An indirect assessment on the impact of connectivity of conductivity classes upon longitudinal asymptotic macrodispersivity

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[1] Solute transport takes place in heterogeneous porous formations, with the log conductivity, $Y = \ln K$, modeled as a stationary random space function of given univariate normal probability density function (pdf) with mean $\langle Y \rangle$, variance σ_Y^2 , and integral scale I_Y . For weak heterogeneity, the above mentioned quantities completely define the first-order approximation of the longitudinal macrodispersivity $\alpha_L = \sigma_Y^2 I_Y$. However, in highly heterogeneous formations, nonlinear effects which depend on the multipoint joint pdf of Y, impact α_I . Most of the past numerical simulations assumed a multivariate normal distribution (MVN) of Y values. The main aim of this study is to investigate the impact of deviations from the MVN structure upon α_L . This is achieved by using the concept of spatial correlations of different Y classes, the latter being defined as the space domain where Y falls in the generic interval $[Y,Y+\Delta Y]$. The latter is characterized by a length scale $\lambda(Y)$, reflecting the degree of connectivity of the domain (the concept is similar to the indicator variograms). We consider both "symmetrical" and "non-symmetrical" structures, for which $\lambda(Y') = \lambda(-Y')$ (similar to the MVN), and $\lambda(Y') \neq \lambda(-Y')$, respectively, where $Y = Y - \langle Y \rangle$. For example, large Y zones may have high spatial correlation, while low Y zones are poorly correlated, or vice versa. The impact of $\lambda(Y)$ on α_L is investigated by adopting a structure model which has been used in the past in order to investigate flow and transport in highly heterogeneous media. It is found that the increased correlation in the low conductive zones with respect to the high ones generally leads to a significant increase in α_L , for the same global I_Y . The finding is explained by the solute retention occurring in low Y zones, which has a larger effect on solute spreading than high Y zones. Conversely, α_L decreases when the high conductivity zones are more correlated than the low Y ones. Dispersivity is less affected by the shape of $\lambda(Y)$ for symmetrical distributions. It is found that the range of validity of the first-order dispersivity, i.e., $\alpha_L = I_Y \sigma_Y^2$, narrows down for non-symmetrical structures.

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1. Introduction

[2] Spreading of solutes in advective transport in heterogeneous porous formations is governed by spatial variations of the hydraulic conductivity K. A large body of work has been published in the last three decades in order relate solute transport to the underlying heterogeneous medium structure. Assuming a stationary log conductivity, $Y = \ln K$, random distribution, one of the earliest and successful approaches proposed in the past is the first-order analysis, which is formally valid for small variations of Y around the mean, i.e., $\sigma_Y^2 \ll 1$, where σ_Y^2 is the log conductivity variance. One of the main results of the first-order analysis is the well-known formula for the longitudinal asymptotic macrodispersivity in mean uniform flows, $\alpha_L = \sigma_Y^2 I_Y$, where I_Y is the longitudinal integral scale of Y [see, e.g., Dagan, 1989; Rubin, 2003]. This result shows that α_L depends on the second-order statistical moments of the Y field through the variance and

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the integral scale of Y, while the precise shape of the Y covariance plays a minor role. Studies based on detailed numerical simulations of solute transport in isotropic formations have shown that the first-order result is a valid approximation for relatively large values of σ_Y^2 , up to $\sigma_Y^2 \simeq 2$ [see, e.g., Bellin et al., 1992; de Dreuzy et al., 2007]. In most of the existing numerical studies, the selected log conductivity field was multi-Gaussian, being generated with the aid of a multivariate normal (MVN) distribution of Y values at different points. Even if this is a convenient working assumption, it is quite restrictive, and (more important) it is not generally supported by field data. Indeed, while the latter indicate that the univariate f(Y) is normal in many aquifers [see, e.g., Dagan, 1989], they are too scarce to enable one to identify the multi-point joint pdf. Here we adopt a different structure model which can be related to natural porous formations [see, e.g., Hoeksema and Kitanidis, 1985; Ritzi, 2000; Ritzi et al., 2000; Labolle and Fogg, 2001]. In addition, it allows for freedom of characterization of heterogeneity of natural porous formations which is lacking in the MVN model. Thus, it is well-known [Journel, 1983] that by selecting MVN fields, the extreme (low and high) values of Y become less spatially correlated than conductivity values close to the mean.

- [3] Deviations from the first-order α_L have been observed in numerical simulations of transport in highly heterogeneous formations (HHF) [Jankovic et al., 2003; De Dreuzy, 2007]. Such differences are related to the statistical moments which are not taken into account by the first-order approach, as expressed for instance by the M-point joint pdf of Y, with M > 2, or equivalently by the different spatial correlation structures of different classes of the hydraulic conductivity. The latter case may lead to random Y fields quite different from MVN, for given f(Y) and C_Y . This finding has stimulated debates on the range of validity of the perturbation expansions, and alternative approaches have been employed [see, e.g., Dentz and Tartakovsky, 2008].
- [4] There is an increasing evidence that the above mentioned properties may have an important role in transport in HHF. A common approach is to investigate formations of given univariate Y and integral scale I_Y , which differ however from the MVN at higher order. This has previously been done by using, within a numerical context, the concept of spatial connectivity of different Y classes. Thus, the distribution of the residual $Y' = Y - \langle Y \rangle$ was divided into classes defined by intervals ΔY . The space domain corresponding to each class was characterized by an integral scale reflecting the degree of correlation of the domain. The concept is similar to the often employed indicator covariance [Journel, 1983], which is sometimes related to the concept of connectivity, although the topic is still a matter of debate [see, e.g., Western et al., 1998; Knudby and Carrera, 2005]. For example, classes in the interval $[Y', Y' + \Delta Y']$ and [-Y', -Y' - ΔY may have the same correlation length scales, coined here as "symmetrical" structures (not necessarily equal to those pertaining to the MVN). A more general structure is the "non-symmetrical" one, in which the two above classes have distinct correlation lengths. For example, large K zones may have high spatial correlation while low K zones are poorly correlated, or vice versa.
- [5] A few studies have examined the impact of different correlation structures, in particular for the non-symmetrical case, i.e., the presence of different connectivities of Y' and

- -Y classes, while obeying the same univariate distribution [Desbarats, 1990; Wen and Gomez-Hernandez, 1998; Guswa and Freyberg, 2002; Zinn and Harvey, 2003; Knudby et al., 2006]. A conclusive result of all studies was that α_L may depend on the particular choice of the Y structure, for the same f(Y) and global integral scale I_Y . This is, of course, a nonlinear effect which is not captured by first-order solutions. The aforementioned studies were of a numerical nature, mostly dealing with 2D transport, and were limited to a few particular cases and computational techniques.
- [6] The aim of the present study is to investigate this issue indirectly by adopting a structure model which has been used in the past to model flow and transport in highly heterogeneous media [Dagan et al., 2003]. The advantage of this model is that it permits to derive an approximate semi-analytical solution for α_L (which is employed in the present work) to provide useful insight on the impact of spatial correlation of Y classes in a simple and systematic manner. The model is able to represent any random heterogeneous medium of given f(Y), and it lends itself to useful generalizations for any correlation structure of the Y classes. Unlike previous investigations [e.g., Dagan et al., 2003], which assumed a unique correlation scale of all Y classes, the present work accounts for a variable degree of correlation of different Y classes in order to mimic different degrees of connectivity. It is worth to underline that, because the different correlation lengths are the same for any spatial direction, the model represents Y fields which are globally isotropic.

2. Mathematical Framework

- [7] We consider spreading of large (ergodic) plumes in three-dimensional heterogeneous formations of isotropic K spatial distribution. Flow is of constant mean velocity U and the flow domain is much larger than any of the correlation scales of generic log conductivity classes; hence, Y is modeled as a stationary, isotropic random field, and we assume that there are no structures (such as lenses of low/ high Y) spanning the entire medium. Such a problem was already solved for a medium consisting of spherical inclusions of different conductivities characterized by the same correlation length for any Y class [Dagan et al., 2003]. Thus, several quantities of interest, such as the dispersion coefficients as well as the breakthrough curves, as function of the conductivity statistical parameters were computed [e.g., Fiori et al., 2003]. The model results were tested against detailed 3D transport numerical simulations in HHF [Jankovic et al., 2003]. More recently, a good agreement with the numerical results of de Dreuzy et al. [2007] was found [Fiori et al., 2008].
- [8] The primary focus of this work is the calculation of the asymptotic longitudinal dispersivity α_L reached by an ergodic plume of a conservative solute after a large travel distance from the injection zone. The general expression of α_L has been derived by *Dagan et al.* [2003], and its final expression is written below:

$$\alpha_{L} = \frac{9}{16} n \int_{0}^{\infty} \int_{-\infty}^{\infty} dR \, dY \frac{\kappa}{2 + \kappa} \frac{X_{M}^{2}(\kappa)}{R} f(Y, R) \qquad \text{with}$$

$$\kappa = \frac{K}{K_{ef}}, \tag{1}$$

where K_{ef} denotes the effective medium conductivity, which is determined from [Dagan, 1989]

$$\int dK f(K) \frac{K_{ef} - K}{2K_{ef} + K} = 0.$$
(2)

Equation (1) is based on determining α_L by summation of contributions from different isolated spheres of conductivity K surrounded by a medium of conductivity K_{ef} . The quantity

$$X_{M}(\kappa) = \frac{2}{3}R(\kappa - 1)\left[\frac{2}{\kappa} + \frac{3}{2 + \kappa} {}_{2}F_{1}\left(\frac{2}{3}, 1, \frac{5}{3}, 2\frac{1 - \kappa}{2 + \kappa}\right)\right]$$
(3)

represents the trajectory residual of the centerline crossing an inclusion (${}_{2}F_{1}$ being the hypergeometric function), whereas f(Y, R) is the joint distribution of Y and the sphere radius R. The latter is proportional to the integral scale pertaining to a given class of hydraulic conductivity, and generally it is correlated with Y. This is observed both at regional (see data collected by *Hoeksema and Kitanidis* [1985]), and at formation scale [see Ritzi, 2000; Ritzi et al., 2000; Labolle and Fogg, 2001] where it is seen that blocks with different conductivity may have different sizes. The particular case considered in the past of constant $R = R^*$ is mathematically expressed by replacing $f(Y, R) = f(Y)\delta(R - R^*)$ in equation (1). Here we extend the analysis to variable R, and to further simplify the problem, without loss of generality, we assume that Y and R are perfectly correlated, i.e., $f(Y, R) = f(Y)\delta[R - \lambda(Y)]$, where $\lambda(Y)$ is a deterministic, given, radius distribution. In such a manner, a given Y class is characterized by an unique size $R = \lambda(Y)$ of its inclusions, proportional to the correlation length for that class. For example, classes of high conductivity may have a larger spatial correlation scale (i.e., larger R) than those of low conductivity, in line with the connectivity approach mentioned in the section 1. The same assumption was made by Firmani et al. [2009] for the calculation of the effective conductivity in heterogeneous 2D formations. Introduction of the above mentioned approximation into equation (1) yields

$$\alpha_L = \frac{9}{16} n \int_{-\infty}^{\infty} dY \frac{f(Y)}{\lambda(Y)} \frac{\kappa X_M^2(\kappa)}{2 + \kappa}.$$
 (4)

This simple expression is employed in the sequel to derive the dispersivity for a few noteworthy structures pertaining to particular $\lambda(Y)$. The simple numerical quadrature appearing in equation (4) is carried out for the widely employed normal f(Y).

3. Dispersivity in Non-symmetrical Structures

[9] As mentioned in section 1, non-symmetrical structures are characterized by a different correlation length for high and low conductivity. In our model, this is expressed by $\lambda(Y') \neq \lambda(-Y')$. The simplest distribution, which still preserves the main features of interest, is the one in which the low and high Y are characterized by different and constant values:

$$\lambda(Y') = \begin{cases} R_H & \text{for } Y' \ge 0 \\ R_L & \text{for } Y' < 0. \end{cases}$$
 (5)

It is easy to verify [see *Dagan et al.*, 2003] that the above medium is characterized by a global correlation scale proportional to $\frac{R_L + R_H}{2}$.

[10] The longitudinal dispersivity can be easily calculated after introducing equation (5) into equation (4) for f(Y) normally distributed, and performing the quadrature. The final result for the dimensionless dispersivity $\frac{\alpha_L}{I_Y}$ depends only on the log conductivity variance σ_Y^2 and the ratio, $\xi = \frac{R_H}{R_L}$ (0 < ξ < ∞), between the correlation scale of the high K and that of the low K.

[11] Figure 1 displays the dimensionless dispersivity $\frac{\alpha_I}{nl_Y}$ (where n is the inclusions volume density, to be taken as n=1) as function of the log conductivity variance σ_Y^2 for a few values of the ratio $\xi < 1$, i.e., for low conductivities zones more connected than the high ones. The first-order result $\frac{\alpha_I}{nl_Y} = \sigma_Y^2$ is also shown for reference. The case $\xi = 1$ (symmetrically correlated structure of uniform R) reproduces the result already presented in the past [see, e.g., *Fiori et al.*, 2003, Figure 3]. It is observed that the increase in the correlation of the low conductive zones with respect to the high ones leads to a significant increase of the longitudinal dispersivity.

[12] The above finding can be explained by considering the different contributions to dispersivity related to large | Y'|. In fact, for high Y' > 0, the sphere's interior velocity $\frac{V_{in}}{U}$ = $\frac{3\kappa}{\kappa+2} > 1$, and the related contributions to α_L in equation (4) are enhanced. However, the fluid velocity reaches the upper bound $\frac{V_{in}}{U} = 3$ for $Y \to \infty$, due to the constraining effect of the surrounding medium, and X_M reaches a finite value as well. On the contrary, $\frac{V_{lm}}{U} \to 0$ for $Y \to -\infty$ $(\kappa \to 0)$. As a consequence, solute particles may spend a very long time in low conductive inclusions, leading to $X_M \rightarrow \infty$ in equation (4) and to a considerable increase of dispersivity (this mechanism is thoroughly discussed by *Dagan et al.* [2003]). The latter is further enhanced when the low conductive areas are highly connected, as in the case $\xi < 1$. Hence, increasing the correlation of low conductive zones implies the existence of large volumes in which the solute hold-up mechanism manifests, leading to a larger dispersion. A similar effect was found by La Bolle and Fogg [2001].

[13] Of particular interest is the behavior for small σ_Y^2 (see the small inset of Figure 1). It was already observed and discussed in the past [Dagan et al., 2003] that for the symmetrical structure ($\xi = 1$) the first order approximation is valid for a broad range of σ_Y^2 values, confirming results based on numerical simulations [Bellin et al., 1992]. This effect was attributed to the mutual cancellation of errors associated with the first-order expansion of the trajectories for Y' > 0 and Y' < 0, which applies to symmetrical systems [Dagan et al., 2003]. The cancellation was shown to stem from the fact that in the neighborhood of Y' = 0, the firstorder approximation of the integrand in equation (4) with R =const overestimates the exact solution for Y' > 0 and underestimates it for Y' < 0. The break of symmetry in equation (5) for $\xi \neq 1$, diminishes the effect, i.e., the errors are no longer balanced at the same extent. As a consequence, the accuracy of the first-order approximation of α_L starts to deteriorate for smaller values of the log conductivity variance, say $\sigma_Y^2 >$ 0.3. This finding is somewhat similar to the results of Wen and Gomez-Hernandez [1998] and Fogg [1990], who also observed a significant impact of the non-symmetrical Y structure even for small σ_Y^2 .

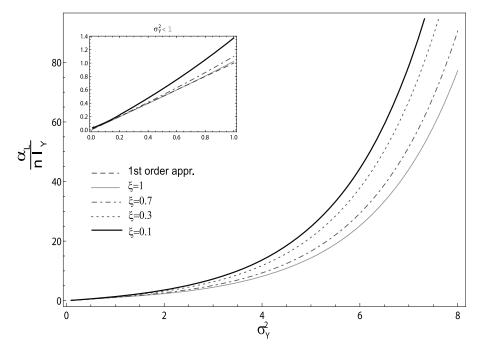


Figure 1. The asymptotic longitudinal dimensionless dispersivity $\frac{\alpha_I}{I_Y}$ as function of the log conductivity variance σ_Y^2 for a non-symmetrical $Y = \ln K$ structure (equation (5)) for the case of low Y more correlated than high Y.

[14] The opposite case, of high conductivity zones more correlated than the low-K ones, i.e., $\xi \ge 1$, is illustrated in Figure 2. It is seen that this non-symmetrical Y structure generally leads to a decrease in the dispersivity. The explanation is that the contribution of high K inclusions is less than those of low K, and the increase in the connectivity

of large K elements does not compensate for the dispersivity reduction caused by the decreased connectivity of the low conductive inclusions. Thus, the final result is an overall decrease of α_L . We note an early departure from the first-order solution also for this case, which is again explained by

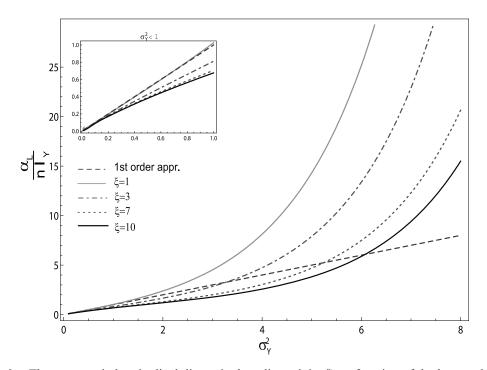


Figure 2. The asymptotic longitudinal dimensionless dispersivity $\frac{\alpha_I}{I_Y}$ as function of the log conductivity variance σ_Y^2 for a non-symmetrical $Y = \ln K$ structure (equation (5)) for the case of low Y less correlated than high Y.

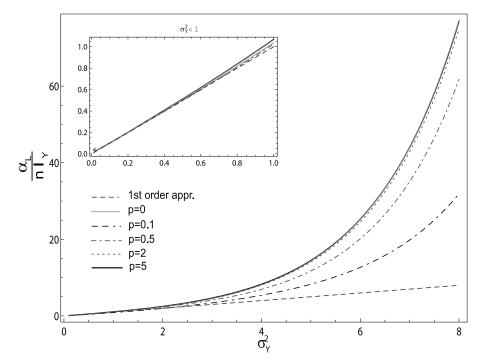


Figure 3. The asymptotic longitudinal dimensionless dispersivity $\frac{\alpha_I}{I_Y}$ as function of the log conductivity variance σ_Y^2 for a symmetrical $Y = \ln K$ structure (equation (6)) for the case of extreme Y less correlated than Y close to the mean $(\frac{R_0}{R_\infty} = 10)$.

the role played by the break of the symmetry in the cancellation of errors, as discussed above.

4. Dispersivity in Symmetrical Structures

[15] In this section we analyze correlation structures of Y which are symmetrical with respect to Y, but not of uniform R. The MVN is an important and widely adopted such configuration, with log conductivity values close to the mean more correlated than the extremes. In our approach, the symmetry condition translates in the identity $\lambda(Y') = \lambda(-Y')$. For illustration purposes, we use the following relationship:

$$\lambda(Y') = R_{\infty} + (R_0 - R_{\infty}) \exp\left(-pY'^2\right),\tag{6}$$

where $0 and <math>\lambda(0) = R_0$, $\lambda(\infty) = R_\infty$. At both the extreme values of the parameter p, i.e., p = 0 and $p = \infty$, the medium degenerates into one with uniform correlation for all classes of conductivity ($R = R_0$ and $R = R_\infty$, respectively). For any other p value there is a smooth transition between $R = R_0$ and $R = R_\infty$. The dimensionless dispersivity, $\frac{\alpha_L}{I_T}$, is calculated by equation (4), and it depends now on the three parameters: σ_T^2 , p and the ratio $\frac{R_0}{R}$.

meters: σ_Y^2 , p and the ratio $\frac{R_0}{R_\infty}$. [16] Figure 3 displays the dimensionless dispersivity $\frac{\alpha_L}{nl_Y}$ as function of σ_Y^2 for the fixed ratio $\frac{R_0}{R_\infty}=10$, and a few values of the coefficient p. Thus K values close to the mean are much more correlated than the extremes, similar to the MVN case, for any finite p. Figure 3 shows a decrease in α_L for intermediate values of p, between 0 and ∞ , for the given large difference between the correlation of Y classes expressed by $\frac{R_0}{R_\infty}$. Such a decrease appears at the highest values of σ_Y^2 , and it is mostly a consequence of filtering out the low-

conductive inclusions, which mostly contribute to solute spreading. This is due to the diminishing of their radius λ relative to R_0 . The effect is pronounced for small values of p, for which the smallest Y inclusions are filtered out while I_Y is large (see equation (6)). At any rate, the differences between the curves are generally smaller than in the previously analyzed non-symmetrical case.

[17] The impact of the symmetrical distribution of R is even smaller when the extreme values are more correlated than the log conductivity close to the mean (case $\frac{R_0}{R_{\infty}} = 0.1$, represented in Figure 4). In this case, the partial filtering out of the inclusions with log conductivity close to the mean (which usually cause a limited spreading) does not change much the overall plume dispersion.

[18] It is interesting to note that for different degree of correlation expressed by the symmetrical $\lambda(Y)$, the mutual cancellation of errors still occurs, as is the case of the first-order approximation, as described in the previous section. Therefore, the first-order result $\alpha_L = I_Y \sigma_Y^2$ is accurate for a broad range of values for σ_Y^2 , in agreement with the numerical results based on MVN fields.

5. Summary and Conclusions

[19] In this work, the impact of the connectivity of hydraulic conductivity on solute transport is investigated by extending the theoretical framework on flow and transport in highly heterogeneous media developed by $Dagan\ et\ al.$ [2003]. The focus is on the calculation of the asymptotic longitudinal dispersivity α_L , reached by an ergodic plume of a conservative solute after a large travel distance from the injection zone. We have adopted the concept of spatial connectivity of different Y classes, each one characterized by dividing the residual $Y' = Y - \langle Y \rangle$ into classes defined by

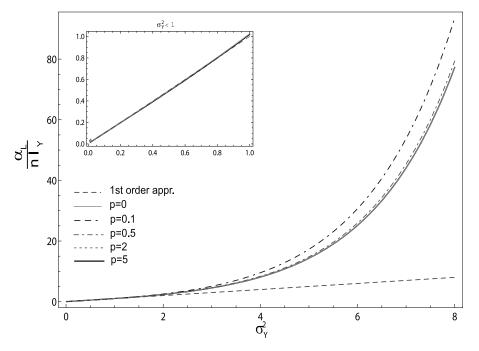


Figure 4. The asymptotic longitudinal dimensionless dispersivity $\frac{\alpha_I}{I_Y}$ as function of the log conductivity variance σ_Y^2 for a symmetrical $Y = \ln K$ structure (equation (6)) for the case of extreme Y more correlated than Y close to the mean $(\frac{R_0}{R_\infty} = 0.1)$.

intervals ΔY . The space domain corresponding to each class was characterized by an integral scale reflecting the degree of correlation of the domain. We have analyzed both non-symmetrical structures, in which large K zones have high spatial correlation while low K zones are poorly correlated, or vice versa, and symmetrical structures, where the extremes have the same correlation scale (similar to the MVN field).

[20] For the non-symmetrical case, the increase in the correlation of the low conductive zones with respect to the high conductive ones leads to a significant increase in the longitudinal dispersivity α_L (Figure 1). The opposite case, of high-conductivity zones more correlated than the low-conductivity ones (Figure 2), leads to a relatively modest decrease in the dispersivity. These findings are explained by the solute retention occurring in low Y zones, which has a larger effect on spreading than high Y zones.

[21] For symmetrical structures, when the conductivities close to the mean are more correlated than the extremes (similar to the MVN case), α_L shows a decrease for certain configurations, provided that there is a marked difference between the correlation of Y classes. The decrease appears at the highest σ_Y^2 and it is mostly a consequence of the filtering out the low conductive inclusions, which are those that mostly contribute to spreading. However, the change of dispersivity is generally much smaller than the non-symmetrical case. The impact is even smaller when the extreme values are more correlated than the mean values.

[22] It is also found that the break of symmetry in the log conductivity structure leads to a narrowing down of the validity of the first-order dispersivity $\alpha_L = I_Y \sigma_Y^2$. The validity of the first-order approximation was in the past attributed to the mutual cancellation of errors associated with the first-order expansion of the trajectories for Y' > 0 and Y' < 0, which applies to symmetrical Y structures [Dagan et al.,

2003]. The break of symmetry of Y diminishes the effect, i.e., the errors are no longer balanced by symmetry to the same extent. As a consequence, the accuracy of the first-order approximation for α_L starts to deteriorate for smaller values of the log conductivity variance. Conversely, the cancellation of errors is still effective for symmetrical structures (Figures 3 and 4), for which the first-order α_L remains a valid approximation for a wide range of σ_Y^2 .

[23] We wish to emphasize that the results obtained here are valid for three-dimensional multi-indicator random log conductivity fields, and their extension to other structures, like the commonly employed multi-Gaussian one, is not straightforward. Previous studies based on two dimensional numerical simulations of multi-Gaussian conductivity fields, with different correlation scales of the Y classes, seem to indicate a different behavior from the one observed here [e.g., Wen and Gomez-Hernandez, 1998; Zinn and Harvey, 2003]. Besides the different random fields, which greatly differ for strongly heterogeneous formations, there are other possible reasons for the above observed difference, such as the numerical issues involved when solving highly heterogeneous flows [see, e.g., Fiori et al., 2008] as well as the presence of Y classes with correlation scales which are not much smaller than (or even comparable with) the domain size leading to well connected, domain-spanning structures. A thorough analysis of the differences between our results, and previous works is beyond the scopes of this note, and it requires further investigations.

[24] Finally, the proposed model is applicable to stationary (statistically homogeneous) porous media. However, there are numerous geological formations where the high degrees of heterogeneity stem from the presence of distinct hydrofacies, therefore defying stationary statistical parameterizations. For such formations, other models (such as the one

proposed by Winter and Tartakovsky [2002]) may be more representative.

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