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An asymptotic analysis for determining concentration uncertainty in aquifer transport

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Abstract

Transport of a conservative solute takes place by advection and by pore-scale dispersion in a formation of spatially variable logconductivity $Y(\mathbf{x}) = \ln K(\mathbf{x})$. The latter is modeled as a normal stationary random space function, characterized by a few statistical parameters, like the mean $\langle Y \rangle$, the variance σ_Y^2 , the horizontal and vertical integral scales I_h and I_v . The local solute concentration $C(\mathbf{x}, t)$, a random function of space and time, is characterized by its statistical moments, like, e.g. the mean $\langle C \rangle$ and the standard deviation σ_C . A simplified analysis for determining the concentration uncertainty is proposed. The proposed methodology, valid for nonreactive solutes, is based on a few simplifications, the most important being: (i) large transverse dimensions of the injected plume compared to the logconductivity correlation lengths, (ii) mild heterogeneity of the hydraulic properties, which allows for the use of the first-order analysis, (iii) highly anisotropic formations, and (iv) mean uniform flow. The concentration uncertainty is represented through the coefficient of variation $CV_C = \sigma_C/\langle C \rangle$ at the plume center, where the expected concentration is maximum. Results for CV_C are illustrated as function of time and on two dimensionless parameters: $\Omega = I_v^2/(I_h \alpha_{dT})$ and $\Lambda = L_1/\sqrt{A_{11} I_h}$, where L_1 is the longitudinal dimension of the initial plume, A_{11} is the longitudinal macro dispersivity, and α_{dT} is the local transverse dispersivity. Summary graphs lead to a quick and simple estimate of the time-dependent concentration uncertainty, as well as its peak and its setting time (i.e. the time needed to reach the peak coefficient of variation). The methodology and its results can be used to assess the concentration uncertainty at the plume center. The problem is quite important when dealing with contaminant prediction and risk analysis.

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1. Introduction

Spatial variability of hydraulic conductivity affects the fate of contaminants in groundwater by a few mechanisms, the main ones being spreading and dilution. Spreading is caused by the variation in groundwater velocity due to permeability changes, leading to shearing of the plume. In contrast, dilution is controlled by pore-scale dispersion (hereinafter denoted as PSD) that determine a solute transfer from

fast moving fingers to neighboring stream tubes and conversely. Because of the anisotropy of the heterogeneous structure of most sedimentary aquifers, manifesting in vertical scales much smaller than the horizontal ones, the major mixing effect is in the vertical direction and is associated with transverse PSD. The interplay between large-scale advection and PSD depends on the Peclet numbers, the most significant being the transverse one $Pe_T = UI_v/D_{dT}$, where U is the mean velocity, I_v is the vertical

correlation scale of the logpermeability field, $D_{dT} = \alpha_{dT}U$ is the coefficient of transverse PSD and α_{dT} is the transverse dispersivity. In the typical field applications, $Pe_T = I_v/\alpha_{dT} = O(10^2)$ (e.g. Dagan, 1989).

It is common to account for the seemingly erratic variations of permeability and for uncertainty, by modeling the permeability and the dependent fields (piezometric head H , velocity \mathbf{V} and concentration C) as random space functions of the coordinate \mathbf{x} . These variables are characterized statistically by their moments, such as: the mean, the variance, the two-point covariance. Thus, at lowest order the local concentration is characterized by $\langle C \rangle$, the ensemble mean, and σ_C^2 , the variance. While $\langle C \rangle$ is slightly affected by PSD, the latter has a major effect upon σ_C , which is a measure of the concentration uncertainty (see, e.g. Fiori and Dagan, 2000).

From the applications perspective, the evaluation of σ_C , supplementing that of $\langle C \rangle$, is quite important. First, the magnitude of the coefficient of variation $\sigma_C/\langle C \rangle$ is indicative of the uncertainty of prediction of the concentration on one hand, and of the irregular distribution of the measured local concentration distribution in a field test (Fiori and Dagan, 1999). Furthermore, if models are to be used for prediction and for risk analysis, when regulations are formulated in terms of maximal admissible local concentrations rather than space or time averages over wells, the statistical moments of concentration are a prerequisite step toward the determination of the probability density function (hereinafter denoted as PDF) of C ; through the latter, an assessment of the probability of exceedance of a given concentration can be made at any point in space and time (see Fiori and Dagan, 1999).

Two types of approaches to determine σ_C^2 were followed recently: the Eulerian one (e.g. Kapoor and Gelhar, 1994; Andricevic, 1998; Kapoor and Kitanidis, 1998) and the Lagrangean one (Fiori and Dagan, 1999, 2000; Pannone and Kitanidis, 1999). In particular, Fiori and Dagan (1999) found a good agreement between their approximate Lagrangean model and Fitts' (1996) analysis of field data, provided that the calibrated value of the pore-scale transverse dispersivity was $\alpha_{dT} \approx 0.5$ mm for the Cape Cod and Borden aquifers. However, the calculations needed to obtain σ_C^2 require

the evaluation of several quadratures, making the procedure quite complicated for field applications.

The main aim of this study is to develop a simplified analysis for σ_C valid for anisotropic formations and large initial plumes. The plan of the paper is as follows: the Lagrangean model of Fiori and Dagan (2000), which is the basis of the calculation for the concentration moments, is briefly recapitulated in Section 2. In Section 3 the approximate formulation is introduced and discussed, and the results are illustrated in Section 4. For the scope of illustration, the methodology is applied in Section 5 to the analysis of the concentration statistics for the Cape Cod field experiment.

2. Mathematical framework

We consider an aquifer of stationary lognormal distribution of the hydraulic conductivity K , i.e. with $Y = \ln K$ characterized by the constant mean $\langle Y \rangle$ and by the two-point covariance $C_Y(\mathbf{x}, \mathbf{y}) = \sigma_Y^2 \rho_Y(r_1/I_h, r_2/I_h, r_3/I_v)$. Here \mathbf{x} and \mathbf{y} are coordinate vectors of two points, the vector $\mathbf{r} = \mathbf{y} - \mathbf{x}$ is the lag, x_1 and x_2 are Cartesian horizontal coordinates, with x_1 parallel to the mean flow direction, and x_3 is vertical. We assume the heterogeneous structure to be of axisymmetric anisotropy (e.g. Gelhar and Axness, 1983), being completely characterized by $\langle Y \rangle$, the variance σ_Y^2 , the horizontal (I_h) and vertical (I_v) integral scales; the anisotropy ratio is defined as $e = I_v/I_h$. Flow is caused by a mean uniform head gradient $\nabla \langle H \rangle = -\mathbf{J}$, resulting in a random velocity field $\mathbf{V}(\mathbf{x}) = \mathbf{U} + \mathbf{u}(\mathbf{x})$, where $\mathbf{U} = \langle \mathbf{V} \rangle$ is constant and the stationary fluctuation \mathbf{u} has a two-point covariance $u_{ij}(\mathbf{r})$ ($i, j = 1, 2, 3$).

A conservative solute of constant concentration C_0 is injected in the formation within a volume v_0 at $t = 0$. Transport takes place due to advection by \mathbf{V} and PSD of constant coefficients $D_{d11} = \alpha_{dL}U$, $D_{d22} = \alpha_{dT}U$ and $D_{d33} = \alpha_{dT}U$, where α_{dL} , α_{dT} are the longitudinal and transverse pore-scale dispersivities, respectively.

The aim here is to compute the concentration variance σ_C^2 , as function of \mathbf{x} and t . This objective is attained in the Lagrangean framework by regarding the plume as a collection of elementary particles defined at the pore-scale, of mass $nC_0 \mathbf{d}\mathbf{a}$, where n is

the constant porosity and \mathbf{a} is the initial coordinate within v_0 , that move along trajectories $\mathbf{X}_t(t; \mathbf{a}) = \mathbf{X}(t; \mathbf{a}) + \mathbf{X}_d(t)$. The random advective component of the trajectory \mathbf{X} satisfies the differential equation $d\mathbf{X}/dt = \mathbf{V}(\mathbf{X}_t)$, whereas the displacement \mathbf{X}_d associated with PSD is of normal distribution, of variance $X_{dij} = 2D_{dij}t$ and independent of \mathbf{V} . The concentration field associated to this representation can be written in mathematical form as

$$C(\mathbf{x}) = C_0 \int_{v_0} \delta(\mathbf{x} - \mathbf{X}_t) d\mathbf{a} \quad (1)$$

Stated in words, the concentration is equal to the initial one C_0 along the particles trajectories $\mathbf{X}_t = \mathbf{X} + \mathbf{X}_d$.

To obtain the statistical moments of C we observe that its randomness stems from two velocity fields: the Darcian \mathbf{V} , that is the independent one, and the diffusive one \mathbf{w}_d , that is modeled as a Wiener process. Following Fiori and Dagan (2000), the moments of C are computed by averaging Eq. (1) over the variable \mathbf{X}_t . Thus, the following expression for the mean concentration is obtained

$$\langle C(\mathbf{x}, t) \rangle = C_0 \int_{v_0} f_1(\mathbf{x}; t, \mathbf{a}) d\mathbf{a} \quad (2)$$

where $f_1(\mathbf{X}_t; t, \mathbf{a})$ is the PDF of $\mathbf{X}_t(t; \mathbf{a})$. In a similar manner, the concentration variance is obtained as follows

$$\begin{aligned} \sigma_C^2(\mathbf{x}, t) &= \langle C^2 \rangle - \langle C \rangle^2 \\ &= C_0^2 \int_{v_0} \int_{v_0} f_2(\mathbf{x}, \mathbf{x}; t, \mathbf{a}, \mathbf{b}) d\mathbf{a} d\mathbf{b} - \langle C \rangle^2 \end{aligned} \quad (3)$$

where $f_2(\mathbf{X}_t, \mathbf{Y}_t; t, \mathbf{a}, \mathbf{b})$ is the joint PDF of $\mathbf{X}_t = \mathbf{X}_t(t; \mathbf{a})$, $\mathbf{Y}_t = \mathbf{X}_t(t; \mathbf{b})$ (we use the notation $\mathbf{Y}_t = \mathbf{Y} + \mathbf{Y}_d$ for the trajectory of a particle originating at \mathbf{b}).

In the following the assumptions of Fiori and Dagan (2000) are adopted in order to evaluate σ_C^2 : (i) the formation is mildly heterogeneous, and a first-order approximation in σ_Y^2 is adopted; as a consequence, f_1 and f_2 are approximately Gaussian and completely defined by the first two moments of the trajectories, and the components \mathbf{X} and \mathbf{X}_d are uncorrelated; numerical studies have shown that the first-order approximation gives reasonable estimates of the spatial moments up to $\sigma_Y^2 \approx 1$ (e.g. Bellin et al., 1992); although it derives rigorously

from the first-order approximation, it should be noted that a recent study demonstrates that the Gaussian assumption is not uniformly valid (Tartakovsky and Guadagnini, 2002); (ii) the initial solute body is a box (parallelepiped) of dimensions L_1, L_2, L_3 . Despite these simplifications, which mostly derive from the application of the first-order theory, the calculation of σ_C^2 is still a formidable task for real applications, requiring a large number of quadratures for both the trajectory moments evaluation and the integrals in Eq. (3). In Section 3, a simplifying procedure for large plumes in highly anisotropic formations is developed.

3. An asymptotic analysis

The following additional conditions are assumed in the sequel:

- (i) the transverse size of the initial plume is large compared to the hydraulic conductivity correlation lengths, i.e. $L_2/I_h \gg 1$ and $L_3/I_v \gg 1$. The latter condition is usually met in practice, because in natural formations the logconductivity vertical correlation length is usually small, being $I_v = O(10^{-1} \text{ m})$ (Dagan, 1989). In contrast, in natural formations the horizontal correlation length is usually larger than the vertical ($I_h = O(10^0 \text{ m})$), and the application of the above condition for the transverse dimension may require relatively large values of L_2 ; the assumption concerning L_2 will be further discussed at the end of Section 4;
- (ii) the porous formation is highly anisotropic, i.e. $e = I_v/I_h \ll 1$ (say, roughly $e \lesssim 0.2$). This is a typical situation for sedimentary aquifers, which are often characterized by thin lenses with horizontal bedding (Dagan, 1989);
- (iii) the advective displacements X_2, X_3 are neglected because they are much smaller than the longitudinal advective displacement X_1 , that is one of the principal quantities that determine the concentration variance (Fiori and Dagan, 2000);
- (iv) the pore-scale displacements X_{d1}, X_{d2} are neglected, the most important component being the vertical one X_{d3} (see Section 1, and Fiori and Dagan, 2000);

(v) the variance of the longitudinal trajectory $X_{11}(t) = \langle [X_1(t; \mathbf{a}) - \langle X_1(t; \mathbf{a}) \rangle]^2 \rangle$ is evaluated through the ‘Fickian’ approximation, i.e. $X_{11} = 2A_{11}Ut$, with A_{11} the longitudinal macrodispersion coefficient (see, e.g. Gelhar and Axness, 1983). The trajectories autocorrelation function $\rho_{11} = Z_{11}/X_{11}$, with $Z_{11}(t; \mathbf{a} - \mathbf{b}) = \langle (X_1 - \langle X_1 \rangle)(Y_1 - \langle Y_1 \rangle) \rangle$ the longitudinal trajectory two-particles covariance, is calculated through the first-order approximation in the logconductivity variance σ_Y^2 . Furthermore, the dependence of the two-particles covariance Z_{11} on the horizontal lag $a_1 - b_1$ is weak and is therefore neglected. The latter assumptions are also employed and discussed in the comparison with field experiments of Fiori and Dagan (1999).

Thus, the components that determine the concentration statistics are essentially: (i) longitudinal, large-scale advection, and (ii) vertical PSD. These two mechanisms are indeed those controlling the concentration variance (Fiori and Dagan, 2000), as shown by the large-time, asymptotic value for the concentration coefficient of variation $CV_C = \sigma_C/\langle C \rangle$ at the plume center of mass, valid for any configuration of the initial plume, obtained by Fiori and Dagan (2000, Eq. 25). In fact, the asymptotic CV_C depends on the longitudinal large-scale advection and on the vertical PSD solely, all the other components (e.g. transverse spreading) being absent. Hence, the large-time limit for CV_C of the present simplified model is the same as the one obtained by the complete formulation of Fiori and Dagan (2000). Since the same does not happen with the variance σ_C^2 , the coefficient of variation appears to be a more robust quantity for assessing concentration statistics. Hence, CV_C is chosen as the representative quantity for assessing concentration uncertainty.

With the above simplifications, the concentration mean and variance can be obtained through Eqs. (2) and (3). Details of the procedure to obtain the concentration moments are reproduced in Appendix A. In particular, expressions (A3) and (A7)–(A9) of in Appendix A give, respectively, the mean and the variance of concentration as function of dimensionless time $t' = tU/I_h$, dimensionless location $\xi_1 = (x_1 - Ut)/\sqrt{A_{11}I_h}$, and two dimensionless parameters:

$\Omega = I_v^2/(I_h\alpha_{dT}) = ePeT$ and $\Lambda = L_1/\sqrt{A_{11}I_h}$. The solution for $\langle C \rangle$ is fully analytical, while σ_C require two quadratures.

4. Discussion of results

Here we wish to illustrate results for $CV_C = \sigma_C/\langle C \rangle$ at the plume center $x_1 = Ut$ (or $\xi_1 = 0$), where the expected concentration is maximum. We chose to represent uncertainty through CV_C for the reasons illustrated in Section 3. In Fig. 1 the concentration coefficient of variation CV_C at the plume center of mass ($\xi_1 = 0$) is represented as function of the dimensionless time $t' = tU/I_h$ for a few values of the parameter $\Lambda = L_1/\sqrt{I_hA_{11}}$ and four values of the parameter $\Omega = I_v^2/(I_h\alpha_{dT})$. Since the permeability anisotropy ratio in natural aquifers is generally very small, being $e < 0.1$, with the conductivity vertical integral scale $I_v = O(10^{-1} \text{ m})$, and the transverse dispersivity is $\alpha_{dT} = O(10^{-4} \text{ m})$ (see, e.g. Dagan, 1989), the parameter Ω may have a large range of variability, say $\Omega = 10^0 - 10^3$. Hence, the following values for Ω have been selected for representing results : 1, 10, 100, 1000. The variability of Λ can also be large since it depends on the particular value of the longitudinal size of the initial plume L_1 .

In accordance with the results for the concentration coefficient of variation obtained in the past (e.g. Kapoor and Gelhar, 1994; Andricevic, 1998; Pannone and Kitanidis, 1999; Fiori and Dagan, 2000), the figures show that CV_C grows initially with time to reach a maximum value, after which the coefficient of variation decays with time and tends to zero for $tU/I_h \rightarrow \infty$, proportional to $\ln t/t$. The behavior reflects the competition of two mechanisms in determining CV_C : large-scale advection generates uncertainty, which is however ‘destroyed’ by dilution, that is in turn controlled by PSD. The two mechanisms are controlled by the parameters Ω and Λ , which therefore determine the value of the peak of CV_C ; the same parameters control the ‘setting time’, i.e. the time needed to reach the peak coefficient of variation. Thus, for a fixed Ω , the peak CV_C is inverse proportional to $\Lambda = L_1/\sqrt{A_{11}I_h}$. In fact, an increase of the macrodispersivity A_{11} leads to an increase of

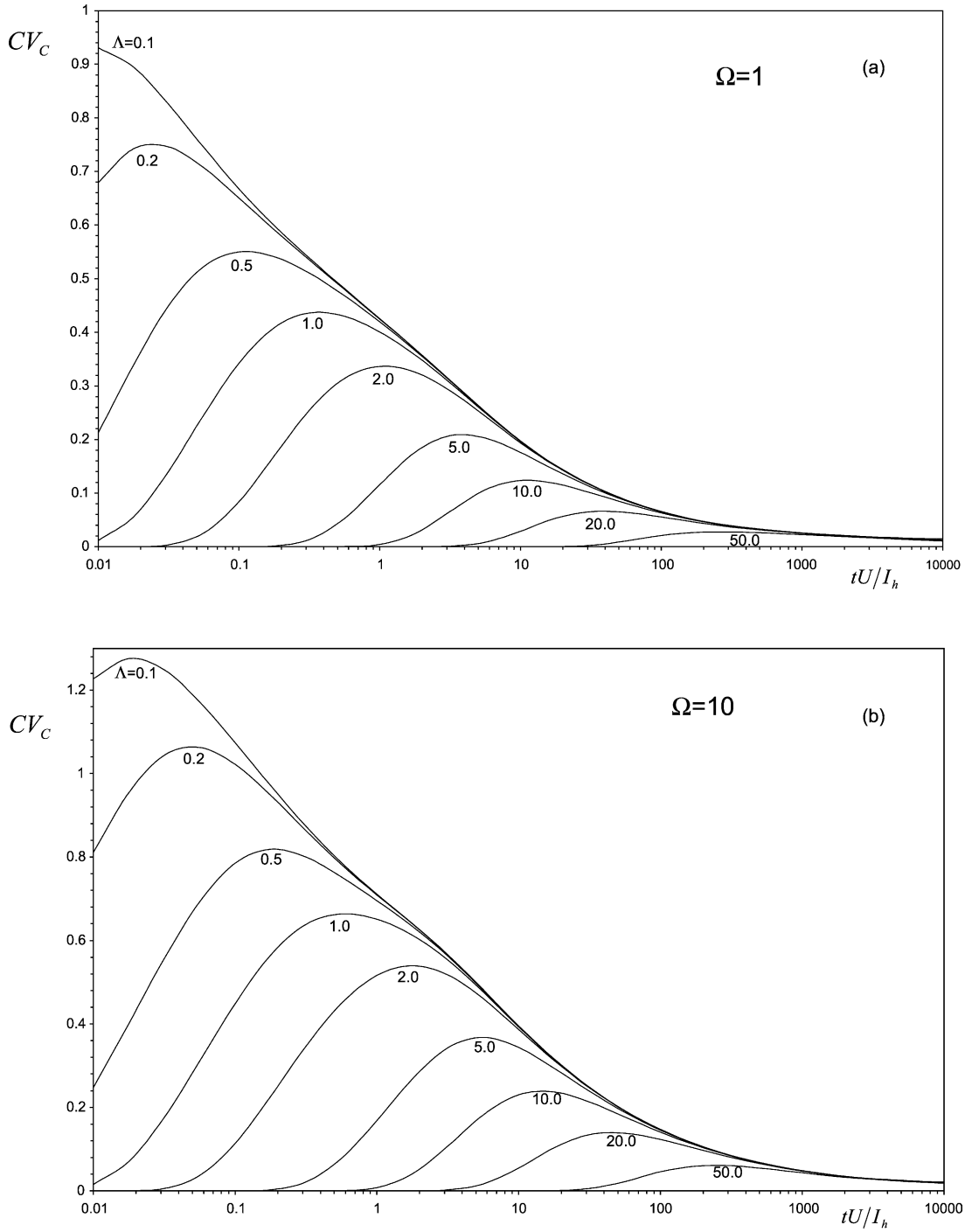


Fig. 1. The concentration coefficient of variation at the plume center of mass $CV_C = \sigma_C/\langle C \rangle$ as a function of dimensionless time tU/I_h for a few values of the parameter $\Lambda = L_1/\sqrt{A_{11}I_h}$, and for $\Omega = I_v^2/(I_h\alpha_{DT}) = 1$ (a), 10 (b), 100 (c), 1000 (d).

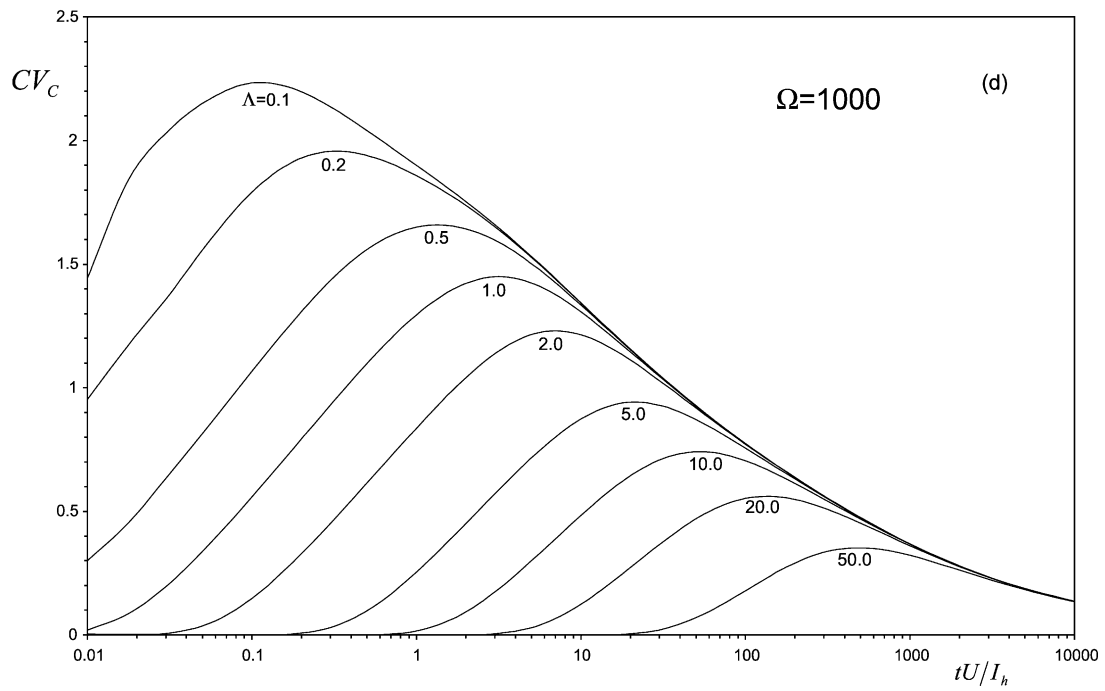
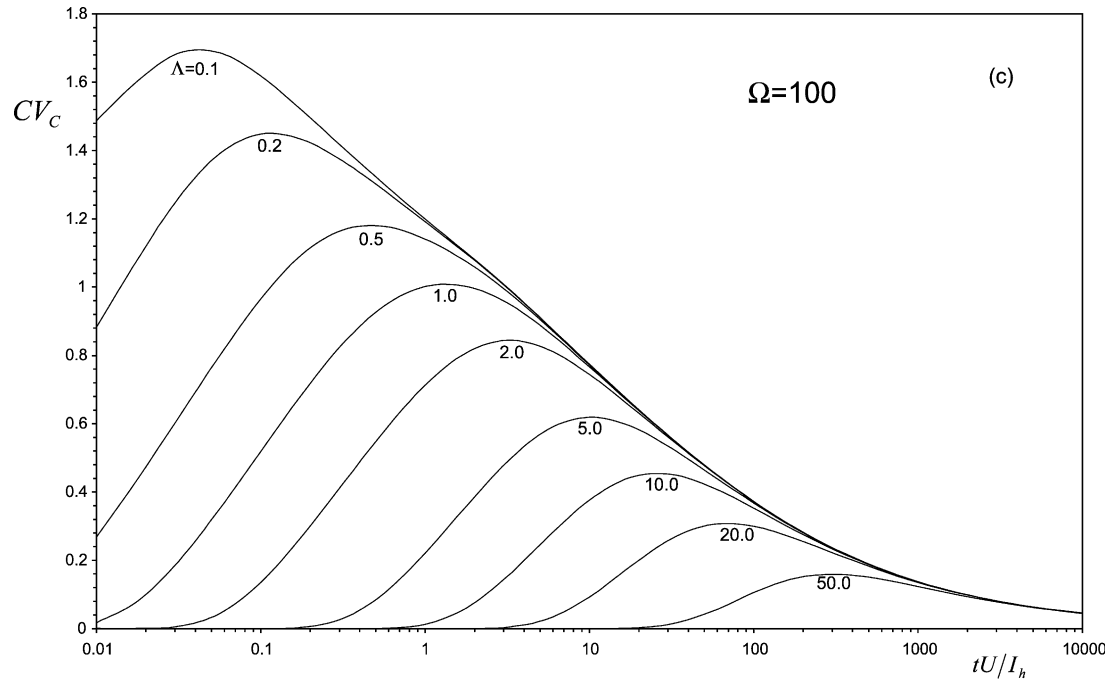


Fig. 1 (continued)

the concentration uncertainty because of the effects of macro-scale advection. For the same reason, the setting time is small because of the fast advection. The plume size L_1 has an opposite effect: if the plume is large, the maximum concentration at the plume center stays high, close to the initial value, and the concentration uncertainty in the surrounding of the plume center is low. Conversely, the setting time will be large, because it takes more time for PSD to influence the center of the plume and play an active role in reducing uncertainty.

The parameter $\Omega = I_v^2 / (I_h \alpha_{dT}) = ePe_T$ controls the effect of PSD, by the means of the transverse dispersivity α_{dT} . The concentration coefficient of variation at the plume center grows with Ω , as the latter is proportional to the transverse Peclet number. This result is in accordance with the behavior illustrated in Fiori and Dagan (2000). The anisotropy ratio $e = I_v / I_h$ is also important, because

it is proportional to the characteristic vertical size of heterogeneity. A small value of I_v makes PSD more effective in diluting the solute and reducing the uncertainty. A similar effect is observed in Fiori (1996) for the concentration spatial moments. A factor similar to Ω was also employed by Naff (1990) for the calculation of the concentration spatial moments. The dependence of the peak concentration coefficient of variation setting time t^* on Ω is weaker, and a decrease of t^* with Ω is generally observed.

To further emphasize results, we represent in Fig. 2 a contour plot for the maximum CV_C at the plume center of mass, as a function of both parameters Ω and Λ . The behavior of the coefficient of variation reflects the one previously explained. An approximate analytical expression for the concentration coefficient of variation at the plume center as function of the dimensionless

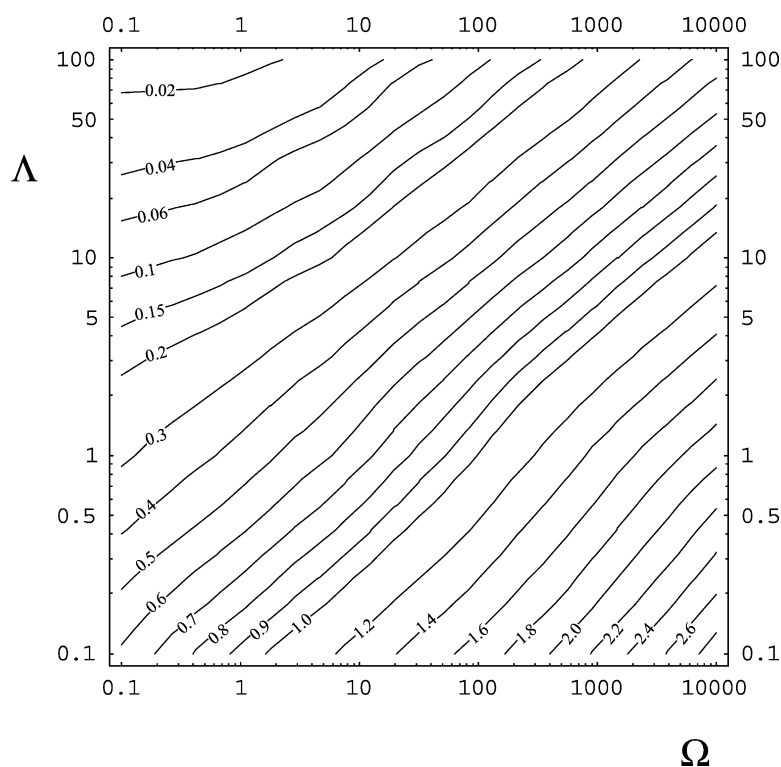


Fig. 2. Contour plot of the peak of the concentration coefficient of variation at the plume center of mass $CV_C = \sigma_C / (C)$ as function of the dimensionless variables $\Omega = I_v^2 / (I_h \alpha_{dT})$ and $\Lambda = L_1 / \sqrt{A_{11} I_h}$.

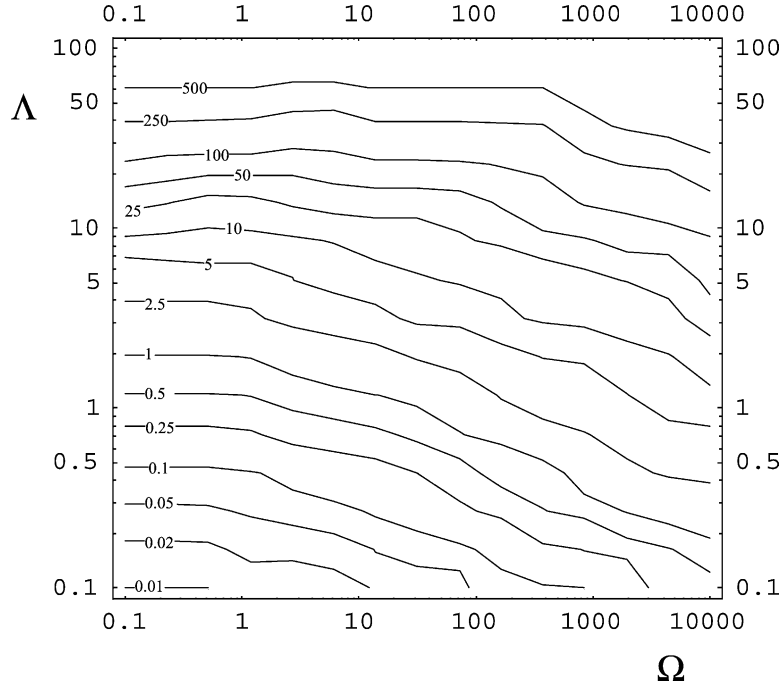


Fig. 3. Contour plot of the dimensionless setting time t^*U/H_h as function of the dimensionless variables $\Omega = I_v^2/(I_h\alpha_{dT})$ and $\Lambda = L_i/\sqrt{A_{11}I_h}$.

variables is given by the following regression formula

$$\begin{aligned} CV_C \cong & 0.435164 - 0.165986 \ln \Lambda + 0.0148895 \ln^2 \Lambda \\ & + 0.0877264 \ln \Omega + 0.00791227 \ln^2 \Omega \\ & - 0.0235874 \ln \Lambda \ln \Omega \quad (R^2 = 0.999) \end{aligned} \quad (4)$$

which is valid when the parameters Λ , Ω lie in the range of Fig. 2. The setting time t^* is represented in Fig. 3 as function of Ω and Λ .

We discuss now a few issues on the applicability of the proposed methodology. As stated in Section 3, one the major limitations of the analysis seems to be the one concerning the transverse size of the plume, which has to be large compared to the logconductivity correlation length; such conditions may not be met in some cases. The approximation takes advantage of the smallness of the transverse and vertical macro dispersivities, A_{22} and A_{33} , compared to the initial size of the plume. The validity of the solution for the usual Peclet numbers encountered in reality can be roughly assessed through the bounds $L_i/\sqrt{2A_{ii}Ut} \gg 1$ ($i = 2, 3$); in particular, a value of 5 can be assumed as

lower limit of the dimensionless quantity $L_i/\sqrt{2A_{ii}Ut}$. However, at large-time the dilution process is ruled by local diffusion, and the approximate solution is bound to be valid when $L_i/\sqrt{2\alpha_{dT}Ut} \ll 1$ ($i = 2, 3$). The two conditions above approximately determine the range of validity of the simplified methodology. The procedure is illustrated in Section 5. If the initial size of the plume obeys the condition $L_i/\sqrt{2A_{ii}Ut} \ll 1$ ($i = 1, 2, 3$), i.e. when the plume is very small compared to the characteristic scale of advection, the approximate solution of Fiori and Dagan (2000) based on point-like injection scan be used, i.e. $CV_C^2 = ((1 - \rho_{11}^2)^{-1} - 1)$, with ρ_{11} the longitudinal trajectory autocorrelation function (see Appendix A).

5. Application example

For the scope of illustration, the concentration uncertainty at the plume center for the Cape Cod experiment (Garabedian et al., 1991) is analyzed. The values for the parameters are taken from

Garabedian et al. (1991), i.e. $C_0 = 640$ mg/l, $L_1 \approx 4$ m, $I_h = 2.9$ m, $I_v = 0.19$ m, $A_{11} = 0.96$ m, $U = 0.41$ m/day. The pore-scale transverse dispersivity was inferred by Fiori and Dagan (1999) as $\alpha_{dT} = 0.0005$ m. The formation anisotropy ratio $e = I_v/I_h = 0.06$ is small enough for methodology to be employed.

The parameters Ω and Λ are calculated by using the above values, obtaining: $\Lambda = L_1/\sqrt{A_{11}I_h} = 2.4$ and $\Omega = I_v^2/(I_h\alpha_{dT}) = 24.9$. Inserting the parameter values in Fig. 2 (or Eq. (4)) and Fig. 3, we obtain the concentration coefficient of variation at the plume center and its setting time t^* , as follows: $CV_C = 0.61$ and $t^*U/I_h = 3.7$, that is $t^* = 26$ days. The values are very close to those obtained by the complete formulation of Fiori and Dagan (1999) ($CV_C = 0.68$ and $t^* = 25$ days), which, however, predicts a secondary peak at later time that is caused by transverse spreading; the latter was not captured by the present analysis.

The accuracy of results can be explained by examining the relevance of the transverse component of transport. The latter can be analyzed through the quantity $L_2/\sqrt{2A_{22}Ut}$, along the lines outlined at the end of Section 4. With $L_2 \approx 4$ m and $A_{22} = 0.018$ m (Garabedian et al., 1991), it is $L_2/\sqrt{2A_{22}Ut} > 5$ when $t < 42$ days; hence, the transverse component can be neglected below that time limit, and the approximate solution gives valid results at $t^* = 26$ days. Conversely, the PSD related factor $L_2/\sqrt{2\alpha_{dT}Ut}$ is much smaller than unity when approximately $t = O(10^5)$ days. Consequently, from t approximately equal to 42 days to the latter time limit, the accuracy of the simplified solution should deteriorate. Similar considerations can be drawn for the vertical components L_3 and A_{33} .

The concentration standard deviation can be further obtained from CV_C . With σ_C^* , $\langle C \rangle^*$, CV_C^* the exact quantities, and σ_C , $\langle C \rangle$, CV_C the approximated ones, we write: $\sigma_C^* = CV_C^* \langle C \rangle^* \approx CV_C \langle C \rangle^*$. In words, standing the robustness of the estimate of CV_C (as explained in Section 3), it is suggested to multiply the latter by the 'exact' expression for the mean concentration, Eq. (A2) (i.e. the one that also considers the transverse and vertical spreading), instead of the simplified one, Eq. (A3). The following additional data are needed for the calculation of $\langle C \rangle$ by Eq. (A2) (see Garabedian et al., 1991): $L_2 \approx 4$ m,

$L_3 \approx 1.2$ m, $A_{22} = 0.018$ m, $A_{33} = 0.0015$ m, $\alpha_{dL} = 0.005$ m (the latter was estimated in Fiori and Dagan, 1999). After substitution in Eq. (A2), for $x_1 = Ut$, $x_2 = x_3 = 0$ (i.e. the quantity is calculated at the plume center) and $t = t^* = 26$ days, we obtain: $\langle C \rangle = 214$ mg/l. The value obtained by the simplified formula (A3) is very close, i.e. $\langle C \rangle = 218$ mg/l. The concentration standard deviation is equal to: $\sigma_C = CV_C \langle C \rangle = 218 \cdot 0.61 = 131$ mg/l.

Assuming a distribution for C , an assessment of the probability of exceedance of a given concentration can be made by using the estimate values for $\langle C \rangle$ and σ_C . For example, adopting the lognormal distribution (e.g. Fiori and Dagan, 2000), the concentration at the plume center relative to 90% cumulative probability (10% exceedance probability) is equal to: $C_{90\%} = 376$ mg/l. It is pointed out that the assessment of the concentration uncertainty can be made at any time by using Fig. 1, or at any time and space by direct integration of the expressions (A3) and (A7)–(A9).

6. Summary and conclusions

A simplified analysis for determining the concentration uncertainty in aquifer transport has been proposed. The aim is to provide for a useful tool in assessing the concentration variance. An assessment, though approximate, of the concentration variance is of fundamental importance when mathematical models are used for contaminant prediction and for risk analysis, in particular when regulations are formulated in terms of maximal admissible local concentrations. The proposed methodology, valid for nonreactive solutes, is based on a few simplifications, the most important being: (i) large transverse dimensions of the injected plume compared to the logconductivity correlation lengths; as a rule of thumb, the following limit may be applied: $L_i/\sqrt{2A_{ii}Ut} \gtrsim 5$ ($i = 2, 3$); (ii) mild heterogeneity of the hydraulic properties (say, $\sigma_Y^2 < 1$), which allows for the use of the first-order analysis; (iii) highly anisotropic formations (roughly, $e = I_v/I_h < 0.2$).

The concentration coefficient of variation CV_C , which is a robust measure of uncertainty, is

obtained as a function of the following parameters: dimensionless time $t' = tU/I_h$, dimensionless location $\xi_1 = (x_1 - Ut)/\sqrt{A_{11}I_h}$, and two dimensionless parameters $\Omega = I_v^2/(I_h\alpha_{dT})$ and $\Lambda = L_1/\sqrt{A_{11}I_h}$. The solution for the mean concentration $\langle C \rangle$ is fully analytical, while CV_C require two quadratures.

In accordance with the results for the concentration coefficient of variation obtained in the past (e.g. Kapoor and Gelhar, 1994; Pannone and Kitanidis, 1999; Fiori and Dagan, 2000), results show that CV_C grows initially with time to reach a maximum value, after which the coefficient of variation decays with time and tends to zero for $t \rightarrow \infty$. The behavior reflects the competition of two mechanisms: large-scale advection generates uncertainty, which is however destroyed by dilution, that is in turn controlled by PSD. The interplay between the two mechanisms is controlled by the parameters Ω and Λ . Thus, for a fixed Ω , the peak CV_C is inversely proportional to $\Lambda = L_1/\sqrt{A_{11}I_h}$. The parameter $\Omega = I_v^2/(I_h\alpha_{dT})$ controls the effect of PSD.

The concentration coefficient of variation can be obtained by integration of expressions (A7)–(A9), together with Eq. (A3). In alternative, Fig. 1 representing CV_C as function of time can be used. The parameters required are the following: (i) longitudinal dimension of the initial plume L_1 , (ii) transverse pore-scale dispersivity α_{dT} , (iii) logconductivity longitudinal and vertical integral scales I_h , I_v , (iv) mean flow velocity U , and (v) longitudinal macro dispersivity A_{11} . The latter can be estimated by the means of the first-order analysis, as follows: $A_{11} = \sigma_Y^2 I_h$, with σ_Y^2 the logconductivity variance (see, e.g. Dagan, 1982; Gelhar and Axness, 1983).

A simpler assessment of the concentration uncertainty can be done through the peak CV_C and the setting time t^* , obtained by Figs. 2 and 3, and approximating the rising part of the curve $CV_C(t)$ by a simple linear interpolation. If the concentration standard deviation $\sigma_C = CV_C \langle C \rangle$ is needed, it is suggested to multiply the concentration coefficient of variation obtained by the simplified formulation by the ‘complete’ expression for the mean concentration $\langle C \rangle$, instead of the simplified one, as suggested in Section 5. In conclusion, the proposed methodology may serve as a simple tool to assess concentration

uncertainty in transport through heterogeneous porous formations.

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Appendix A

The mean concentration $\langle C \rangle$ for the complete formulation, i.e. without the simplifications introduced in the present work, is obtained by integration of Eq. (2). We assume a Gaussian shape for the trajectory PDF f_1 (see Section 2), i.e.

$$f_1(\mathbf{X}_t; t, \mathbf{a}) = (8\pi^3 X_{t11} X_{t22} X_{t33})^{-1/2} \times \exp \left[-\frac{(X_{t1} - Ut - a_1)^2}{2X_{t11}} - \frac{(X_{t2} - a_2)^2}{2X_{t22}} - \frac{(X_{t3} - a_3)^2}{2X_{t33}} \right] \quad (\text{A1})$$

with $X_{iii} = X_{ii} + 2D_{dii}t$ ($i = 1, 2, 3$) the second order moment of the i -component of trajectory (Dagan, 1989). Insertion of the Gaussian f_1 (A1) into Eq. (2) and further integration over a_1, a_2, a_3 , leads to the following expression for the mean concentration $\langle C \rangle$ (Fiori and Dagan, 2000)

$$\langle C(\mathbf{x}, t) \rangle = \frac{C_0}{8} \left\{ \operatorname{erf} \left[\frac{x_1 - Ut + L_1/2}{\sqrt{2X_{t11}}} \right] - \operatorname{erf} \left[\frac{x_1 - Ut - L_1/2}{\sqrt{2X_{t11}}} \right] \right\} \times \left\{ \operatorname{erf} \left[\frac{x_2 + L_2/2}{\sqrt{2X_{t22}}} \right] - \operatorname{erf} \left[\frac{x_2 - L_2/2}{\sqrt{2X_{t22}}} \right] \right\} \times \left\{ \operatorname{erf} \left[\frac{x_3 + L_3/2}{\sqrt{2X_{t33}}} \right] - \operatorname{erf} \left[\frac{x_3 - L_3/2}{\sqrt{2X_{t33}}} \right] \right\} \quad (\text{A2})$$

The approximate mean concentration is derived from Eq. (A2) by introducing the following limits: $L_2 \rightarrow \infty$, $L_3 \rightarrow \infty$, $D_{d11} \rightarrow 0$, $X_{11} = 2A_{11}Ut$, with A_{11} the longitudinal macrodispersion coefficient (Dagan, 1982;

Gelhar and Axness, 1983). The result is the following

$$\langle C \rangle = \frac{C_0}{2} \left\{ \operatorname{erf} \left[\frac{\Lambda + 2\xi_1}{4\sqrt{t'}} \right] - \operatorname{erf} \left[\frac{-\Lambda + 2\xi_1}{4\sqrt{t'}} \right] \right\} \quad (\text{A3})$$

where the following variables have been introduced: $t' = tU/I_h$ is the dimensionless time; $\xi_1 = (x_1 - Ut)/\sqrt{A_{11}I_h}$ is the dimensionless longitudinal coordinate of the point where the mean concentration is evaluated; $\Lambda = L_1/\sqrt{A_{11}I_h}$. The latter is a relevant parameter that characterizes the shearing of the plume relative to its initial size. Viewing the plume as a collection of sheets of solute, each sheet slides and separates from the surrounding ones because of convection, and Λ is roughly inverse proportional to the area of the sheet that is in contact with the uncontaminated fluid. In that area, PSD is more effective in diluting the plume, hence in reducing the concentration uncertainty.

The concentration variance is obtained by Eq. (3) following the simplifications outlined in Section 3. According to the first-order analysis, the joint PDF $f_2(\mathbf{X}_t, \mathbf{Y}_t)$ that appears in Eq. (3) is multivariate normal, and it can be split in the product of three bivariate normal distributions, one for each component of the total trajectories $\mathbf{X}_t, \mathbf{Y}_t$. Thus the trajectory joint PDF is written as

$$f_2(\mathbf{X}_t, \mathbf{Y}_t) = f_2(X_{1t}, Y_{1t})f_2(X_{2t}, Y_{2t})f_2(X_{3t}, Y_{3t}) \quad (\text{A4})$$

where the generic component $f_2(X_{it}, Y_{it})$ is a bivariate normal PDF that has the following expression

$$f_2(X_{it}, Y_{it}) = \frac{1}{2\pi\sqrt{X_{ii}^2 - Z_{ii}^2}} \times \exp \left[-\frac{(X_{it} - \langle X_{it} \rangle)^2 + (Y_{it} - \langle Y_{it} \rangle)^2}{2X_{ii}} + \frac{(X_{it} - \langle X_{it} \rangle)(Y_{it} - \langle Y_{it} \rangle)}{Z_{ii}} \right] \quad (\text{A5})$$

where $Z_{ii}(t; \mathbf{a} - \mathbf{b})$ is the two-particles trajectory second order moment, which is a measure of the degree of correlation of the trajectories X_{it} and Y_{it} of two distinct solute particles that originated at $\mathbf{x} = \mathbf{a}$ and $\mathbf{x} = \mathbf{b}$ (Fiori and Dagan, 2000).

According to the model assumptions, discussed in Section 3, the following simplifications are introduced

in Eq. (A5): $X_{22} = X_{33} = Z_{22} = Z_{33} = D_{d11} = D_{d22} = 0$, leading to the simplified joint PDFs

$$f_2(X_{t1}, Y_{t1}) = \frac{1}{2\pi\sqrt{X_{11}^2 - Z_{11}^2}} \times \exp \left[-\frac{(X_{t1} - Ut - a_1)^2 + (Y_{t1} - Ut - b_1)^2}{2X_{11}} + \frac{(X_{t1} - Ut - a_1)(Y_{t1} - Ut - b_1)}{Z_{11}} \right]$$

$$f_2(X_{t2}, Y_{t2}) = \delta(X_{t2} - a_2)\delta(Y_{t2} - b_2)$$

$$f_2(X_{t3}, Y_{t3}) = \frac{1}{4\pi D_{d33}t} \exp \left[-\frac{(X_{t3} - a_3)^2 + (Y_{t3} - b_3)^2}{4D_{d33}t} \right] \quad (\text{A6})$$

where the following substitutions have been made: $\langle X_{t1} \rangle = Ut + a_1, \langle Y_{t1} \rangle = Ut + b_1, \langle X_{t2} \rangle = a_2, \langle Y_{t2} \rangle = b_2, \langle X_{t3} \rangle = a_3, \langle Y_{t3} \rangle = b_3$.

Substitution of Eqs. (A4) and (A6) in (3) and further integration, leads to the following relation

$$\sigma_C^2(\xi_1, t) = \langle C^2 \rangle - \langle C \rangle^2 = 4C_0^2 \lim_{L'_3 \rightarrow \infty} \int_0^{L'_3} \int_0^\Lambda \Phi_1(r'_1)\Phi_3(r'_3)dr'_1 dr'_3 - \langle C \rangle^2 \quad (\text{A7})$$

where

$$\Phi_1(r'_1) = \frac{1}{4\sqrt{2\pi t'(1 - \rho_{11})}} \exp \left[-\frac{r'^2_1}{8t'(1 - \rho_{11})} \right] \times \left\{ \operatorname{erf} \left[\frac{\Lambda - r'_1 + 2\xi_1}{2\sqrt{2t'(1 + \rho_{11})}} \right] - \operatorname{erf} \left[\frac{-\Lambda + r'_1 + 2\xi_1}{2\sqrt{2t'(1 + \rho_{11})}} \right] \right\} \quad (\text{A8})$$

$$\Phi_3(r'_3) = \frac{1}{4\sqrt{2\pi t'/\Omega}} \exp \left[-\frac{r'^2_3\Omega}{8t'} \right] \times \left\{ \operatorname{erf} \left[\frac{L'_3 - r'_3 + 2\xi_3}{2\sqrt{2t'/\Omega}} \right] - \operatorname{erf} \left[\frac{-L'_3 + r'_3 + 2\xi_3}{2\sqrt{2t'/\Omega}} \right] \right\} \quad (\text{A9})$$

where $r'_1 = r_1/\sqrt{A_{11}I_h}, r'_3 = r_3/I_v, \rho_{11} = Z_{11}(t; r'_3)/X_{11}(t)$ is the trajectory autocorrelation function, $L'_3 = L_3/I_v$

the dimensionless transverse size of the initial plume, and $\Omega = I_v^2 / (I_h \alpha_{dT}) = ePe_T$. The latter is an important parameter, proportional to the transverse Peclet number, that determines the ratio between large-scale advection (which tends to increase uncertainty) and PSD (which contributes to a reduction of uncertainty); both mechanisms are controlled by the particular aquifer setting through the permeability correlation scales I_h, I_v . Thus, Ω and Λ are the two fundamental parameters for determining the concentration variance.

The autocorrelation function of longitudinal trajectories $\rho_{11} = Z_{11}/X_{11}$, that appears in Eqs. (A8) and (A9), is needed for the calculation of the concentration variance. Assuming an exponential correlation for the logconductivity field (Gelhar and Axness, 1983), the trajectory moments X_{11} and Z_{11} can be obtained by first-order expressions (14) and (15) of Fiori and Dagan (2000). Introducing in the expressions the model approximations, as done before for the concentration moments, we obtain after manipulation the following expressions for the longitudinal trajectory variance

$$\begin{aligned} \frac{X_{11}(t'; \Omega)}{\sigma_Y^2 I_h^2} &= \frac{16\Omega}{\pi^2} \int_0^\infty \int_0^1 \int_0^{\pi/2} \frac{1}{(1 + \rho^2)^2 (\rho^2 F_3^2 + \Omega^2 u^2)^2} \\ &\times \{ \Omega^3 u^2 - \Omega \rho^2 F_3^2 + \Omega^2 \rho^2 u^2 F_3 t' + \rho^4 F_3^3 t' \\ &- \Omega \exp[-\rho^2 F_3 t' / \Omega] \cdot [(\Omega^2 u^2 - \rho^2 F_3^2) \\ &\cos(\rho u t') + 2\Omega \rho u F_3 \sin(\rho u t')] \} d\rho du d\psi \end{aligned} \quad (\text{A10})$$

with $F_3 = \sin^2 \psi (1 - u^2)$.

In a similar fashion, the two-particles covariance Z_{11} is given by

$$\begin{aligned} \frac{Z_{11}(t', r'_3; \Omega)}{\sigma_Y^2 I_h^2} &= \frac{8}{\pi^2} \int_0^\infty \int_0^1 \int_0^{\pi/2} \frac{\cos[\rho r'_3 \sqrt{F_3}]}{(u^2 + \rho^2 F_3^2 / \Omega^2)(1 + \rho^2)^2} \\ &\times \left\{ 1 + \exp\left[-\frac{2\rho^2 F_3 t'}{\Omega}\right] \right. \\ &\left. - 2 \exp\left[-\frac{\rho^2 F_3 t'}{\Omega}\right] \cos(\rho u t') \right\} d\rho du d\psi \end{aligned} \quad (\text{A11})$$

Since the moments depend linearly on the logconductivity variance σ_Y^2 at first-order,

the autocorrelation function $\rho_{11} = Z_{11}/X_{11}$ does not depend on that parameter. Both X_{11} and Z_{11} are solved by numerical quadratures (see Fiori and Dagan, 2000).

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