Spatial and Temporal Moment Approaches to Quantify Laboratory-Scale Macrodispersion in Stratified Porous Formations

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Abstract: In this study, intermediate scale dye tracer experiments were conducted in a 1 m length, 1 m height and 0.03 m thickness sandbox to quantify macrodispersion phenomena in stratified porous formations and to elucidate the effect of the layering on the degree of macrodispersion. Spatial and temporal moment approaches based on a snapshot of tracer distribution and NaCl concentration at a point, respectively, were applied to identify the macrodispersivities in longitudinal and lateral directions. The experimental results indicated that the longitudinal macrodispersivity depends on the travel distance of solute and the degree of heterogeneity in stratified formations with two or four layers, while the transverse macrodispersivity decreases with the increase of travel distance and depends on the magnitude of the initial solute distribution. A difference between longitudinal macrodispersivity estimates using spatio-temporal moments was also clarified.

Keywords: Macrodispersion; Stratified porous formation; Spatial and temporal moments; Image analysis; Intermediate scale experiment

1 Introduction

Contamination of groundwater resources has been one of the major environmental concerns during the past few decades due to concerns on adverse effect on surrounding environment and public health. Management of contaminated groundwater resources has been a difficult challenge despite of the degree of contamination because of the limited resources that can be committed to remediate a large number of contaminated sites (He *et al.*, 2008). In addition, the observation scale issue regarding dispersion phenomena in natural aquifers has been observed to occur at much larger dispersion of contaminants than that observed for homogeneous porous media under laboratory conditions, indicating that larger rates of solute dispersion in fields are related to the heterogeneities of natural formations (Rajaram and Gelhar, 1993).

Physically dominant mechanism of dispersion of contaminants in aquifers seems to be the spatial variability of groundwater flow velocities associated with primarily with the spatial distributions of hydraulic conductivity. This mechanism is commonly referred to as macrodispersion (Dagan, 1984; Chrysikopoulos *et al.*, 1992; Gelhar *et al.*, 1992) despite of the conservative and non-conservative solutes. Understanding the characteristic behind such transport pathways related to macrodispersion is a crucial step to ensuring the reliable prediction of a contaminant behavior (Uffink, 1985; Tompson, 1993). Various experimental studies have been extensively carried out to investigate the mechanisms of solute

movement in porous media (Guedes de Carvalho and Delgado, 2003; Jones and Smith, 2005; Inoue *et al.*, 2011). However, many applications involving long-term predictions for regional flow systems and large scale tracer experiments to estimate dispersion coefficients are impractical because of not only the limitation in space and time but physical and social apprehensions.

Macrodispersivities of solutes are often identified in the field by monitoring breakthrough curves at a limited number of observation wells (Uffink, 1990; Adams and Gelhar, 1992). However, in general, interpretation of breakthrough curves obtained in a field tracer test even in a laboratory tracer test is extremely difficult to infer the transverse dispersivity (Olsson and Grathwohl, 2007). Contrary to the temporal moments, a snapshot of a cloud of solute particles in time expresses the dispersive variation in the aquifers, providing the estimates of spatial moments in the lateral direction as well as in the longitudinal direction under the prescribed heterogeneity and stratification of hydraulic conductivity of the geologic medium (Fadili *et al.*, 1999; Fernández-García *et al.*, 2008; Inoue *et al.*, 2010).

The main model problem employed in this paper is the case of solute transport in multilayered porous formation, which is a common problem in practical and theoretical concern. The objectives of this study are to identify the longitudinal and transverse macrodispersivities in the laboratory using a non-invasive technique in conjunction with a dye tracer and to elucidate the solute transport behavior in two-dimensional stratified porous formations under saturated conditions. Spatial and temporal moment approaches linked with image data of a dye tracer behavior and variations of NaCl concentration were utilized to rely on the difference between the two approaches. Although perfect layering over large horizontal distances is an idealization which is quite

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Table 1: Physical properties of sand materials

	K3	K4	K5	K7
Mean particle size (cm)	0.15	0.081	0.48	0.011
Uniformity coefficient (—)	2.10	1.65	1.34	1.25
Porosity (—)	0.42	0.42	0.42	0.42
Hydraulic conductivity at 15°C (cm/s)	1.47	0.751	0.224	0.0334

Figure 1: Experimental setup of intermediate-scale dye tracer experiments

improbable in natural aquifers, the model may apply to flow and transport at short distances and serve as a simple tool to interconnect the findings between the field- and laboratory-scale dispersion phenomena even in laboratory-scale phenomena.

2 Intermediate-scale tracer experiments

2.1 Materials

Dye tracing technique has been widely used for many years in order to characterize water flow and/or solute transport behavior in porous media in a laboratory or in a field (Aeby *et al.*, 2001; Vanderborght *et al.*, 2002; Citarella *et al.*, 2015). For the purpose of visualization of solute transport phenomena, in this study, Brilliant Blue FCF, which is a synthetic color having the nature of readily visible in soils, moderate mobile, and low toxicity (Flury and Flühler, 1995; Forrer *et al.*, 1999), was used as a dye tracer. NaCl was also mixed with dye tracer solution of Brilliant Blue FCF in order to measure the solute concentration of NaCl in a flow field. The initial concentrations of Brilliant Blue FCF and NaCl were adjusted to 0.40 mg/cm³ and 1.0 mg/cm³, respectively. The initial concentration of dye tracer was determined to be low enough to avoid density-induced flow effects. In this study,

four types of silica sand materials having a small uniformity coefficient were employed to comprise a few stratified porous formations. Physical properties of these sands are summarized in Table 1.

2.2 Experimental apparatus and experimental cases

Two-dimensional solute transport experiments were carried out in a quasi two-dimensional and vertically placed water flow tank with the dimensions of 100 cm width, 100 cm height and 3 cm thickness. The water flow tank consisted of the glass plate and the perforated acrylic plate to allow the visualization of stratified porous formations and dye tracer behavior while monitoring points to observe the piezometric heads and NaCl concentrations were established. Two constant head reservoirs were connected to the water flow tank at the both sides and were used to adjust the hydraulic gradient. Schematic diagram of experimental apparatus is shown in Figure 1.

In tracer experiments, in order to elucidate the effect of the layering on the degree of solute macrodispersion in both the longitudinal and lateral directions, six stratified porous formations were of interest and were comprised using different types of soil materials to reflect two-layered or four-layered porous formations. In two-layered porous formations, Layer 2 and Layer 3 shown in Figure 1 were filled with two types of soil materials according to the combination of K4 and K5, K5 and K7 and K7 and K3. These three types of porous formations were referred to as “two-layered type” and were denoted as K4-5, K5-7 and K7-3. In a similar manner, four-layered porous formations were consisted of four distinct layers of soil materials and were also referred to as “four-layered type”. In this study, all soil materials were filled from Layer 1 to Layer 4 with the descending order of the mean particle size, which was expressed as K3-4-5-7. In addition, different allocation of all soil materials, which was shown as K3-5-7-4, was also employed. As the third porous formation with four layers, K4-7-3-4 was employed to compare the degree of solute dispersion under the replacement of soil materials comprising the layering. In Table 2, experimental cases and the corresponding layering formations are listed as well as the assigned injection ports of the solution with dye and NaCl. Addition to these multilayered formations, as the base cases of solute transport in homogeneous porous media, each soil material comprised a homogeneous porous formation. Despite of the experimental cases, the porosity not only of the each layer but the entire flow domain was adjusted to 0.42 during the soil packing.

Table 2: Experimental cases

Case	Injection port(s)
<i>Homogeneous types</i>	
K3	c
K4	c
K5	c
K7	c
<i>Two-layered types</i>	
K7-3	b ~ d
K4-5	b ~ d
K5-7	b ~ d
<i>Four-layered types</i>	
K3-4-5-7	a ~ e
K3-5-7-4	a ~ e
K4-7-3-4	a ~ e

2.3 Experimental methodology

Soil materials were completely washed and saturated before packing to avoid entering air and to conduct experiments under the saturated condition. During the process of soil packing, the water flow tank was filled with water and soil material of concern from the bottom toward the top of the water flow tank in 6 cm or 4.5 cm layers. As shown in Figure 1, the soil material K4 was filled in 6 cm layers up to 12 cm height from the bottom through the process of compaction prior to filling the next layer. Subsequent four layers whose area was the primal stratified area were comprised by prescribed soil materials in 4.5 cm layers. In order to reduce the effect of regional boundary on the solute transport phenomena, prescribed soil materials were packed into the middle part in each layer while the material K4 was filled into the upstream and downstream sides in each layer. When filling soil materials at the boundary between the middle and upstream or downstream parts of the layer, two thin boards were used to avoid the mixture of different materials. The rest of the upper area was filled by K4 in the same manner as the other areas in 6 cm layers.

While maintaining saturated condition of porous media, water was applied to the flow tank under a specific hydraulic gradient controlled by constant head water reservoirs connected to the upstream and downstream sides. A steady saturated flow field was established in the flow tank when fluctuations in the observed drainage rate, which was effluent from the constant head water reservoir at the downstream side, and piezometer reading at water pressure measurement points could become negligible. After steady state conditions had been established, a needle is inserted through the injection port with 0.5 cm of radius on the face of acrylic plate in order to create a two-dimensional transport state. The solutions with dye mixed with NaCl was released from three injection ports of b, c and d, which are depicted in Figure 1, in “two-layered type”, or five injection ports from a to e in “four-layered type” porous formations, respectively. The solution with 25 cm³ in each injection port was injected for 30

Figure 2: Representative images of dye tracer transport in K3-4-5-7

seconds to construct a pulse source. During the experiment, the profiles of solute migration were recorded using a digital camera approximately 100 cm located away from the front side of the water flow tank. A series of transport experiments were conducted in a constant temperature room at 25°C under the water temperature condition of $21 \pm 2^\circ\text{C}$.

2.4 Relation between dye concentration and image intensity

Figure 2 shows a representative image obtained during the tracer experiments in four-layered porous formation. Tracer experiments may be regarded as effectively two-dimensional since dye tracer was injected across the full 3 cm thickness of the flow tank. Each of the pixels representing an image has a pixel intensity which describes how bright the corresponding pixel is. Data recorded by the digital camera successfully exhibited different pixel intensities in dye tracer distributions, showing different concentration variations within the dye tracer distribution. Moreover, image processing techniques used in this study relate the color intensity of solutes to the dye concentration in the porous media. Under the same light condition in the laboratory, calibrations were performed and analyzed with solutes of known concentrations of dye in order to establish a relationship between the color intensity and dye concentration. Established piece-wise regression curve is shown in Figure 3. A few studies pointed out that non-linear variation of the relation between the color intensity and dye concentration, which depends on the soil brightness and the dye, appears and has little effect on the subsequent estimation process (Huang *et al.*, 2002; McNeil *et al.*, 2006; Inoue *et al.*, 2011).

3 Quantification of the macrodispersivity

3.1 Spatial moment approach

Spatial moment techniques have a potential to provide insight into a wide range of solute transport processes (Tomp-

Figure 3: Relation between the pixel intensity and the dye tracer concentration

son and Gelhar, 1990; Fernández-García *et al.*, 2005b). In this study, spatial moments of dye concentrations distributed in space are calculated based on digital images at given times as follows:

$$M_{ij}(x, z, t) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} c(x, z, t) x^i z^j dx dz \quad (1)$$

where x and z are the Cartesian coordinates, which stand for the horizontal and vertical directions, respectively. Also, c is the solute concentration, t is the time, M_{ij} is the spatial moments associated with the dye distribution at a certain time, and i and j are the spatial order in the x and z coordinates, respectively. Based on the regression curves relevant to the dye concentration and pixel intensity, the pixel intensity distribution can be converted to a concentration distribution by the calibration, providing an analogy between Eq.(1) and Eq.(2) (Inoue *et al.*, 2011)

$$M_{ij}(x, z, t) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} H(x, z) I(x, z, t) x^i z^j dx dz \quad (2)$$

where $H(x, z)$ is the area per unit pixel and $I(x, z, t)$ is the intensity at a corresponding pixel. The centroid of concentration distribution is calculated as the normalized first-order spatial moment.

$$X_G(x, z, t) = \frac{M_{10}}{M_{00}}, \quad Z_G(x, z, t) = \frac{M_{01}}{M_{00}} \quad (3)$$

where X_G and Z_G are the centroid locations of plume concentration distribution in the x and z coordinates, respectively. Longitudinal and transverse macrodispersivities from spatial moments of the distributed tracer plume are calculated as (Inoue *et al.*, 2011)

$$A_L(x, z, t) = \frac{\frac{M_{20}}{M_{00}} - X_G^2}{2\xi_c}, \quad A_T(x, z, t) = \frac{\frac{M_{02}}{M_{00}} - Z_G^2}{2\xi_c} \quad (4)$$

where A_L is the longitudinal macrodispersivity, A_T is the transverse macrodispersivity and ξ_c is the travel distance of the center of tracer plume in the mean flow direction at a given time t . This approach is applicable not only to conservative solutes but to reactive tracers.

3.2 Temporal moment approach

The temporal moment approach was used to characterize the breakthrough data at all NaCl measurement locations. The j th normalized absolute temporal moments is very useful descriptors of breakthrough curves (BTCs). The first-order temporal moment corresponds to the mean arrival time of solute, while the second-order temporal moment is an analogy with the statistical dispersion (Fernández-García *et al.*, 2005b)

$$\mu_j(x, z, t) = \frac{\int_0^{\infty} t^j c_m(x, z, t) dt}{\int_0^{\infty} c_m(x, z, t) dt} \quad (5)$$

where $\mu_j(x)$ is the j th normalized absolute temporal moments, $c_m(x, z, t)$ represents the time variation of the NaCl concentrations at the monitoring locations and j is the non-negative integer corresponding to the order of concern. Also, the j th central temporal moments around the mean can be computed based on the first normalized absolute temporal moments

$$\mu'_j(x, z, t) = \frac{\int_0^{\infty} (t - \mu_1(x, z, t))^j c_m(x, z, t) dt}{\int_0^{\infty} c_m(x, z, t) dt} \quad (6)$$

where $\mu'_j(x)$ is the j th central temporal moments.

Longitudinal macrodispersivity using temporal moments is calculated as:

$$A_L(x, z, t) = \frac{\zeta_P}{2} \frac{\mu'_2(x, z, t)}{(\mu_1(x, z, t))^2} \quad (7)$$

where A_L is the longitudinal macrodispersivity and ζ_P is the distance from the source of solute injection. This approach as well as spatial moment approach is also applicable not only to conservative solutes but to reactive tracers.

4 Results and discussion

4.1 Quantification of the longitudinal macrodispersivity

Longitudinal macrodispersivity values obtained from spatial moments in each porous formation are shown in Figure 4 as a function of the displacement distance of plume centroid from the source D_G , which is expressed as:

$$D_G(x, z, t) = \sqrt{X_G^2 + Z_G^2}. \quad (8)$$

In this figure, longitudinal macrodispersivity values identified by the temporal moment approach at the corresponding observation point from the initial release source of solute are also exhibited, which will be described later. Longitudinal macrodispersivity estimates in homogeneous porous formations remain constant during the solute transport, whereas longitudinal macrodispersivity estimates in two- or four-layered porous formations increase with the increase of the displacement distance of solute. This is attributed to the spatial distribution of hydraulic conductivity comprising flow fields, expresses the scale effect of solute transport (Dagan,

Figure 4: Longitudinal macrodispersivity variation identified with spatial and temporal moment approaches

Figure 5: Transverse macrodispersivity variation identified using the spatial moment approach

1984; Fernández-García *et al.*, 2005a) and exhibits the evidence occurring macrodispersion phenomena in heterogeneous porous formations. Inherently, longitudinal macrodispersivity in heterogeneous porous media becomes larger than that in homogeneous porous media due to the variation of seepage velocity in a space (Dagan, 1984; Inoue *et al.*, 2010), leading to the distinct definition of microscopic and macroscopic dispersion phenomena, or micro- and macro-dispersion phenomena. Thus, these results shown in Figure 4 demonstrate the validity of estimates in spatial moments under controlled laboratory conditions.

In two-layered types, longitudinal macrodispersivity values identified with spatial moments in Case K5-7 are the lowest estimates among three experimental cases, while Case K7-3 provides the highest values of the longitudinal macrodispersivity. In general, the degree of solute dispersion depends on the particle size and the heterogeneity of flow field, which reflects the hydraulic conductivity distribution (Tompson, 1993; Xu and Eckstein, 1997). In Case K7-3, the larger difference of the hydraulic conductivity between two soil materials than other cases results in the largest estimates of the longitudinal macrodispersivity. On the other hand, Case K5-7 involves the lowest two soil materials associated with the mean particle size, leading to the lowest estimates among three experimental cases. It is particularly worth noting that Case K4-5 with a slight difference of the hydraulic conductivity and mean particle size between two materials exhibits the increase tendency.

As for the four-layered types, the results of Case K3-4-5-7 and Case K3-5-7-4 obtained from spatial moments show similar values. This is because these two cases have the same heterogeneity. Although the allocation of soil materials is

different from each other, solute released from five injection ports moves an entire region of four-layered stratified formations. This means that the degree of solute dispersion along a regional flow in Case K3-4-5-7 is identical to that in Case K3-5-7-4. Contrary to this finding, the heterogeneous flow field in Case K4-7-3-4 where soil material K5 is replaced with K4 provides smaller estimates. This point indicates the effect of porous formations comprising stratification, or the effect of the heterogeneity of a flow field and is consistent with previous modeling and laboratory results (Uffink, 1985; Inoue *et al.*, 2011).

Besides the estimates obtained from spatial moments, longitudinal macrodispersivity estimates identified with temporal moments using breakthrough curves at NaCl measurement locations are also plotted in Figure 4. In homogeneous types, longitudinal macrodispersivity estimates using temporal moments are almost identical to the macrodispersivity estimates obtained from spatial moments. This may suggest that single monitoring point leads to a plausible result in homogeneous porous formations if a direction from a solute injection port to a monitoring point is parallel to the regional flow direction. On the other hand, in two-layer types except for K4-5, a marked difference between longitudinal macrodispersivity estimates from spatial and temporal moments appears. The difference becomes larger with the increase of the number of layers. This indicates that single monitoring location is insufficient to capture the primal characteristics of solute transport as breakthrough curves and to identify a reliable estimate. Hence, transect allocation of monitoring points perpendicular to a regional flow direction may be required to capture solute migration passing through a plane or a line connecting several monitoring points in

Figure 6: Schematic drawing of initial location of solute and its initial length, L_z , in the z direction in each layered type

heterogeneous porous formations (Fernández-García *et al.*, 2005b).

4.2 Quantification of the transverse macrodispersivity

The results of transverse macrodispersivity estimates obtained from spatial moments are shown in Figure 5 as a function of the displacement distance of solute. The absolute values of the transverse macrodispersivity show a dependency on the number of layers. This is attributed to the geometrical length in the z direction at the initial release time of solute. In other words, the number of injection ports employed in the solute release affects the variation of transverse macrodispersivity estimates. Figure 6 expresses the differences of initial location and geometrical length of solute. In this study, each porous formation with own number of layers has own initial solute length, L_z , in the z direction because the solute must migrate in an entire region in stratified porous formation when quantifying and obtaining reliable macrodispersivity estimates in the corresponding heterogeneous flow field (Gelhar *et al.*, 1992; Rahman *et al.*, 2005). Therefore, the values of the second order spatial moments in the z direction, σ_{zz} , are affected by the initial source length, L_z , and are anticipated to become larger associated with the larger source as the number of layers increases.

In Figure 7, the results of the variation of second order spatial moments in the z direction, σ_{zz} , are shown as a function of elapsed time for three representative cases in each layer system. As aforementioned above, the second order spatial moments in the z direction, σ_{zz} , not only depend on the number of layers but remain constant despite of the number of layers. This means that the values of the transverse macrodispersivity differ from the number of layers and exhibit a decrease tendency as solute migrates because the travel distance of the center of dye tracer plume in the mean flow direction in Eq. (4) increases. Consequently, in all experimental cases as shown in Figure 5, the transverse macrodispersivity exhibits a strong dependency on the number of layers and the decrease tendency during the course of solute transport although a slight variation of estimates among the experimental cases in the corresponding layered types appears. Also,

Figure 7: Variation of the second order spatial moments of σ_{zz} in each layered type

these results demonstrate that the effect of initial allocation of solute on the transverse macrodispersivity disappears and the transverse macrodispersivity asymptotically will reach a certain value of the transverse dispersivity at the microscopic level.

4.3 Longitudinal macrodispersivity versus the degree of heterogeneity

Spreading of solutes in advective transport in heterogeneous porous formations is governed by spatial variations of the hydraulic conductivity. Mercado (1967) suggested one relation between macrodispersivity and spatial variations of the hydraulic conductivity based on the theory for the perfectly stratified porous media in the form as

$$A_L = \frac{1}{2} \left(\frac{\sigma_K}{\bar{K}} \right)^2 \bar{x} \quad (9)$$

where σ_K is the standard deviation of hydraulic conductivity and shows the degree of heterogeneity, $\bar{K}$ is the mean of hydraulic conductivity, $\bar{x}$ is the travel distance of the center of tracer plume in the mean flow direction. For all results in each porous formation, the variation of the longitudinal macrodispersivity is shown in Figure 8 as a function of the degree of heterogeneity. In this figure, the variation of longitudinal macrodispersivity based on the theory of Mercado (1967) is also depicted. The experimental estimates are smaller than the Mercado's results given by Eq. (9). In Mercado's model, primary cause of solute transport was limited to the advection, leading to the elongation of solute spreading along a flow direction. In addition, a physical expression of solute mixing passing through different layers was not involved in Eq. (9). These points provide the difference between the experimental and Mercado's results. On the other hand, there is a non-linear increase and the longitudinal macrodispersivity will approach a constant value. This finding reflects the outcome of Gelhar *et al.* (1992) and Fiori *et al.* (2010), suggesting the reliability of experimental estimates.

Figure 8: The variation of longitudinal macrodispersivity with the degree of heterogeneity

5 Conclusions

In this study, the behavior of macrodispersion in stratified porous formations was assessed through intermediate-scale solute transport experiments. Longitudinal and transverse macrodispersivities were identified using spatial and temporal moment approaches based on the dye tracer distribution and NaCl concentration, respectively. The following findings have been clarified.

1. Image processing was a non-intrusive approach, was able to directly characterize the solute movement in porous media and allowed to link with the spatial moment approach.
2. At larger displacement of solute, the macrodispersivity estimates obtained from spatial moments of the distributed dye tracer plume in two- and four-layer porous formations were larger than those in homogeneous porous formations.
3. The experimental results revealed that the macrodispersivity estimates in two- and four-layer types exhibit the increase tendency with the increase of the travel distance, or the scale effect.
4. In homogeneous porous formation, longitudinal macrodispersivity estimates from temporal moments were almost identical to the macrodispersivity estimates obtained from spatial moments at a single measurement point. On the other hand, in two-layered porous formations, there was a marked difference between longitudinal macrodispersivity estimates derived from spatial and temporal moments, showing the limitation using only one point observation in temporal moment approach.
5. Longitudinal macrodispersivity depends on the degree of heterogeneity even in the laboratory scale transport, while the layering shows less effect on the variation of the transverse macrodispersivity in each layered porous formation.

Acknowledgments

Part of this study was supported by JSPS Grants-in-Aid for Scientific Research Grant Number 23780246.

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Discussion open until June 30, 2017