$

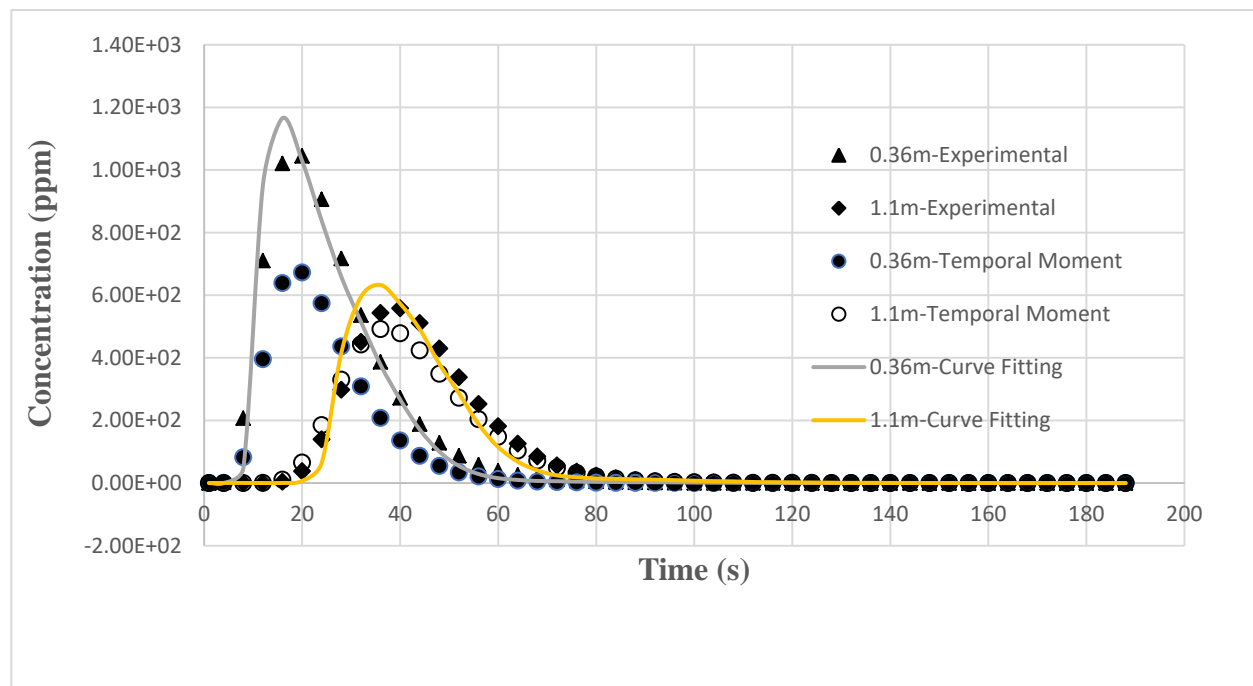
Where: the x_i is the real parameter, \bar{x} is the mean of parameter and $std(x_i)$ is the standard deviation of the parameter.

The figures of the sensitivity analysis is illustrated in Fig. 3(a) to (d). Among the examined parameters, the pore velocity has the most important and reverse role in the moment equations. Since all the moments change very intensely by increasing of the pore velocity. Increasing of the length scale exhibits a linear increasing tendency for all the moments

but in skewness coefficient, it operate a reverse none-linear tendency.



(a)



(b)

Fig. 2. Experimental and analytical BC curves for a) injection mass equal to 5000 (mgr), median diameter of media $d=1.8$ cm and entrance discharge equal to 0.26 (l/s) b) Injection mass equal to 15000 (mgr), median diameter of media $d=1.1$ cm and entrance discharge equal to 0.37 (l/s).

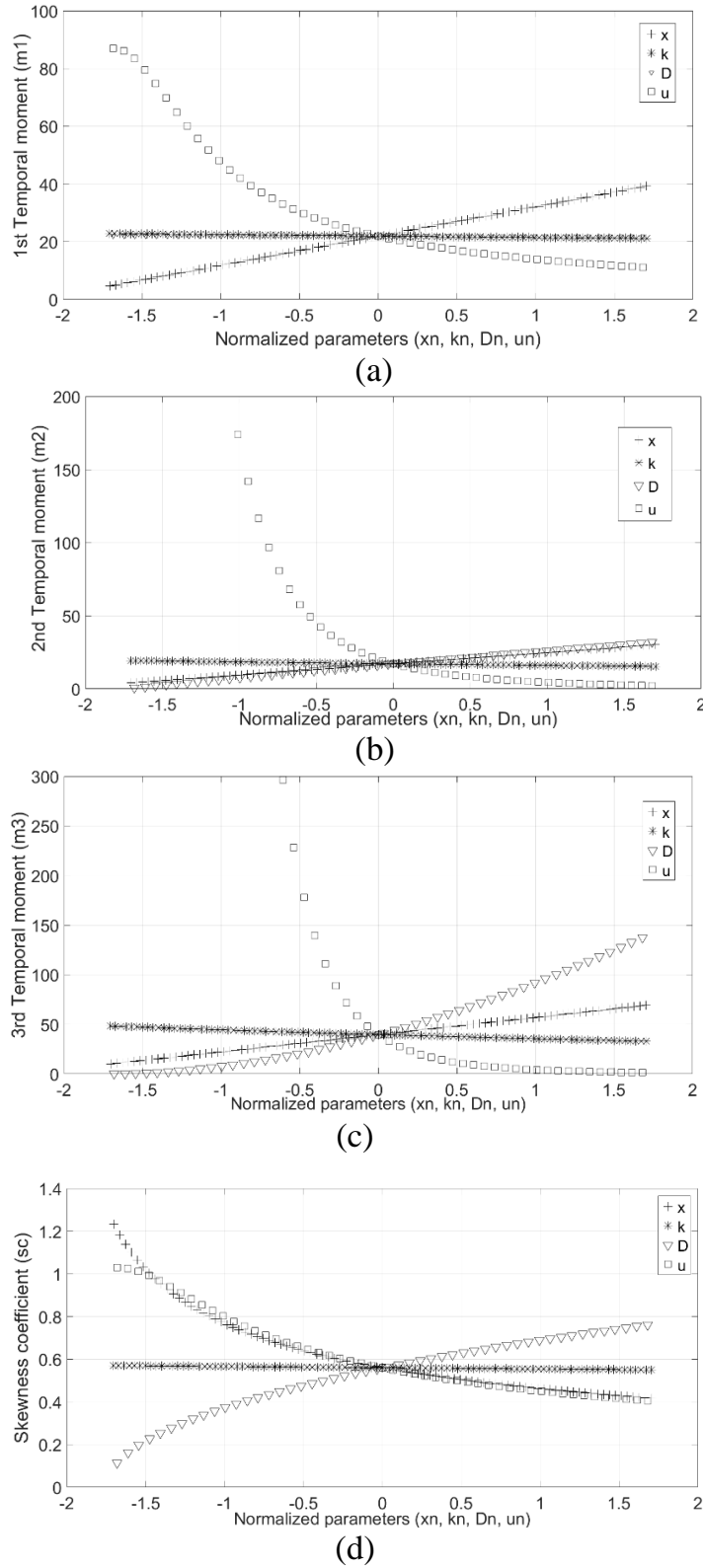


Fig. 3. Sensitivity analysis for moment equations a) 1st temporal moment versus normalized parameters b) 2nd temporal moment versus normalized parameters c) 3rd temporal moment versus normalized parameters d) skewness coefficient versus normalized parameters.

The decay rate reveals a gentle decreasing manner in the all moment and skewness coefficient equations such that it has the lowest impact on the model equations. The parameter of the dispersion coefficient has very interesting role in which in the 1st and 2nd moment equation it has linear trend but decreasing in the 1st and increasing in the 2nd moment. For 3rd moment equation, the dispersion coefficient shows non-linear increasing manner. Moreover, in the skewness coefficient equation, non-linear increasing manner with respect to the dispersion coefficient has been observed.

4. Summary and Conclusions

Sometimes, the injected mass as tracer in different type flow fields cannot be recovered totally. Consequently, the reactive equations is beneficial for application. In this research, the theoretical temporal moments were derived from direct integration from analytical equation of the advection-dispersion including first-order reactive term. The magnitudes of the decay rate and dispersion coefficients have been extracted for one experimental data series applying two different methods named least square curve fitting and temporal moment. The outcomes reveals proficiency of the both of these methods. Notwithstanding, the rebuilt breakthrough

curves by the coefficients of LSCF shows a better agreement with the experimental BC curves. The results revealed that the magnitudes of the both of the mentioned coefficients illustrated spatial variation along the porous media. All geometrical and hydraulically parameters which enhances mechanical dispersion processes causes enhancing in the both of the coefficients. The sensitivity analysis were demonstrated for the obtained moment equations by changing of the 4 involved parameters. It is concluded that the velocity parameter is the most effective parameter among others which shows a reverse effect in the different order moment equations. Furthermore, the degradation rate is the less effective parameter in the all the moment and skewness coefficient equations which indicate reverse linear decreasing manner.

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