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Localized Point Mixing Rate Potential in Heterogeneous Velocity Fields

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Abstract Mixing is driven by an interplay between diffusion and flow-induced concentration gradients. Accurately describing the effect of flow heterogeneity on the mixing state of a plume is a challenging problem that has important repercussions on the modeling of plume dilution and chemical reactions. In this technical note, we propose a simple, semi-analytic measure to quantify the local mixing potential at a point based on the local properties of the flow-induced strain. Specifically, it is the trace of the local strain matrix squared, $\mathrm{Tr}(\boldsymbol{s}^2)$, which we demonstrate controls mixing from a point source over small times. Due to its mathematically similar influence to a shear flow on mixing, we propose an *ansatz* to attempt to use this metric as a predictor for mixing. We test its performance via random walk particle tracking simulations in heterogeneous Darcy flow through lognormal permeability fields. The *ansatz* appears to work better and better the more heterogeneous the flow, unlike more traditional approaches that rely on weak heterogeneity assumptions. While we cannot rigorously demonstrate why this is, we find it sufficiently promising that it may guide future model development.

Keywords Mixing · Heterogeneous media · Flow deformation metrics

1 Introduction

Mixing is the process that locally homogenizes a solute plume. It is controlled by the interplay between the formation of concentration gradients by velocity gradients, and their smoothing by diffusion. A fluid parcel moving in a heterogeneous flow suffers deformation; for an incompressible flow, stretching along one direction is always accompanied by compression

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in another, enhancing mixing (Le Borgne et al. 2010; Bolster et al. 2011a, b; Engdahl et al. 2014). Understanding this link between flow-induced deformation and solute dilution is key to understanding and quantifying mixing in real-world systems in an efficient and accurate manner.

Quantifying mixing is a problem of great practical and theoretical interest (Dentz et al. 2011). For one, mixing brings different chemical species together and is the main driver of multispecies, rate-limited reactions; thus, any method that purports to quantify collocation-driven chemical reactions and/or mesoscale concentrations must accurately capture mixing properties (De Simoni et al. 2005, 2007; Chiogna et al. 2012; de Anna et al. 2013; Benson and Meerschaert 2008; Porta et al. 2016; Sanchez-Vila et al. 2010; Paster et al. 2014, 2013, 2015). Real-world aquifers are often heterogeneous, leading to complex and scale-dependent solute plume structure. Resolving all relevant scales is demanding, if not impossible, and upscaled descriptions become necessary. However, many current models fail to capture the complex scale dependency of plume structure adequately (Burchard and Rennau 2008; Le Borgne et al. 2011, 2010).

In recent years, several theories for mixing have emerged, improving our understanding and predictive ability (Le Borgne et al. 2015; de Barros et al. 2012; Bolster et al. 2011b; Le Borgne et al. 2010, 2013; Borgne et al. 2014). However, many limitations still remain. While it is clear that the underlying flow structure can influence mixing, a strong quantitative link between well-defined and easily computable flow properties and mixing remains elusive, especially in highly heterogeneous settings. For weakly heterogeneous systems, classical perturbation approaches appear to do well (de Barros et al. 2015; Bolster et al. 2011b). While some quantitative approaches work for highly heterogeneous systems [e.g., lamellar models (Le Borgne et al. 2015)], they are typically not as well suited to describe point sources, requiring a line or planar source for sufficient sampling of the background flow at all times. The goal here is to provide an efficient measure of mixing based on local flow properties, and shed light on fundamental flow and transport elements that control it.

Several measures to describe the mixing state of a plume exist. Here we focus on the dilution index (Kitanidis 1994), an entropy-based measure, which represents an effective volume occupied by the plume. It is closely related to other measures of mixing [e.g., scalar dissipation (Le Borgne et al. 2010)]. To quantify and explain how these global measures are driven by local flow properties, a number of metrics have been proposed. In 2d, this includes the Okubo–Weiss parameter (de Barros et al. 2012), based on the eigenvalues of the local strain and rotation. Its time history as a plume moves through a medium correlates with the behavior of the dilution index (de Barros et al. 2012), but a strong quantitative link over long times is yet to be identified. Another measure in the same vein is the largest eigenvalue of the right Cauchy–Green deformation tensor (Engdahl et al. 2014). Intuitively, these measures rest on the idea that it is the local stretching, compressing, and shearing effects induced by the underlying flow field that play the main role in driving dilution. In this technical note, we develop a new measure of dilution based on a similar concept, the time history of the plume as it samples the shearing effects of the local flow field.

2 Equations of Motion

We assume transport at the scale of interest can be described by the advection dispersion equation (ADE),

$$\frac{\partial c}{\partial t} + \nabla \cdot [\boldsymbol{v}(\boldsymbol{x})c] - D\nabla^2 c = 0\,. \tag{1}$$

The underlying velocity field $\boldsymbol{v}$ is steady and incompressible. For simplicity, and to focus on the role of a heterogeneous advection term, we consider dispersion as homogeneous and isotropic, characterized by a constant dispersion coefficient D. Additionally, we consider this equation to be in dimensionless form. Following de Barros et al. (2012) and approximating the local velocity field by a Taylor expansion around a point within a moving coordinate system, rendering that point stationary, the ADE may be approximated as

$$\frac{\partial c}{\partial t} + \nabla \cdot [\boldsymbol{\varepsilon}\boldsymbol{x}c] - D\nabla^2 c = 0, \tag{2}$$

where $\boldsymbol{\varepsilon}$ is the deformation tensor at $\boldsymbol{x}$, $\varepsilon_{ij} = (\nabla \boldsymbol{v})_{ji}|_{\boldsymbol{x}}$. For a pulse initial condition, $c(\boldsymbol{x}, t = 0) = \delta(\boldsymbol{x})$, the solution is (e.g., Van Kampen 2007)

$$c(\boldsymbol{x}, t) = \frac{\exp\left\{-\boldsymbol{x} \cdot [2\boldsymbol{\kappa}]^{-1}\boldsymbol{x}\right\}}{\sqrt{(2\pi)^d \det[\boldsymbol{\kappa}(t)]}}, \tag{3}$$

where $\boldsymbol{\kappa}$ solves equation

$$\frac{\mathrm{d}\boldsymbol{\kappa}}{\mathrm{d}t} = \boldsymbol{\varepsilon}\boldsymbol{\kappa}^T + \boldsymbol{\kappa}\boldsymbol{\varepsilon}^T + 2\boldsymbol{D}. \tag{4}$$

For isotropic dispersion $\boldsymbol{D} = D\delta_{ij}$, where δ_{ij} represents the Kronecker delta. The superscript T denotes matrix transposition. For this setup, the dilution index is given by

$$E(t) = \exp\left(-\int c(\boldsymbol{x}, t)\log[c(\boldsymbol{x}, t)]\,\mathrm{d}\boldsymbol{x}\right) = (2\pi e)^{d/2}\sqrt{\det[\boldsymbol{\kappa}(t)]}, \tag{5}$$

where d is the number of spatial dimensions. The dilution index is a global measure of dilution (i.e., integrated over the whole space). Up to here our approach follows that of de Barros et al. (2012), but in what follows we deviate from it.

Solving (4) directly is challenging, so we take an alternate Lagrangian approach (Risken 1989; Van Kampen 2007). The equivalent Langevin equation to (2) for the movement of a solute particle is

$$\frac{\mathrm{d}\boldsymbol{x}}{\mathrm{d}t} = \boldsymbol{\varepsilon}\boldsymbol{x} + \sqrt{2D}\boldsymbol{\xi}, \tag{6}$$

where $\boldsymbol{x}$ is the position of a particle and $\boldsymbol{\xi}$ is a d-dimensional vector of independent, delta-correlated Gaussian-distributed Langevin forces with zero mean and unit variance. Note that this is a Langevin equation for an Ornstein–Uhlenbeck process in multiple dimensions. Its solution is

$$\boldsymbol{x}(t) = \int_0^t e^{\boldsymbol{\varepsilon}u}\boldsymbol{\xi}(t-u)\,\mathrm{d}u. \tag{7}$$

In our moving coordinate frame, the first moments of the position are all zero, i.e., $\mu_i(t) = \langle x_i(t)\rangle = 0$, and variances $\sigma_{ij}(t) = \langle x_i(t)x_j(t)\rangle$ play the role of κ, as they are also governed by (4); they both represent the spread of the solute plume around its mean position. Thus, the dilution index $E = (2\pi e)^{d/2}\sqrt{\det\boldsymbol{\sigma}}$. From (7),

$$\boldsymbol{\sigma}(t) = 2D\int_0^t e^{\boldsymbol{\varepsilon}u}e^{\boldsymbol{\varepsilon}^T u}\,\mathrm{d}u. \tag{8}$$

We remind the reader at this point that the exponential of a matrix $\boldsymbol{A}$ is defined by its Taylor-series expansion (Bellman 1997):

$$e^{\boldsymbol{A}} = \sum_{i \geqslant 0} \frac{\boldsymbol{A}^i}{i!} = \mathbb{1} + \boldsymbol{A} + \frac{\boldsymbol{A}^2}{2} + \cdots, \tag{9}$$

where $\mathbb{1}$ is the identity matrix, and the commutator of two matrices $\boldsymbol{A}$ and $\boldsymbol{B}$ is given by:

$$[\boldsymbol{A}, \boldsymbol{B}] = \boldsymbol{A}\boldsymbol{B} - \boldsymbol{B}\boldsymbol{A}. \tag{10}$$

Note that evaluating the above integral is complicated by the fact that the integrand is the product of matrix exponentials. For matrix exponentials, if $[\boldsymbol{A}, \boldsymbol{B}] = 0$ (i.e., matrices $\boldsymbol{A}$ and $\boldsymbol{B}$ commute), then $e^{\boldsymbol{A}} e^{\boldsymbol{B}} = e^{\boldsymbol{A}+\boldsymbol{B}}$; otherwise this may not hold. Recall that the deformation tensor $\boldsymbol{\varepsilon}$ can be decomposed into strain $\boldsymbol{s} = (\boldsymbol{\varepsilon} + \boldsymbol{\varepsilon}^T)/2$ (symmetric part) and rotation $\boldsymbol{\xi} = (\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^T)/2$ (antisymmetric part), $\boldsymbol{\varepsilon} = \boldsymbol{s} + \boldsymbol{\xi}$. Thus, $\boldsymbol{\sigma}$ is most easily computed when $[\boldsymbol{\varepsilon}, \boldsymbol{\varepsilon}^T] = 2[\boldsymbol{\xi}, \boldsymbol{s}] = 0$. While this provides some interesting end-member cases, for arbitrary heterogeneous flows, we do not anticipate $[\boldsymbol{\varepsilon}, \boldsymbol{\varepsilon}^T] = 0$ to hold.

However, even when they do not commute, closed form analytical solutions for $\boldsymbol{\sigma}$ can be found, see "Appendix." In two dimensions, the final result for the dilution index agrees with previous derivations (de Barros et al. 2012). In three dimensions, the result depends not only on the strain and rotation eigenvalues, but also on the orientation of vorticity relative to the principal axes of strain. The result may be obtained in closed form, but it is complex and provides little physical insight.

3 Linking Dilution to Flow Properties

For late times, the local expansion of the velocity field $\boldsymbol{v} = \boldsymbol{\varepsilon}\boldsymbol{x}$ around the injection point becomes dubious as particles will move far away from this point, violating assumptions in the expansion. Thus, let us focus on early times. Consider (8) and evaluate the integral through an early-time Taylor expansion of the exponentials ($e^{\boldsymbol{\varepsilon} t} \approx \mathbb{1} + \boldsymbol{\varepsilon} t + \boldsymbol{\varepsilon}^2 t^2/2$). This leads to:

$$E(t) = \frac{(4\pi e D t)^{d/2}}{V_0} \sqrt{1 + \frac{\mathrm{Tr}(\boldsymbol{s}^2) t^2}{6}}. \tag{11}$$

Here, the important term to focus on is $\mathrm{Tr}(\boldsymbol{s}^2)$, the trace of the square of the strain matrix. In (11), the leading-order behavior is controlled by dispersion. A quadratic contribution is missing; it corresponds to the combined effect of dispersion and compressibility, which is absent for incompressible flow. Note also how to this order rotation plays no role: Rotation by itself cannot affect mixing. In fact, any local effects of rotation on mixing are due to interactions between dispersion, rotation and strain, and enter the formulas through the commutator of rotation and strain, due to the symmetric nature of $\boldsymbol{\sigma}$. Because they arise from the interaction between three processes, their effect is of higher order.

Recall, our purpose is to construct a metric for calculating the dilution index, determined by the plume's sampling of gradients in the underlying velocity field, while taking into account that the deformation tensor varies throughout the flow field. To this end, consider for a moment the canonical shear flow with velocity $v_x = \alpha y$, for which

$$s_{ij} = \frac{1}{2}\begin{pmatrix} \alpha & 0 & 0 \\ 0 & \alpha & 0 \\ 0 & 0 & 0 \end{pmatrix}, \qquad \xi_{ij} = \frac{1}{2}\begin{pmatrix} 0 & \alpha & 0 \\ -\alpha & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{12}$$

For this setup, the dilution index at all times can be shown to be (Bolster et al. 2011a)

$$E(t) = (4\pi eDt)^{d/2}\sqrt{1+\frac{(\alpha t)^2}{12}}. \tag{13}$$

We see that the dilution index for this simple flow has the same structure as (11) and they are identical if we define an effective shear rate corresponding to the local strain s:

$$\alpha = \sqrt{2\,\mathrm{Tr}(\boldsymbol{s}^2)}. \tag{14}$$

Thus we argue that the $\mathrm{Tr}(\boldsymbol{s}^2)$ provides a local measure of how efficient a given point is at mixing due to local velocity gradients.

4 Ansatz: Interpreting a Heterogeneous Flow as an Effective Shear

While we have provided an additional metric to quantify the mixing intensity at a point in a heterogeneous flow that is applicable to any incompressible flow, we noted in "Introduction" that several such metrics already exist and one has to ask what the benefit of one more might be. Ideally we want to be able to use metrics like this to build effective upscaled models that can predict mixing over scales larger than over which this local expansion and early-time approximation holds.

With this in mind, we propose the following *ansatz*: Compute the dilution index of a plume in a heterogeneous flow by representing it as an effective, time-variable shear flow as sampled by the plume and defined based on (14). It is critical to note here that this is not a formal derivation or proof of an effective model. Generalizing Bolster et al. (2011a), the Greens function for the ADE with a time-variable shear flow with constant isotropic dispersion can be solved exactly via a combination of Fourier transforms and the method of characteristics, leading to dilution index:

$$E(t) = (4\pi eDt)^{d/2}\sqrt{1+A(t)(2B(t)-A(t))+C(t)-B^2(t)}, \tag{15}$$

where:

$$A(t) = \int_0^t \alpha(u)\,\mathrm{d}u, \quad B(t) = \frac{1}{t}\int_0^t u\alpha(u)\,\mathrm{d}u, \quad C(t) = \frac{1}{t}\int_0^t A^2(u)\,\mathrm{d}u. \tag{16}$$

The dimensionless functions A, B and C encode the time history of the plume's sampling of shear effects through different integrals that involve the effective shear rate α. For α we will take an effective shear rate defined according to Eq. (14). Since our effective shear rate varies throughout a spatially extended plume in a heterogeneous background flow, we must also provide an averaging procedure. We consider two variants, the value at the plume center of mass and a plume average. Considering the plume in terms of Lagrangian particles, with $\boldsymbol{x}^{(i)}$ the position of the ith particle, and $\boldsymbol{x}_{cm}$ the position of the plume center of mass, these two definitions for the effective shear rate lead to:

$$\alpha_{cm}(t) = \sqrt{2\,\mathrm{Tr}(\boldsymbol{s}^2\,|_{\boldsymbol{x}_{cm}(t)})} \quad \alpha_{av}(t) = \sum_i \sqrt{2\,\mathrm{Tr}(\boldsymbol{s}^2\,|_{\boldsymbol{x}^{(i)}(t)})}. \tag{17}$$

5 Numerical Simulations

To test the performance of our *ansatz*, we simulate transport through heterogeneous velocity fields, using standard random walk particle tracking methods (Risken 1989). We present two-dimensional simulations, but the model formulation holds in three dimensions. We follow a standard approach of solving Darcy flow through random realizations of lognormally distributed, multi-Gaussian-correlated hydraulic conductivity fields, often used as idealizations to study flows in hetereogeneous aquifers (e.g., Dagan and Neuman 2005; Gelhar, LW 1993). Flow was solved with a finite volume formulation. We set the correlation length to $l = 1$ and considered several variances, $\sigma^2_{\log \boldsymbol{K}} = 0.1, 0.5, 1, 2, 4$, for the lognormal conductivity field, representing different degrees of heterogeneity. A sample conductivity and velocity field are presented in Fig. 1. Flow was driven by a head drop along the x direction. The resulting velocity field was normalized to unit spatial average along the horizontal direction, $\overline{v_x} = 1$.

For transport, the dispersion coefficient was chosen so as to consider two values of Péclet number, $Pe = \overline{v_x} l/D = 10^2, 10^3$. The initial condition was a point source. In all cases, 5 million particles were used. Simulations were run up to a time $t_{\max} = 10^2 l/\overline{v_x}$, so as to focus on early, non-ergodic dynamics. To obtain the dilution index directly from the particle tracking simulations, we reconstruct the concentration field using a Gaussian kernel smoothing method (Morariu et al. 2008; Fernàndez-Garcia and Sanchez-Vila 2011). Spatial variables are presented normalized by the correlation length, and temporal variables are normalized by $l/\overline{v_x}$.

Figure 2 shows a typical sample time series of the two effective shear rates as determined from our simulations. As expected, the plume average approach smooths out the more drastic fluctuations observed for the center of mass approach. These are then used to compute (15) for comparison with the actually measured dilution indices. Figure 3 shows the directly measured dilution index along with the predictions of our *ansatz* for each of our Péclet number and variance combinations, discussed in further detail below. Interestingly, the averaging character of the quantities A, B and C in Eq. (15) lead to similar results for the dilution index estimated based on α_{av} and α_{cm}. This means that knowledge of either the center of

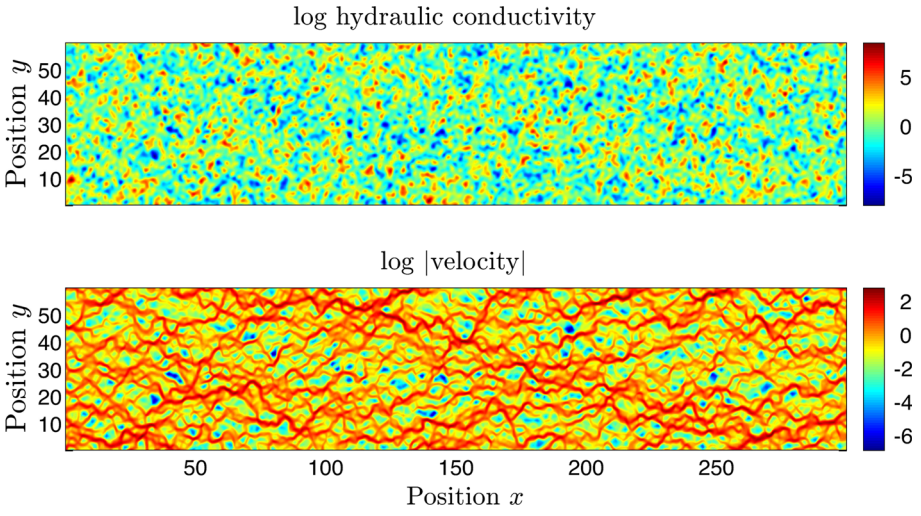

Fig. 1 Example lognormal, Gaussian-correlated hydraulic conductivity field with variance $\sigma^2_{\log \boldsymbol{K}} = 4$ and associated Darcy flow field

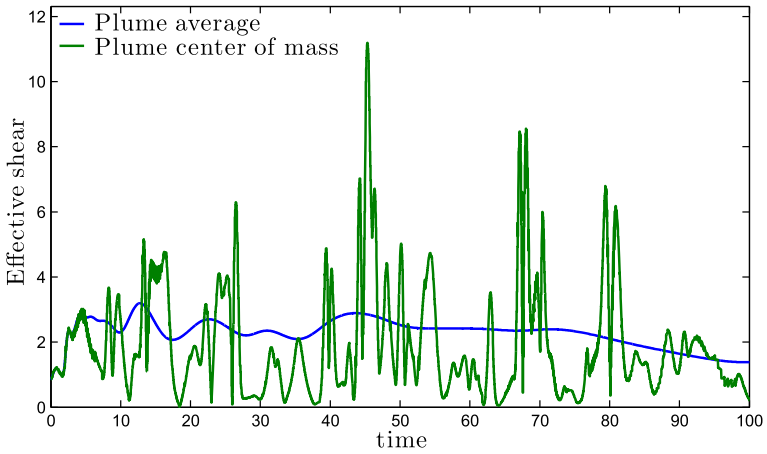

Fig. 2 Typical time series of effective shear, using the plume average and center of mass approaches [see Eq. (17)]. Results shown correspond to a transport realization with $Pe = 10^3$ and a hydraulic conductivity field with variance $\sigma^2_{\log \boldsymbol{K}} = 4$

mass trajectory or plume-averaged properties is sufficient to provide a reasonable basis for the application of our *ansatz*.

In the presence of weak flow heterogeneity ($\sigma^2_{\log \boldsymbol{K}} \lesssim 1$), we found that our *ansatz* significantly overpredicted the observed dilution index at late simulation times, but its performance appears to improve with heterogeneity strength. Results were qualitatively similar for the two simulated values of Péclet number (10^2 and 10^3), with our method generally providing slightly better estimates for the higher value. This is in stark contrast to standard perturbation approaches, which rely on the assumption of weak heterogeneity (e.g., de Barros et al. 2015). At present, we lack a satisfying explanation for the breakdown of our *ansatz* at low heterogeneities and future efforts will focus on establishing formal conditions for its applicability, in order to better understand the origins of its limitations. Nonetheless, it appears to provide a promising approach for strong heterogeneity, which typically poses the greatest challenge for traditional theories. Unlike perturbation solutions our approach is not bottom up; it does not clearly link predictions with properties of the measured conductivity field and relies on the time series produced by numerical simulations.

6 Discussion and Conclusions

Departing from an ADE formulation, we derived a formal local, short-time equivalence between the effects on mixing of an arbitrary incompressible, steady flow and a shear flow with an effective shear rate determined by the trace of the local strain tensor squared. Based on this equivalence, we proposed an *ansatz* to quantify the dilution index, a proxy for mixing, of a point source solute plume in a heterogeneous velocity field. In stark contrast to more classical perturbation-based approaches, our ansatz appears to work well for systems with higher degrees of heterogeneity than for those with weak heterogeneity. While we do not have a robust explanation or set of criteria for this, we find promise in the approach, as highly heterogeneous systems tend to be more difficult to model.

Although we have considered homogeneous and isotropic dispersion for theoretical simplicity and in order to highlight the role of the velocity field, the formalism presented here may

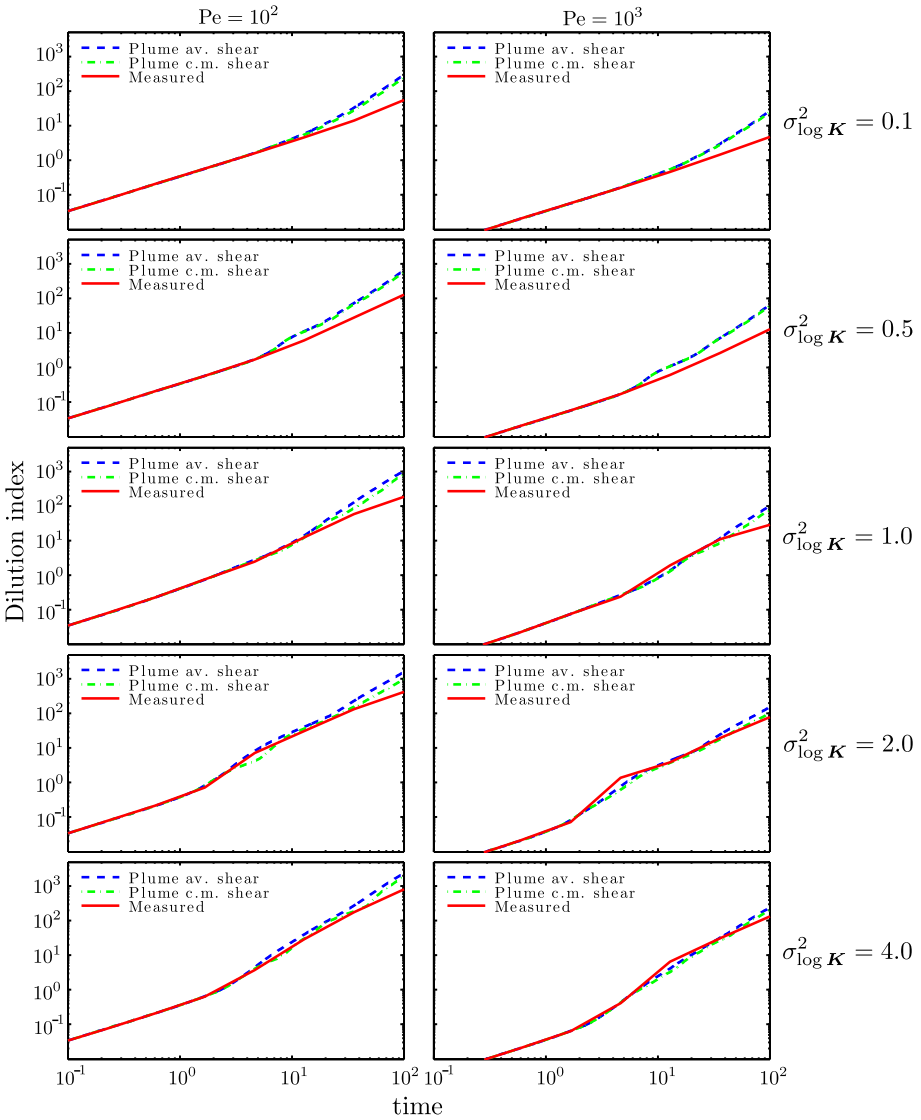

Fig. 3 Evolution of the dilution index as measured directly from simulations and predicted by the two variants of our effective shear approach. *Each column* corresponds to a Péclet number and *each line* to a hydraulic conductivity variance

be generalized to accommodate more complex dispersion tensors. The numerical results were presented for two-dimensional Darcy flow through lognormal, Gaussian-correlated conductivity fields, but the theoretical framework is in principle applicable to three dimensions and arbitrary flow fields. It also suggests an approach to predicting dilution based on theoretical considerations about the structure of the underlying flow field, which is of particular importance given the high degree of uncertainty often associated with real site characterization. Examining the performance of our approach under these generalizations will be the subject of future work. Additionally, the work we presented here focuses on a point source initial

condition and one must be cautious in generalizing any conclusions to other initial and/or boundary conditions given the nonlinear nature of mixing, e.g., for steady-state plumes (Liedl et al. 2005; Cirpka and Valocchi 2007; Knutson et al. 2007; Bolster et al. 2007; Cirpka et al. 2012; Hochstetler et al. 2013) .

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Appendix: Full Solution for Dilution Index with Arbitrary Constant Deformation

For an arbitrary deformation tensor for which strain and rotation do not necessarily commute, we will calculate the exponentials of $\boldsymbol{\varepsilon}$ and $\boldsymbol{\varepsilon}^T$ via a result that follows from the Cayley–Hamilton theorem. Because the exponential function is analytic in the whole complex plane, we can write:

$$\begin{aligned} e^{\boldsymbol{\varepsilon} u} &= a_\mu(u)\boldsymbol{\varepsilon}^\mu, \\ e^{\lambda_i u} &= a_\mu(u)\lambda_i^\mu, \end{aligned} \tag{18}$$

where μ runs over 0 to $d-1$ (and summation over repeated indices is implied), and the λ_i, $i = 1, \ldots, d$, are the eigenvalues of $\boldsymbol{\varepsilon}$. Since $\boldsymbol{\varepsilon}$ and $\boldsymbol{\varepsilon}^T$ have the same eigenvalues, the same expressions hold if $\boldsymbol{\varepsilon}$ is replaced by $\boldsymbol{\varepsilon}^T$. The time-dependent a_μ can be determined by solving the linear system encoded in the second line, and they are found to be:

$$a_\mu(u) = b_{\mu j}\, e^{\lambda_j u}, \tag{19}$$

where:

$$b_{\mu j} = \frac{1}{\prod_{i\neq j}(\lambda_j - \lambda_i)} \times \begin{cases} 1, & \mu = d-1 \\ -\sum_{i\neq j}\lambda_i, & \mu = d-2 \\ \prod_{i\neq j}\lambda_i, & \mu = d-3 \end{cases}. \tag{20}$$

These formulas are valid in 2- and 3-d (a_{-1} in $2d$ should be ignored). Then:

$$\begin{aligned} \boldsymbol{\sigma}(t) &= c_{ij}(t) b_{\mu i} b_{\nu j} \boldsymbol{\varepsilon}^\mu \boldsymbol{\varepsilon}^{T\nu}, \\ c_{ij}(t) &= \frac{e^{(\lambda_i+\lambda_j)t} - 1}{\lambda_i + \lambda_j}. \end{aligned} \tag{21}$$

Note there is no sum implied in the definition of the c_{ij}. Also, these formulas are still valid if a pair of eigenvalues sums to zero, in which case they are to be understood in the limit. For such a pair, $c_{ij} = t$.

We will now focus on the two-dimensional case. The three-dimensional case can be tackled using similar techniques, but the algebra is substantially more complicated, and fully characterizing the system requires knowledge of the eigenvalues of strain, the absolute value of the vorticity and also the orientation of the vorticity vector, for example with respect to

the principal axes of strain. The final result, while it can be explicitly written, yields little physical insight and/or utility. However, for two dimensions we have for the eigenvalues of strain, rotation and deformation $\lambda_{s,i} = \pm\lambda_s$, $\lambda_s \in \mathbb{R}$, $\lambda_{\xi,i} = \pm\lambda_\xi$, $\lambda_\xi \in \imath\mathbb{R}$, and $\lambda_i = \pm\lambda$, $\lambda = \sqrt{|\lambda_s|^2 - |\lambda_\xi|^2} \in \mathbb{R} \cup \imath\mathbb{R}$, which are all easily seen to be true for 2-d incompressible flow ($\imath$ is the imaginary unit). We can now use the results above to find:

$$\boldsymbol{\sigma}(t) = \frac{2D}{4\lambda}\left[[\sin\, h(2\lambda t) + 2\lambda t]\mathbb{1} + [\cos\, h(2\lambda t) - 1]\frac{\boldsymbol{\varepsilon} + \boldsymbol{\varepsilon}^T}{\lambda} + [\sin\, h(2\lambda t) - 2\lambda t]\frac{\boldsymbol{\varepsilon}\boldsymbol{\varepsilon}^T}{\lambda^2}\right]. \tag{22}$$

Using $\boldsymbol{\varepsilon} = \boldsymbol{s} + \boldsymbol{\xi}$ we find:

$$\begin{aligned}\boldsymbol{\sigma}(t) = \frac{2D}{4\lambda}\Bigg[&[\sin\, h(2\lambda t) + 2\lambda t]\mathbb{1} + [\cos\, h(2\lambda t) - 1]\frac{2\boldsymbol{s}}{\lambda} \\ &+ [\sin\, h(2\lambda t) - 2\lambda t]\frac{\boldsymbol{s}^2 - \boldsymbol{\xi}^2 + [\boldsymbol{\xi}, \boldsymbol{s}]}{\lambda^2}\Bigg].\end{aligned} \tag{23}$$

Now from the results for commuting strain and rotation we expect that if $[\boldsymbol{\xi}, \boldsymbol{s}] = 0$, rotation should play no part, so the $\boldsymbol{\xi}^2$ term is troublesome. However, the application of the Cayley–Hamilton theorem above means that there is a relation between $\boldsymbol{\varepsilon}$ and $\boldsymbol{\varepsilon}^T$ and their corresponding powers. Specifically, using Eq. (18) for $\boldsymbol{\varepsilon}$ and $\boldsymbol{\varepsilon}^T$ and looking at the coefficients of t^2 we find the relation:

$$\boldsymbol{\xi}^2 = \lambda^2\mathbb{1} - \boldsymbol{s}^2. \tag{24}$$

Using this relation, we can rewrite $\boldsymbol{\sigma}$ as:

$$\boldsymbol{\sigma}(t) = \frac{2D}{\lambda}\left\{\lambda t\mathbb{1} + \frac{\cos\, h(2\lambda t) - 1}{2\lambda}s + \frac{\sin\, h(2\lambda t) - 2\lambda t}{4\lambda^2}(2\boldsymbol{s}^2 + [\boldsymbol{\xi}, \boldsymbol{s}])\right\}. \tag{25}$$

In this form, we can clearly identify the effect on spreading of pure diffusion, and the coupling between diffusion and strain, and diffusion, strain and rotation.

Let us now consider the (direct-oriented) basis where $\boldsymbol{s}$ diagonalizes. This basis always exists, because $\boldsymbol{s}$ is symmetric. Note that if $\boldsymbol{s} = 0$ it diagonalizes in any basis and our expressions will still hold. We will use hatted indices (e.g., $\hat{i}$) to denote components in this basis. The matrices s and s^2 diagonalize trivially in this basis, but one can also show without too much effort that:

$$[\boldsymbol{\xi}, \boldsymbol{s}]_{\hat{i}\hat{j}} = -\lambda_s Re(\lambda_\xi)\begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix}, \tag{26}$$

where Re denotes the real part. Incidentally, this shows that in 2-d we have $[\boldsymbol{\xi}, \boldsymbol{s}] = 0$ iff there is no rotation or no strain. We thus find:

$$\begin{aligned}\sigma_{\hat{i}\hat{j}}(t) = &\frac{2D}{\lambda} \\ &\times\begin{pmatrix}\lambda t + \frac{\cos\, h(2\lambda t)-1}{2\lambda}\lambda_s + \frac{\sin\, h(2\lambda t)-2\lambda t}{4\lambda^2}2\lambda_s^2 & -\frac{\sin\, h(2\lambda t)-2\lambda t}{4\lambda^2}2\lambda_s Re(\lambda_\xi) \\ -\frac{\sin\, h(2\lambda t)-2\lambda t}{4\lambda^2}2\lambda_s Re(\lambda_\xi) & \lambda t - \frac{\cos\, h(2\lambda t)-1}{2\lambda}\lambda_s + \frac{\sin\, h(2\lambda t)-2\lambda t}{4\lambda^2}2\lambda_s^2\end{pmatrix}.\end{aligned} \tag{27}$$

We can now find the determinant, which is frame independent as long as direct orientation is kept, directly using this basis. Some manipulation gives:

$$\det \boldsymbol{\sigma}(t) = 4D^2 \left[t^2 + \lambda_s^2 \frac{\sin\, h^2(\lambda t) - \lambda^2 t^2}{\lambda^4} \right], \tag{28}$$

and thus the dilution index is:

$$E(t) = 4\pi e D \sqrt{t^2 + \lambda_s^2 \frac{\sin\, h^2(\lambda t) - \lambda^2 t^2}{\lambda^4}}. \tag{29}$$

This expression recovers the solutions for pure rotation and pure strain found under the assumption of commuting strain and rotation. Note also that $\sin\, h^2(x) - x^2$ is positive for all $x \in \mathbb{R} \cup \imath\mathbb{R}$, which implies that strain always has an enhancing effect on mixing.

The case $\lambda = 0$, or equivalently $|\lambda_s| = |\lambda_{\boldsymbol{\xi}}|$, represents shear flow and must be understood as the limit of the formulas above. It leads to:

$$\boldsymbol{\sigma}(t) = 2Dt \left\{ \mathbb{1} + t\boldsymbol{s} + \frac{t^2}{3}(2\boldsymbol{s}^2 + [\boldsymbol{\xi}, \boldsymbol{s}]) \right\}, \tag{30}$$

$$\sigma_{\hat{i}\hat{j}}(t) = 2Dt \begin{pmatrix} 1 + \lambda_s t + \frac{2}{3}\lambda_s^2 t^2 & -\frac{2}{3}\lambda_s Re(\lambda_{\boldsymbol{\xi}}) t^2 \\ -\frac{2}{3}\lambda_s Re(\lambda_{\boldsymbol{\xi}}) t^2 & 1 - \lambda_s t + \frac{2}{3}\lambda_s^2 t^2 \end{pmatrix}, \tag{31}$$

$$E(t) = 4\pi e D t \sqrt{1 + \frac{1}{3}\lambda_s^2 t^2}. \tag{32}$$

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