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ON THE VALIDITY OF DIFFUSION/DISPERSION TESTS IN SOILS

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In the paper, the validity of dispersion/diffusion parameters identified in the column tests is discussed. Using the experimental data reported in the literature it is shown that in some cases, the continuum approach (the Fickian mode of transport) can not be applied to interpret the test results. In such cases the transport parameters obtained by fitting the experimental results to the macroscopic model (the advection-dispersion equation) are experiment-dependent and can not be considered as intrinsic parameters. In order to illustrate the problem, the calculations of the scale separation parameter introduced in the homogenization theory are performed in four particular cases.

NOTATIONS

$c(x,t)$	concentration of the pollutant,
D_m	molecular diffusion coefficient,
D	dispersion coefficient,
D^*	effective diffusion coefficient,
ERV	an elementary representative volume,
l	characteristic microscopic length,
L	sample length,
$\mathcal{L}$	characteristic macroscopic length,
Pe_m	molecular Peclet number,
P	Peclet number,
U	mean pore water (or seepage) velocity,
x, t	dimensional variables,
X, T	non-dimensional variables,
ε	homogenization (or scale separation) parameter,
ε_g	geometrical scale separation parameter.

1. INTRODUCTION

The evaluation of transport coefficients for the dispersion equation is commonly made through laboratory measurements. Such tests, as for example the classical column test, are typically performed with a view to applications in the field of environmental engineering for describing the pollutant migration in soils, or in the framework of the design of barriers for the waste storage.

It is an important point to realise that prior to the parameter identification, the choice of the dispersion equation as the relevant governing equation was done. It means that the model, consisting of the dispersion equation and the appropriate initial and boundary conditions, was chosen to be further applied in order to derive the approximate behaviour of a particular system, with a known precision of the modelling. Although it is beyond the scope of the paper, we would like to stress the importance of this stage. The selection of the model should be based on the estimations of the relative "weights" of different phenomena existing in the system. This, in turn, is impossible unless the analysis of the practical situation *in situ*, using all the information available about the current and the anticipated states of the system, is done.

In the process of parameter identification, the laboratory tests are carried out, in which the known initial and boundary conditions are imposed on the selected sample of the medium. It provokes the transport phenomena to occur, that means the propagation of the solute in the soil. The response of the system i.e. the concentration as a function of space and time variables is monitored. Then, the model coefficients are fitted so that the difference between the concentration predicted by the model and that observed in the experiment is sufficiently small.

In the paper, an important question related to the identification of transport parameters is posed. It is well known that the macroscopic models, like for example the diffusion/dispersion model, are valid, provided there exists a good separation of certain scales. In the experimental studies, soil samples are necessarily limited in size and are subjected to large concentration gradients, especially during the early stages of the tests. As a consequence, the scale separation weakens. Therefore, fitting the dispersion coefficient to the concentration profile obtained from the macroscopic model is questionable and might give results that will be experiment-dependent, which practically means that they will not be applicable in other conditions than the ones investigated. This problem is studied in the framework of the multiple scale expansion method which is often used to derive the macroscopic equivalent models for finely heterogeneous materials, such like porous media, composite materials etc., BENSOUSSAN *et al.* [8], SANCHEZ–PALENCIA [22], AURIAULT [2]. In order to make the paper more comprehensive, it is proposed to base the analysis on the general findings of this method, without going into details. As those results can be intuitively recognised, the reader does not need to be familiar with the homogenization technique. Nevertheless, the reader is encouraged to refer to the previous papers [5, 6].

The paper is organised as follows: in Sec. 2 the homogenization approach is briefly presented. Section 3 contains the analysis of three column tests while in Sec. 4 the effective diffusion experiment is investigated. Finally, Sec. 5 is devoted to the general discussion and the comparison between the deterministic and the stochastic approach.

The problem is of great practical importance and might have serious consequences for the conceptual preparation of the tests of identification of the transport parameters. The aim of this paper is also to give to the laboratory experiments a new aspect in view of the homogenization theory.

2. MODELLING OF DIFFUSION/DISPERSION IN SOILS BY MEANS OF THE CONTINUUM APPROACH

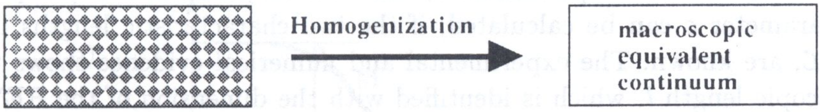

FIG. 1. Micro-macro passage.

When a diffusion/dispersion equation is applied to calculate the concentration of the solute in the soil column, it means that the continuum approach to the problem is adopted. According to this approach, each of the two phases present in the saturated soil, i.e. the solid matrix and the pore water in the void space, is treated as a continuum field that fills up the entire volume. The two interacting fields are superposed to form a macroscopic continuum model, for which the macroscopic equation, governing the diffusion/dispersion phenomena is derived, Fig. 1. The resulting model comprises the effective macroscopic parameters, for example the dispersion coefficient, that characterise the averaged properties of the model medium; they are to be determined in an experimental way. The main advantage of this approach is that the diffusion/dispersion problem does not have to be solved in the microscopic scale, for the complex pore geometry, that is, in almost all cases, practically impossible. Instead, we shift to the macroscopic problem in which the local scale is "forgotten". This upscaling process, which is called homogenization, requires one basic and necessary assumption to be satisfied, i.e. the existence of a representative elementary volume (REV) of the characteristic length l, very small with respect to the characteristic macroscopic length $\mathcal{L}$

$$l \ll \mathcal{L}. \tag{2.1}$$

In order to quantify the scale separation, and thus the applicability of the macroscopic model, a small homogenization parameter ε is defined

$$\varepsilon = \frac{l}{\mathcal{L}} \ll 1. \tag{2.2}$$

If the condition (2.2) is not satisfied, it is concluded that the soil cannot be treated as a continuum in the sense of the homogenization theory and the macroscopic model is not valid. Such "non-homogenizable" problems are frequently encountered in the engineering practice, though not always recognised.

On the other hand, ε and ε^2 measure the approximation of the equivalent macroscopic model for diffusion and dispersion, respectively. For the purpose of this paper, $\varepsilon \leq 0.1$ was chosen as a criterion of homogenizability of the problem. It means that we impose the approximation of the diffusion model to be less than 10%, and 1% for the dispersion model.

The essential problem related to the modelling of diffusion/dispersion in soils is the estimation of the parameter ε, that can be performed only *a posteriori*. The parameter ε can be calculated, if the two characteristic lengths involved, l and $\mathcal{L}$, are known. The experimental and numerical evidence shows that the microscopic length l, which is identified with the dimension of the REV, could be roughly approximated as (ANGUY *et al.* [1])

$$l \approx 10\delta, \tag{2.3}$$

where δ is the mean grain size or, more generally, the dimension of the heterogeneity. Obviously, the proper evaluation of l represents a great problem itself, depending on many different factors, such as for example the geometrical features of the medium in the microscale and the kind on the phenomenon being investigated. Nevertheless, for the purpose of the paper, the approximation (2.3) is assumed to provide a sufficiently good estimation of the order of magnitude of l. Note that this approximation fails, when the grain size distribution is relatively broad. The determination of the macroscopic length $\mathcal{L}$ is even less straightforward. The difficulty in the evaluation of $\mathcal{L}$ arises from the consideration that it is strongly dependent not only on a particular phenomenon (diffusion or/and dispersion) but also on the "intensity" of the phenomenon, expressed in terms of the macroscopic gradient of the concentration. As the latter varies in the sample and with time, it yields $\mathcal{L}$ being a function of position and time. To capture this problem, the reasoning proposed by BOUTIN and AURIAULT [9, 10] will be adopted. According to this method, in the case of the column test, $\mathcal{L}$ can be calculated from the macroscopic concentration field by the following formula:

$$\mathcal{L} = \left| \frac{\partial}{\partial x}\left(\frac{c}{c_0}\right) \right|^{-1}, \tag{2.4}$$

where c_0 means the concentration at the column inlet. The principle of this formula is presented in Fig. 2b. Note that $\mathcal{L}$ is generally different from the sample length L. It coincides with the sample length when the concentration gradient $\partial c/\partial x$ is constant all over the sample, Fig. 2a. Therefore, ε is generally different

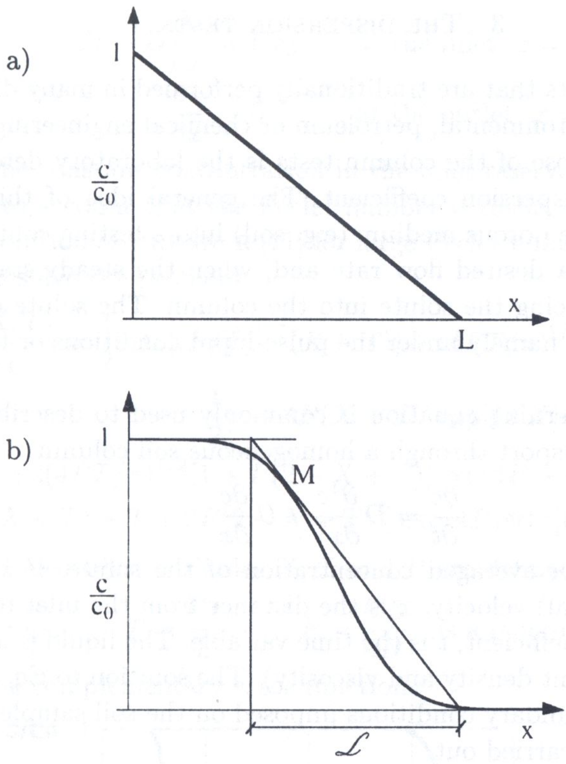

FIG. 2. Macroscopic length. a) static flow, b) transient flow.

from the geometrical scale separation parameter $\varepsilon_g = l/L$. The sample length L, which is naturally associated with the macroscopic characteristic length by many authors, will appear explicitly in the estimation of ε for the particular initial and boundary value problems considered in this paper (see the following chapters). Finally, the homogenization parameter ε is expressed as

$$\varepsilon = l\left|\frac{\partial}{\partial x}\left(\frac{c}{c_0}\right)\right|. \tag{2.5}$$

It should be stressed that the value of ε plays a doubly fundamental role in the modelling. On the one hand, it determines the domain of validity of the modelling, that enables the correct choice between the diffusion/dispersion models offered by the homogenization method using the asymptotic expansion technique [5, 6]. On the other hand, it gives the precision of modelling that has to be kept in mind in the process of test preparations or interpretation of the test results.

The concepts presented above will be discussed in more details by using the experimental data of four investigations of dispersion/diffusion coefficients, reported in the literature.

3. THE DISPERSION TESTS

The dispersion tests that are traditionally performed in many different fields, like for example: environmental, petroleum or chemical engineering, are the column tests. The purpose of the column tests is the laboratory determination of the hydrodynamic dispersion coefficient. The general idea of this experiment consists in placing the porous medium (e.g. soil) into a testing column, pumping the water into it at a desired flow rate and, when the steady-state conditions are achieved, introducing the solute into the column. The solute can be added in two different ways, namely under the pulse-input conditions or the step-input condition.

The following governing equation is commonly used to describe the one-dimensional solute transport through a homogeneous soil column

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} - U\frac{\partial c}{\partial x}, \tag{3.1}$$

where c is the volume-averaged concentration of the solute, U is the average pore-water (interstitial) velocity, x is the distance from the inlet to the column, D is the dispersion coefficient, t is the time variable. The liquid is assumed to be homogeneous (constant density and viscosity). The solution to Eq. (3.1) depends on the initial and boundary conditions imposed on the soil sample, i.e. the type of experiment being carried out.

Let us analyse four dispersion/diffusion experiments reported in the literature. The purpose of the analysis is to investigate the validity of the presented results from the point of view of the continuum approach employed to interpret the measured quantities.

3.1. *The experiments of Bues and Aachib*

The experiment conditions were the following [12]:

• soil	quartz sand,
• mean grain diameter	$\delta = 1.425\,\mathrm{mm}$,
• solution	calcium chloride $CaCl_2$,
• column	Plexiglas column $L = 2\,\mathrm{m}$,
• stepwise input	
• molecular Péclet number	$\mathrm{Pe}_m = 300$ (author's notation),

where $\mathrm{Pe}_m = U\delta/D_m$, U is the mean pore velocity, δ is the mean grain diameter and D_m is the molecular diffusion coefficient.

The concentration $c(x,t)$ satisfies the initial-boundary value problem (3.1) – (3.4)

$$c(x,0) = 0, \tag{3.2}$$

$$Uc - D\frac{\partial c}{\partial x} = Uc_0 \quad \text{at the inlet } x = 0, \tag{3.3}$$

$$\frac{\partial c}{\partial x} = 0 \quad \text{at the outlet } x = L, \tag{3.4}$$

where c_0 is the constant concentration in the inlet reservoir.

Taking into account that the Péclet number is rather elevated, the available asymptotic solution of BRENNER [11] for large Péclet number and/or small t will be applied to solve the problem:

$$\begin{aligned} \frac{c(X,T)}{c_0} = {} & \frac{1}{2}\operatorname{erfc}\left[(P/T)^{1/2}(X-T)\right] + (4PT/\pi)^{1/2}\exp[-P(X-T)^2/T] \\ & - \frac{1}{2}[1+4P(X+T)]\exp[4PX]\operatorname{erfc}\left[(P/T)^{1/2}(X+T)\right] \\ & + 2(4PT/\pi)^{1/2}[1+P(2-X+T)]\exp(4P - \{[P(2-X+T)^2]/T\}) \\ & - 2P[2(2-X+T)+T+2P(2-X+T)^2]\exp(4P)\operatorname{erfc}\left[(P/T)^{1/2}(2-X+T)\right], \end{aligned} \tag{3.5}$$

where the dimensionless variables X, T and P are defined by

$$X = \frac{x}{L}, \quad T = \frac{Ut}{L}, \quad P = \frac{UL}{4D} \quad \text{is a macroscopic Péclet number,} \tag{3.6}$$

and erfc is the complementary error function.

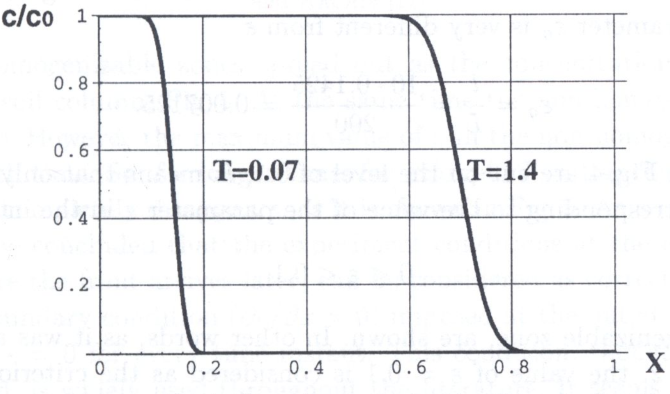

FIG. 3. Concentration profiles in the soil column for the dimensionless times $T = 0.07$ and $T = 1.4$. BUES and AACHIB experiment [12].

The calculations of the concentration profiles and the homogenization parameter ε (scale separation parameter) are performed for the test data given in Fig. 3 in [12]. For example: for $\mathrm{Pe}_m = (U \cdot \delta)/D_m = 300$, the dispersion coefficient was found to be $D = \sim 600 \cdot D_m$, where $D_m = 1.335 \cdot 10^{-5}\ \mathrm{cm}^2/\mathrm{s}$ is the molecular diffusion of CaCl_2 in water (SHACKELFORD and DANIEL [24]). The macroscopic Péclet number P is then evaluated as

$$P = \frac{\mathrm{Pe}_m \cdot D_m \cdot L}{\delta \cdot 4 \cdot D} = \frac{300 \cdot D_m \cdot 200}{0.1425 \cdot 4 \cdot 600 \cdot D_m} = 175.4. \tag{3.7}$$

In Fig. 3 the concentration profiles, Eq. (3.5), in the soil column for $P = 175.4$ and for two dimensionless times $T = 0.07$ and $T = 1.4$ are presented. These times correspond to the real times $t = 498\,\mathrm{s}$ and $t = 996\,\mathrm{s}$, respectively. In Fig. 4

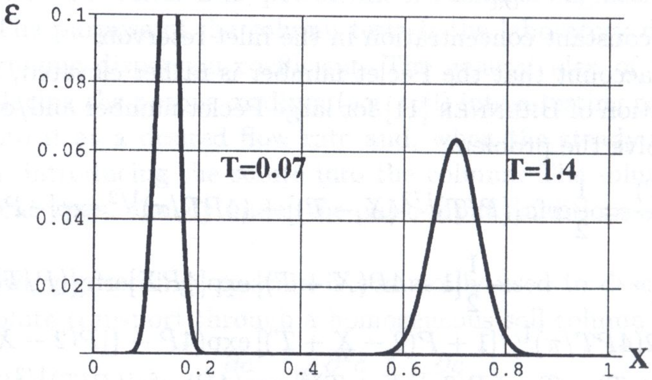

FIG. 4. Variations of ε in the soil column for the dimensionless times $T = 0.07$ and $T = 1.4$. BUES and AACHIB experiment [12].

the corresponding scale separation parameter ε is plotted. The calculations are performed according to the definition of ε provided by the formula (2.5), for the dimensionless times $T = 0.07$ and $T = 1.4$. Remark that the geometrical scale separation parameter ε_g is very different from ε

$$\varepsilon_g = \frac{l}{L} = \frac{10 \cdot 0.1425}{200} = 0.007125. \tag{3.8}$$

The graphs in Fig. 4 are cut on the level of 0.1. It means that only the parts of the graphs corresponding to the value of the parameter ε in the interval

$$0 < \varepsilon < 0.1 \tag{3.9}$$

i.e. the homogenizable zone, are shown. In other words, as it was already mentioned in Sec. 2, the value of $\varepsilon = 0.1$ is considered as the criterion (or as the threshold) of the homogenizability of the problem.

The conclusions may be formulated as follows:

1. From the point of view of the homogenization theory, we are interested in such experiment conditions that would authorise us to apply the continuum approach (the homogenised macroscopic model) to interpret the results. In other words, we are searching zones, in which the concentration gradients are relatively small, thus the measurements could be used to fit the dispersion-advection equation and to determine the dispersion coefficient. We also say that in these zones there exists good separation of the two scales present in the problem (the microscopic and the macroscopic scale), so the homogenizability condition is satisfied.

As it can be seen from Fig. 5, the dispersion zones, where the problem is homogenizable, are very narrow at $T = 0.07$. The presence of non-homogenizable zone renders the front propagation description incompatible with the continuum approach. Therefore, the concentration front at $T = 0.07$ can not be used for the determination of the dispersion coefficient. The situation improves when $T = 1.4$ because the whole concentration front reveals homogenizable features.

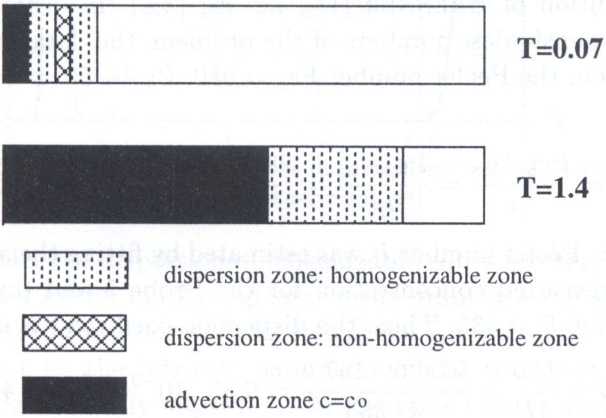

FIG. 5. Evolution of the homogenizable zones in a soil column for the experiment of BUES and AACHIB [12].

2. The homogenizable zones spread out as the concentration front propagates in the soil column, Fig. 5. At the same time the non-homogenizable zone also increases. However the maximum value of ε in the non-homogenizable zone decreases with time. So, for longer time, for example $T = 1.4$, the non-homogenizable zone completely disappears, as it is shown in Fig. 5. As a consequence of this, it can be concluded that the experiment conditions at the end of the soil sample, where the front arrives later, can be considered as correct.

3. The boundary condition ($\partial c/\partial x = 0$) imposed at the outlet of the column implies that $\varepsilon = 0$ for every time instant. This condition, though questionable and criticised, is widely used throughout the literature. It seems to be difficult to comment this assumption on the basis of the homogenization theory.

3.2. *The experiments of Han, Bhakta and Carbonell*

The experiment conditions were the following [17]:

• porous medium	urea formaldehyde spheres;
• mean grain diameter	$\delta = 0.35\,\mathrm{cm}$;
• solution	sodium chloride NaCl;
• column	Plexiglas column $L = 1.50\,\mathrm{m}$;

- stepwise input,
- molecular Péclet number $\mathrm{Pe}_p = 980$ (authors notation),

where $\mathrm{Pe}_p = U\, l_\beta / D_m$, U is the mean pore velocity, l_β is the characteristic length $l_\beta = \delta n/(n-1)$, where n is the porosity ($n = 0.41$) and D_m is the molecular diffusion coefficient, $D_m = 1.545 \cdot 10^{-5}\,\mathrm{cm}^2/\mathrm{s}$.

Since in this experiment again the Péclet number was relatively high, we applied the solution of BRENNER [11], i.e. Eq. (3.5). In order to evaluate the values of the dimensionless numbers of the problem, the data presented in Fig. 5 in [17] were taken: the Péclet number $\mathrm{Pe}_p = 980$, Probe 5 : $x = 147.6$ cm. Then, we get

$$U = \frac{\mathrm{Pe}_m D_m}{l_\beta} = \frac{\mathrm{Pe}_p D_m}{l_\beta} = \frac{980 \cdot 1.545 \cdot 10^{-5}}{0.2432} = 0.0622\,\mathrm{cm/s}\,. \tag{3.10}$$

The macroscopic Péclet number P was estimated by fitting the appropriate value to obtain the measured concentration for the Probe 5 and time $t = 1000$ s. It was obtained that $P = 335$. Thus, the dispersion coefficient is evaluated as

$$D = \frac{UL}{4P} = \frac{0.0622 \cdot 147.6}{4 \cdot 335} = 6.85 \cdot 10^{-3}\mathrm{cm}^2/\mathrm{s} = 443 \cdot \mathrm{D_m}\,. \tag{3.11}$$

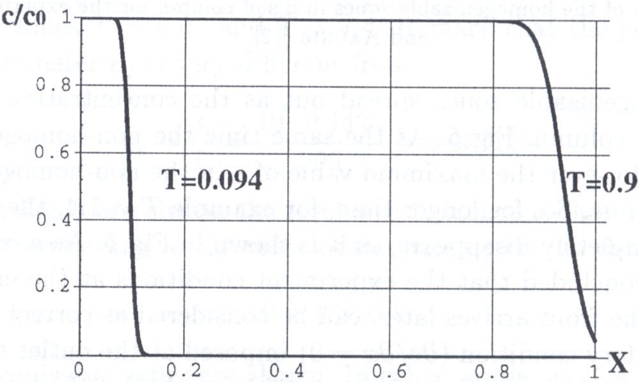

FIG. 6. Concentration profiles in the soil column for the dimensionless times $T = 0.094$ and $T = 0.94$. HAN *et al.* experiment [17].

In Figs. 6 and 7 the concentration profile and the homogenization parameter ε are shown, for the dimensionless times $T = 0.094$ and $T = 0.94$ (the corresponding real times are $t = 226$ s and $t = 2226$ s, respectively). The geometrical scale separation parameter ε_g is estimated as

$$\varepsilon_g = \frac{10 \cdot 0.35}{150} = 0.023\,. \tag{3.12}$$

Figure 8 presents the homogenizable zones at the beginning and at the end of the experiment. In comparison with Fig. 5, the conditions of this experiment are

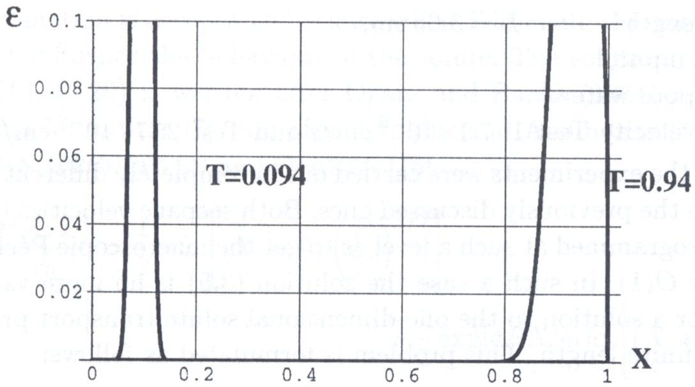

FIG. 7. Variations of ε in the soil column for the dimensionless times $T = 0.094$ and $T = 0.94$. HAN *et al.* experiment [17].

even less favourable: the dispersion homogenizable zones are very narrow, being the results of higher Péclet number, greater mean grain size and shorter column. Such conditions make the laboratory investigations questionable. The dispersion coefficient may not be the intrinsic parameter of the transport problem. The situation would diametrically improve, if for the same porous medium (soil) and for the same Péclet number, the column was longer.

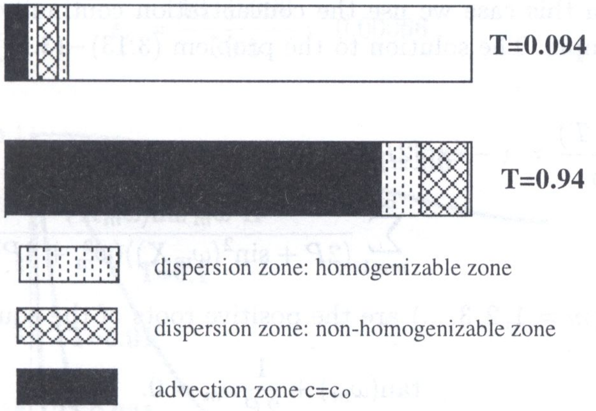

FIG. 8. Evolution of the homogenizable zones in a soil column for the experiment of HAN *et al.* [17].

3.3. *The experiments of Shackelford and Redmond*

The experiment conditions were the following [23]:

• porous medium	processed kaolin,
• mean grain diameter	assumed $\delta = 0.002\,\mathrm{mm}$ (39% silt, 61% clay),
• solution	sodium chloride NaCl,

- sample length $L = 3.05\,\text{cm}$,
- stepwise input,
- average pore water ,

(seepage) velocity Test 1: $7.1 \cdot 10^{-6}$ cm/s and Test 2: $7 \cdot 10^{-7}$ cm/s.

In this case the experiments were carried out in completely different conditions with respect to the previously discussed ones. Both seepage velocities (Test 1 and Test 2) were programmed at such a level as to get the macroscopic Péclet number P of the order $O(1)$. In such a case the solution (3.5) is no more valid and we have to look for a solution to the one-dimensional solute transport problem in a soil sample of finite length. This problem is formulated as follows:

$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} - U\frac{\partial c}{\partial x} \tag{3.13}$$

with the initial and boundary conditions

$$c(x,0) = 0, \tag{3.14}$$

$$c(0,t) = c_0\,, \tag{3.15}$$

$$\frac{\partial c}{\partial x}(L,t) = 0. \tag{3.16}$$

Remark that in this case we use the concentration continuity condition at the inlet to the sample. The solution to the problem (3.13) – (3.16) can be obtained in the form

$$\frac{c(X,T)}{c_0} = 1 - \exp[2PX - PT] \cdot \sum_{m=1}^{\infty} \frac{4P\omega_m \sin(\omega_m X)}{(2P + \sin^2(\omega_m X))(\omega_m^2 + 4P^2)} \exp\left[-\omega_m^2 \frac{T}{4P}\right], \tag{3.17}$$

where the ω_m $(m = 1, 2, 3, \ldots)$ are the positive roots of the equation

$$\tan(\omega_m) + \frac{1}{2P}\omega_m = 0. \tag{3.18}$$

The dimensionless variables appearing in Eq. (3.17) are defined by

$$P = \frac{UL}{4D}\,, \qquad T = \frac{Ut}{L}, \qquad X = \frac{x}{L}\,. \tag{3.19}$$

When T is small, the convergence of the series (3.17) is very slow. To overcome this difficulty, Eq. (3.17) is replaced by the solution for the semi-infinite medium with the boundary condition (3.20) instead of (3.16)

$$c(\infty, t) = 0. \tag{3.20}$$

This approximation is acceptable because when T is small, the outlet of the sample does not influence the behaviour of the solute. The solution to the problem (3.13) - (3.15), (3.20) is written after Ogata and Banks (1961) as in the paper by BEAR and VERRUIJT [7], and after Lapidus and Amundson (1952) as in the paper by VAN GENUCHTEN and PARKER [15]:

$$\frac{c(X,T)}{c_0} = \frac{1}{2}\operatorname{erfc}\left[(X-T)\left(\frac{P}{T}\right)^{1/2}\right] + \frac{1}{2}\exp[4PX]\operatorname{erfc}\left[(X+T)\left(\frac{P}{T}\right)^{1/2}\right]. \tag{3.21}$$

The macroscopic Péclet numbers are calculated using the values of the dispersion coefficients obtained for the ions Cl^- [23]

$$\text{Test 1}: \qquad P = \frac{7.1\cdot 10^{-6}\cdot 3.05}{4\cdot 3.9\cdot 10^{-6}} = 1.38\,; \tag{3.22}$$

$$\text{Test 2}: \qquad P = \frac{7\cdot 10^{-7}\cdot 3.05}{4\cdot 2.8\cdot 10^{-6}} = 0.19\,. \tag{3.23}$$

The geometrical scale separation parameter is estimated

$$\varepsilon_g = \frac{10\cdot 0.002}{30.5} = 0.00066\,. \tag{3.24}$$

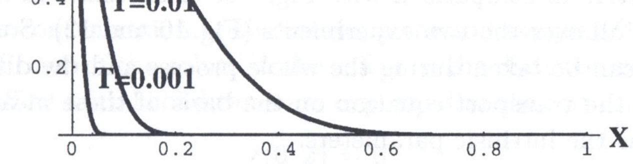

FIG. 9. Concentration profiles in the soil sample for $P = 1.38$, for the dimensionless times $T = 0.001$, 0.01, 0.1 and 1.0. SHACKELFORD and REDMOND experiment [23].

The concentration and the homogenization parameter as a function of X for the dimensionless times $T = 0.001$, 0.01, 0.1 and 1.0 for the Test 1 and the Test 2 are plotted in Figs. 9 - 12. The calculations were performed in accordance with Eq. (3.21) for $T = 0.001$ and 0.01, and with Eq. (3.17) for $T = 0.1$ and 1.0.

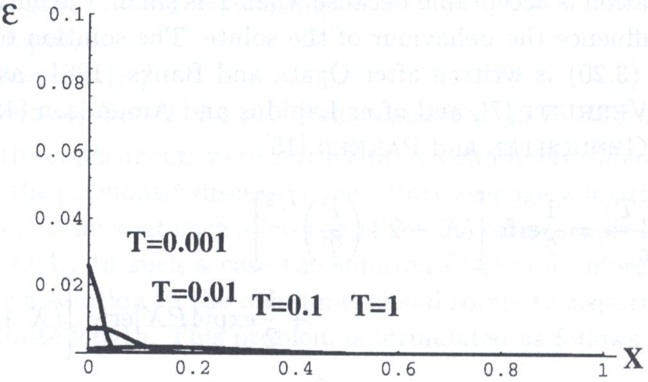

FIG. 10. Variations of ε in the soil sample for $P = 1.38$, for the dimensionless times $T = 0.001$, 0.01, 0.1 and 1.0. SHACKELFORD and REDMOND experiment [23].

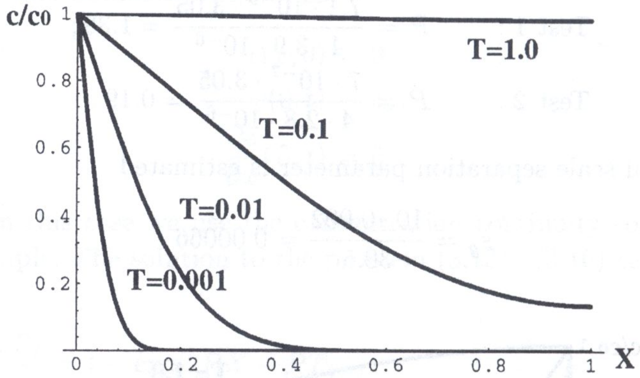

FIG. 11. Concentration profiles in the soil sample for $P = 0.19$, for the dimensionless times $T = 0.001$, 0.01, 0.1 and 1.0. SHACKELFORD and REDMOND experiment [23].

It can be seen that in these tests the concentration curves, Fig. 9 and 11, have different characters in comparison with Fig. 3 or 6. It results in the homogenizable conditions all over the two experiments (Fig. 10 and 12). So, the "correct" measurements can be taken during the whole process and the dispersion coefficients fitted to the transport equation on the basis of these measurements can be considered as the intrinsic parameters.

4. EFFECTIVE DIFFUSION TESTS

There are several methods of the laboratory measurements of effective diffusion coefficient, like the half-cell method, the steady-state method, and both the column and reservoir methods with constant and decreasing source concentra-

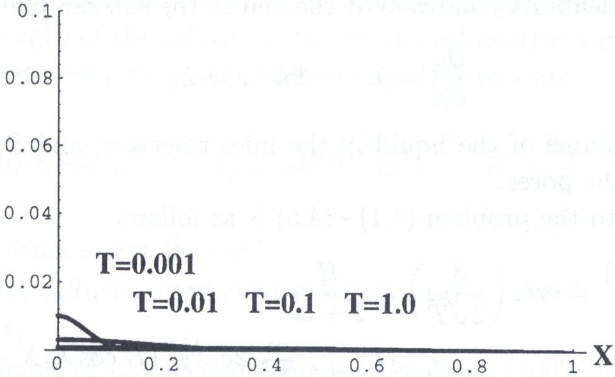

FIG. 12. Variations of ε in the soil sample for $P = 0.19$, for the dimensionless times $T = 0.001$, 0.01, 0.1 and 1.0. SHACKELFORD and REDMOND experiment [23].

tions. Let us consider the test called single reservoir with decreasing source concentration (SRDC) method. Such tests are frequently performed in the geotechnical and environmental laboratories because they are relatively easy to use and the experimental set-up and the boundary conditions are analogous to the environmental applications in which contaminants are contained by soil mineral barriers (SHACKELFORD and DANIEL [24]). In the SRDC method a reservoir containing the solute of the initial concentration c_{00} is placed on top of a cell of soil. Due to the gradient of the solute concentrations between the reservoir and the soil pore water, diffusion of the solute to the soil occurs. The changes in the concentration are monitored and utilised to evaluate the effective diffusion coefficient.

The mathematical formulation of this problem (i.e. the macroscopic diffusion model) is the following

$$D^* \frac{\partial^2 c}{\partial x^2} = \frac{\partial c}{\partial t}, \tag{4.1}$$

where c is the concentration of the solute in the soil sample, D^* is the effective diffusion coefficient of the solute in the soil, x is the distance from the inlet and t is the time. The initial conditions are expressed in the form

$$c(0, x) = 0, \tag{4.2}$$

$$c(0, 0) = c_{00}, \tag{4.3}$$

c_{00} is the initial concentration in the inlet reservoir.

The assumption of a uniform concentration c_R in the inlet reservoir leads to the flux boundary conditions

$$V \frac{\partial c_R}{\partial t} = D^* \frac{\partial c_R}{\partial x} S, \qquad c = c_R \qquad \text{for } x = 0, \tag{4.4}$$

while the impermeability condition at the end of the soil sample yields

$$\frac{\partial c}{\partial x} = 0, \qquad \text{for } x = L; \tag{4.5}$$

V is the total volume of the liquid in the inlet reservoir, and S is the average cross-section of the pores.

The solution to the problem (4.1) – (4.5) is as follows:

$$\frac{c(X,T)}{c_{00}} = \operatorname{erfc}\left(\frac{X}{2\sqrt{T}}\right) - \frac{H}{1+H} + \sum_{n=1}^{\infty} \frac{2e^{-\lambda_n^2 T}(H\cos\lambda_n X - \lambda_n \sin\lambda_n X)}{H + H^2 + \lambda_n^2}, \tag{4.6}$$

where the dimensionless variables T, X and H are defined as

$$X = \frac{x}{L}, \qquad T = \frac{Dt}{L^2}, \qquad H = \frac{V_p}{V}, \tag{4.7}$$

$$V_p = SL, \tag{4.8}$$

and the λ_n are given by

$$H \operatorname{tg} \lambda_n + \lambda_n = 0, \qquad \text{where } n = 1, 2, 3, \ldots . \tag{4.9}$$

Then, the definition (2.5) can be used to calculate the scale separation parameter ε

$$\varepsilon = \varepsilon \left[\frac{e^{-X^2/4T}}{\sqrt{\pi T}} + \sum_{n=1}^{\infty} \frac{2\lambda_n e^{-\lambda_n^2 T}(H\sin\lambda_n X + \lambda_n \cos\lambda_n X)}{H + H^2 + \lambda_n^2}\right], \tag{4.10}$$

where ε_g is the ratio of the geometrical scales l and L,

$$\varepsilon_g = \frac{l}{L}. \tag{4.11}$$

With a view to see the changes of ε as a function of time and position in a soil sample, let us now analyse the experimental results of a diffusion test performed by SHACKELFORD and DANIEL [24]. Since the volume of the liquid in the reservoir V is not given by the authors explicitly, it has to be deduced from the other information provided in the paper. As an example, we take the test results presented in Fig. 5 (a) in [24] (Test Series 1, Test 1) and we note that for $t \to \infty$ (the end of the test) the solution to the problem, Eq. (8) in [24], reduces to

$$\frac{c_\infty}{c_0} = \frac{\alpha}{1+\alpha}, \tag{4.12}$$

where α is a dimensionless coefficient, depending on the "effective length of the reservoir, the length of the diffusion cell and the volumetric water content of the soil". The needed test parameters estimated in this way are

$$V = 1038.86\,\mathrm{cm}^3, \qquad V_p = 540.01\,\mathrm{cm}^3, \qquad H = \frac{V_p}{V} = 0.5198.$$

Further information about the test:

- the effective diffusion coefficient determined from the Cl^- analysis is $D^* = 4.5 \cdot 10^{-6}\,\mathrm{cm}^2/\mathrm{s}$,
- the soil sample of $L = 58$ mm is made of kaolinite (100% silt and clay). The geometrical scale separation parameter can be estimated as

$$\varepsilon_g = \frac{10 \cdot 0.075}{58} = 0.0129.$$

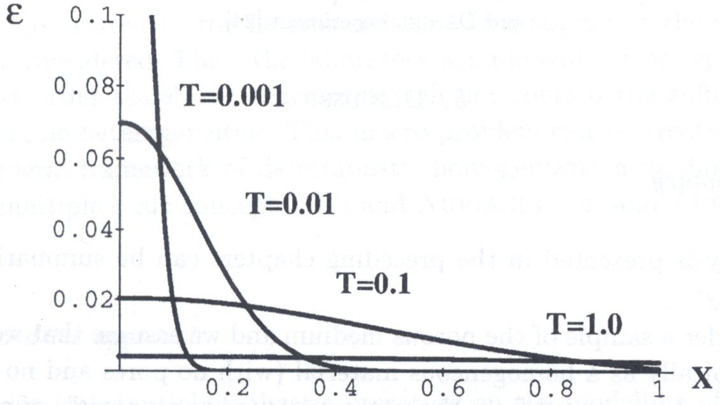

FIG. 13. Scale separation parameter ε versus space variable for different dimensionless times. $L = 116$ cm. SHACKELFORD and DANIEL experiment [24].

In Fig. 13 the scale separation (the homogenization) parameter ε in the soil sample is shown, for different dimensionless times $T = 0.001$, 0.01, 0.1 and 1.0. It corresponds to the real times $t = 8.3$ h, 83 h, 830 h and 8300 h, respectively. It can be deduced that at $T = 0.001$, the part of the curve where $\varepsilon \leq 0.1$ is a narrow segment in the sample, while for $T \geq 0.01$ the situation is homogenizable all over the sample. In Fig. 14 the isolines of ε as functions of X and T are presented, so the homogenizable zones as functions of position and time can be easily distinguished.

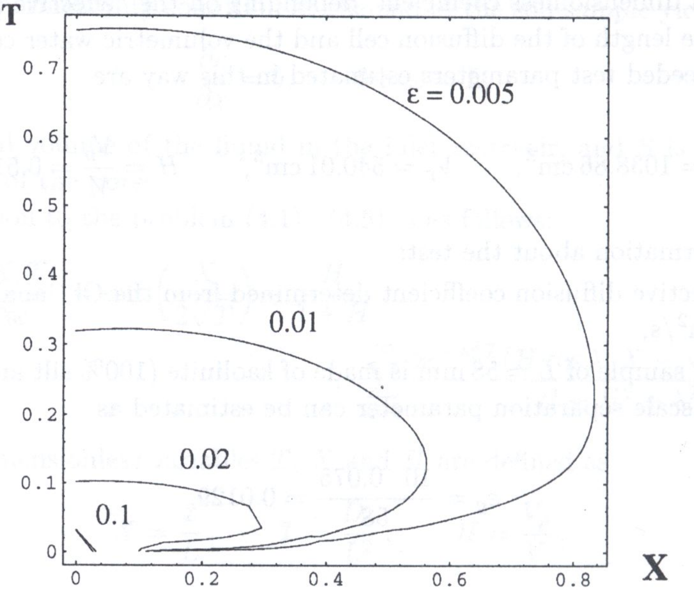

FIG. 14. Contour plots of the scale separation parameter ε. $L = 116$ cm. SHACKELFORD and DANIEL experiment [24].

5. DISCUSSION

5.1. *Summary*

The analysis presented in the preceding chapters can be summarised in the following way:

We consider a sample of the porous medium and we assume that we can treat it macroscopically as a homogeneous material (with no pores and no grains). It means that we have passed from the microscopic (or "pore scale") of observation to the laboratory (or "Darcy scale") of observation. Therefore, according to [5, 6] we can apply the "homogenised" advection-dispersion equation (3.1) (or the "homogenised" diffusion equation (4.1)) to describe the chemical transport occurring in this medium. This equation can also be used in the inverse problem of fitting the experimental data (the concentration measurements) to determine the transport parameters. The only constraint is the condition of scale separation. Using the homogenization approach we say that Eq. (3.1) is valid, and thus the experimentally determined transport coefficients are the intrinsic parameters, if the scale separation condition is satisfied in each point of the wave front. However, this condition can only be verified *a posteriori*, based on the laboratory test results and the calculations of $\varepsilon(x, t)$ presented in this paper.

5.2. *Field applications*

The important question that has not been directly posed in this paper is whether the classical advection-dispersion equation (3.1) with the laboratory-fitted dispersion coefficients provides an intrinsic description of the solute transport through natural aquifer materials which exhibit broad range of heterogeneities. The attempts to apply this model to field conditions have led to several difficulties. The examination of over 130 longitudinal dispersivities D/U (reported in NEUMAN [19]), deduced by means of the continuum Fickian theory expressed by Eq. (3.1), from laboratory and field tracer studies in a variety of porous media proved that there is a systematic increase in the longitudinal dispersivity with the scale of the study, known in the hydrological literature as the "scale effect". The experimental data recalled in NEUMAN *et al.* [21] show that in the laboratory the longitudinal dispersivity ranges from 10^{-4} to 10^{-1}m for relatively uniform fine-to-coarse gravel. In the field tracer experiments it varies from 10^{-2} to 10 m and may exceed 10^2 m. When the longitudinal dispersivity is obtained based on the documented cases of aquifer pollution or chemistry on a regional scale, its value ranges from less than 10 m to more than 100 m. It is clear that the "scale effect" is related to the heterogeneities which appear at the scale larger than the one considered. Thus, the laboratory sample will not be representative for the "next order scale" i.e. the sample will not contain the sufficient number of large scale heterogeneities. This macro-problem can be treated following the same general framework of deterministic homogenization method by introducing the multiple scale concept, MEI and AURIAULT [18] and AURIAULT and BOUTIN [4].

5.3. *Stochastic approach*

There exists substantial stochastic literature on the modelling of the chemical transport within a heterogeneous porous medium, regarding the Fickian dispersion, micro-macroscale processes and the REV. Efforts to understand the physical mechanism and to describe the scale dependence of the dispersion process observed in field situations have led to a conclusion that chemical transport in geological media is strongly influenced by spatial variations in permeability. Such variations produce fluctuations in the subsurface fluid velocity which in turn cause the solute to disperse over distances considerably greater than those normally observed in laboratory column experiments (NEUMAN [20]). The stochastic approaches focus on the transport theories that could be applied to highly nonuniform geological media as encountered in the hydrological practice. Several theoretical studies have analysed the dispersion mechanism as dependent on three-dimensional correlation structure of the medium. Among many contributions, the stochastic methods proposed by GELHAR and AXNESS [16], DAGAN [14] and NEUMAN [19, 20] should be mentioned. GELHAR and AXNESS [16] developed a stochastic predictive model of macrodispersion in aquifers. This model is applicable to transport within identifiable geologic unit and suitable to describe long-term, large displacement behaviour. This theory indicates that a "classical gradient (Fickian) relationship" is valid for asymptotic condition of large displacement. NEUMAN [19] considered a fractal analysis of the scale effect and proposed a universal scaling rule that accounts for self-similarity of log hydraulic conductivities in the mean sense over a large length scales in a broad variety of geological media under diverse conditions of flow and transport. In 1993 NEUMAN [20] analysed the transport of solute in a random velocity field and developed a non-local theory using the ensemble moments conditions on local hydraulic measurements. His results apply to "multiscale velocity fields of the kind expected to arise in geologic media with evolving heterogeneities, including fractal media above some cut-off". In 1994 DAGAN [14] derived an exact nonlinear correction to the transverse macrodispersion coefficient for transport in heterogeneous formations. All these intensive research studies are oriented toward large-scale field applications.

In the stochastic framework it was found that the Fickian mode of dispersion (the classical advection-dispersion equation i.e. Eq. (3.1)) is attained asymptotically in a homogeneous medium of infinite extent i.e. the longitudinal dispersivity tends asymptotically to a constant, as the distance increases. Consequently, the Fickian behaviour is not expected to be valid near the solute sources [16]. As stated by Neuman (1993): "Stochastic analysis of transport often admits the applicability of Fick's law on some "local scale" [Gelhar and Axness (1983), Dagan (1987), Neuman *et al.* (1987), Sposito and Barry (1987), Neuman and Zhang (1990) see [20]]. However, this scale is seldom specified in an unambiguous manner." It should be pointed out that the question of applicability of the continuous Fick's model can be answered both qualitatively and quantitatively in the framework of the homogenization upscaling technique (see for example: [6, 3]).

5.4. Deterministic versus stochastic approach

Although generally it is difficult to compare the deterministic and stochastic approaches, one can find some common points when interpreting the column test results. The results published in the literature show that in a given tracer experiment, the dispersivity coefficient grows with the distance from the sampling point towards an asymptotic value [see for example: Martin (1971), Peaudecerf and Sauty (1978), Sudicky and Cherry (1979), Sudicky *et al.* (1983) cited in NEUMAN *et al.* [21]]. This behaviour is consistent with the results of the stochastic analysis (DAGAN [14]). In the experiment of Bues and Aachib we can observe the asymptotic value of the dispersion coefficient for the regime $\mathrm{Pe}_m = 300$ at the end of the sample (Fig. 3 in BUES and AACHIB [12]). In all the other cases i.e. for other Péclet numbers, the asymptotic behaviour is not well pronounced. On the other hand, according to the deterministic homogenization analysis presented in Sec. 3.1, the asymptotic behaviour corresponds to a "homogenizable condition", Fig. 5, and therefore it provides the dispersion coefficient that can be considered as the intrinsic parameter. The two methods converge to the same conclusion. Similarly, the experiment conditions of HAN *et al.* [17] for $\mathrm{Pe}_m = 980$, can not be called "homogenizable conditions", see Sec. 3.2 and Fig. 8, and indeed Fig. 9 in HAN *et al.* [17] shows that the asymptotic value has not yet been achieved.

6. CONCLUSIONS

The existing experimental data on dispersion/diffusion in soil columns were exploited to analyse the validity of the continuum approach employed to interpret these results. The obtained values of the homogenization (or scale separation) parameter proved that some experimental situations do not ensure the conditions of good scale separation, that is the necessary condition of the existence of the macroscopic transport model (the Fick's transport equation). In such situations the parameter identification test is not correct and will lead to the estimation of the parameters that are not the intrinsic parameters. The results will be experiment-dependent and thus, it will not be applicable to practical situations. This fact implies that in order to work out the concept of an adequate parameter identification test, it would be helpful to repeat the analysis proposed in this paper in a similar manner but for the particular test conditions.

It should be recognised that at present it is very difficult to answer many questions related to the problem of transport in porous media. One of the fundamental questions is the problem concerning the REV. Although in the recent stochastic framework "one must question the utility of associating medium properties with the REVs as has been the custom in subsurface hydrology for several decades" (NEUMAN [19]), the deterministic homogenization approach is inherently related to the REV through the scale separation condition. Therefore, it is hoped that substantial efforts will be devoted to the development of techniques of determination of the REV for the transport problems. The other important issue that has not been addressed in this paper concerns the measuring technique which is connected to the question of scale. CUSHMAN [13] posed the question:

"what is actually being measured by an instrument, i.e. are we measuring a "point" property, a weighted spatial average, a temporal point average, a spatial and temporal average etc.?" Throughout this paper it was silently assumed that the measurements of concentration (the chemical analyses) were done in the conditions enabling us to obtain the proper input values for the model equation (3.1) or (4.1), i.e. the proper average concentrations c.

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