Solute dispersion for stable density-driven flow in randomly heterogeneous porous media

Aronne Dell'Oca¹, Monica Riva^{1,2}, Jesus Carrera³, Alberto Guadagnini^{1,2}

¹Dipartimento di Ingegneria Civile e Ambientale, Politecnico di Milano, Piazza L. Da Vinci 32,
20133 Milano, Italy

²Department of Hydrology and Atmospheric Sciences, University of Arizona, Tucson, AZ 85721,
USA

³ GHS UPC-CSIC, IDAEA, CSIC, Barcelona, 08034, Spain

Correspondence to: aronne.delloca@polimi.it

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18 Abstract

19 We present a theoretical investigation on the processes underpinning the reduced longitudinal
20 spreading documented in stable variable density flows, as opposed to constant density settings,
21 within heterogeneous porous media. We do so by decomposing velocity and pressure in terms of
22 stationary and dynamic components. The former corresponds to the solution of the constant density
23 flow problem, while the latter accounts for the effects induced by density variability. We focus on a
24 stable flow configuration and analyze the longitudinal spread of saltwater injected from the bottom
25 of a column formed by a heterogeneous porous medium initially fully saturated by freshwater. We
26 adopt a perturbation expansion approach and derive the equations satisfied by section-averaged
27 concentrations and their ensemble mean values. These formulations are respectively characterized
28 by a *single realization* and an *ensemble* dispersive flux, which we **determine** through appropriate
29 closure equations. The latter are solved via semi-analytical and numerical approaches. Our
30 formulations and associated results enable us to discriminate the relative impact on the density-
31 driven solute displacement of (a) covariance of the permeability of the porous medium, (b) cross-
32 covariance between permeability and concentration, which is in turn linked to the coupling of flow
33 and transport problems, and (c) cross-covariance between the dynamic and stationary velocities.

34

1. Introduction

Proper understanding and quantification of the feedback between space-time variability of fluid density and the ensuing flow and transport settings is relevant for a variety of environmental and industrial problems. These include, e.g., coastal aquifer management (e.g., [1, 18, 29, 36, 51, 62, 51]), enhanced oil recovery strategies (e.g., [21, 27, 56]), design and engineering of safe CO₂ storage protocols (e.g., [19, 40, 48, 65]) as well as quantification of solute transport in fractured media for site remediation and/or groundwater source protection (e.g., [8, 53, 57]). In this framework, the effect of density contrasts between miscible fluids has been shown to influence the spreading of contaminants along directions parallel (e.g., [13], [24], [39], [66]) and normal to mean flow velocity (e.g., [2, 22, 38, 46, 45, 47, 55, 61, 64]). A notable feature is that settings associated with variable density are characterized by reduced spreading (when compared to constant density) for stable configurations (where the light fluid lies above the dense fluid), and by enhanced spreading for unstable configurations.

Reduced spreading is typically attributed to the occurrence of stabilizing effects. A local increase in velocity causes a perturbation in the concentration front that does not affect velocities in constant density settings. However, the ensuing density increase tends to drag the fluid, resulting in a reduction of the velocity peak and of the overall spreading of concentrations. The results of this stabilizing effect have been documented in nearly homogenous (e.g., [28, 30, 59]) as well as in heterogeneous (e.g., [9, 34, 35, 37]) porous media.

The complexity of the pore scale geometry and flow patterns, which lead to spatial variability of permeability at the continuum scale (e.g., [6, 63]), ultimately governs the variability of solute concentration fields typically observed in **natural porous formations** (e.g., [50, 56]). A common practice to address this complexity **in constant density scenarios** relies on decomposing the velocity and concentration fields in terms of the sum of a mean value and a zero-mean fluctuation (i.e., a classical perturbation approach). This decomposition is non-unique and can be performed by considering spatial (e.g., [3, 15, 49, 63]), temporal (e.g., [6, 17]) or ensemble (e.g., [12, 41, 42, 43,

44]) averaging techniques. Regardless the nature of this decomposition, a key research goal is the formulation and solution of an effective model satisfied by a representative (mean/average) concentration. A dispersive flux, given by the average of the cross product between velocity and concentration fluctuations, typically arises in such effective models.

Here, we focus on the interaction between the effects of (a) buoyancy and (b) heterogeneity induced by the spatial variability of permeability on density-dependent flow and transport behavior. Our key aim is to provide a physically-based quantitative analysis of the above documented reduction of the width of the dispersion zone in stable flow configurations. The essence of the matter is illustrated in Figure 1, obtained using the procedures described in Section 2. Saltwater is continuously injected within a porous domain initially saturated with freshwater. The permeability of the porous medium is modelled as a random function of space characterized by a multivariate log-Gaussian distribution. A realization of permeability is displayed in Figure 1a. Figures 1b, 1c, and 1d display a snapshot of solute concentration, for three diverse values of density contrast, as quantified through the gravity number, N_g , which reflects the relative importance of buoyancy and viscous forces (see Section 2.1 for additional details). These plots evidence two major elements: (a) the progressive reduction of the width of the dispersion zone and (b) the tendency of the concentration profile to resemble a configuration typical of homogenous media for increasing values of N_g . It is then natural to ask: (a) Can we detect the basic physical mechanisms at the heart of this behavior? (b) What is the feedback between the effects of buoyancy and physical heterogeneity of the domain in the concentration distribution? and (c) Can we quantify the relative importance of buoyancy and permeability heterogeneity on the pattern of the observed spreading process? These are precisely the key questions we address in this work.

Previous studies have addressed the above questions. Hassanizadeh and Leijnse [28] proposed an extension of Fick's law to model the local dispersive flux, i.e., the dispersive flux appearing in the upscaled (from pore- to continuum-scale) transport equation. These authors modeled the local dispersive flux as a nonlinear function of the local concentration gradient through the introduction

87 of a coefficient of proportionality (termed β by them). This model has been shown to accurately
 88 reproduce a set of laboratory experiments in homogenous media under a stable configuration [59],
 89 as well as breakthrough curves obtained from a suite of two-dimensional numerical simulations in
 90 heterogeneous media, [31]. However, a clear and unambiguous link between β and the underlying
 91 physical processes is still missing. Watson et al. [59] and Landman et al. [32] observed that β
 92 depends on the mean flow rate. Landman et al. [31] documented a notable dependence of β on the
 93 fluid density contrast in heterogeneous formation and introduced two upscaling models (from pore
 94 to continuum and from continuum to continuum scale) based on homogenization theory. Both
 95 models render a good reproduction of the numerical results of Landman et al. [32] and provide a
 96 relatively simple nonlinear relationship between the dispersive flux and the gradient of section
 97 averaged concentrations. Welty and Gelhar [60] introduced an analytical expression for the
 98 asymptotic (long-time) longitudinal macro-dispersivity under the assumption that the velocity field
 99 is a second-order stationary random process. These authors show that macro-dispersion is a function
 100 of the gradient of the ensemble mean concentration, flow rate, displacement distance, gravity, fluid
 101 density and viscosity and log-permeability correlation scale and variance. However, as we highlight
 102 in Section 3, the velocity field **is** not (statistically) stationary in a variable density problem, due to
 103 the coupling between flow and transport equations. In this framework, the objectives of this work
 104 are (i) to obtain an upscaled equation for flow and transport in a variable density context, (ii) to
 105 **highlight** the factors that control the dispersive flux, and (iii) to investigate the features of the
 106 reduced solute dispersion that is observed under stable displacement conditions.

107 **2. Flow and transport Model and problem set up**

108 Fluid flow in porous media is described by conservation of mass and Darcy's law, in the form

$$109 \quad \frac{\partial \phi \rho^*}{\partial t^*} + \nabla^* \cdot (\rho^* \mathbf{q}^*) = 0, \quad \mathbf{q}^* = -\frac{\mathbf{k}^*}{\mu^*} \cdot (\nabla^* p^* + \rho^* \mathbf{g}^* \nabla^* z^*), \quad (1)$$

110 where ρ^* [ML⁻³], μ^* [M T⁻¹ L⁻¹] and p^* [M T⁻² L⁻¹] are fluid density, viscosity, and pressure,

111 respectively, g^* [LT^{-2}] is gravity, z^* [L] is elevation, t^* [T] is time, $\mathbf{q}^*$ [L T^{-1}] is Darcy's flux, $\mathbf{k}^*$
 112 [L^2] is the permeability tensor and ϕ [-] is porosity, which we treat as a constant scalar.

113 We consider solute transport to be locally governed by the advection-dispersion equation

$$114 \quad \frac{\partial(\phi \rho^* C^*)}{\partial t^*} + \nabla^* \cdot (\rho^* \mathbf{q}^* C^*) - \nabla^* \cdot (\phi \rho^* \mathbf{D}^* \cdot \nabla^* C^*) = 0. \quad (2)$$

115 Here C^* [-] is solute concentration, and $\mathbf{D}^*$ [L T^{-2}] is the local dispersion tensor, modeled as

$$116 \quad \mathbf{D}^* = (D_m^* + \alpha_T^* |\mathbf{v}^*|) \mathbf{I} + (\alpha_L^* - \alpha_T^*) \frac{\mathbf{v}^* \mathbf{v}^*}{|\mathbf{v}^*|}, \quad (3)$$

117 where D_m^* is molecular diffusion, α_T^* and α_L^* [L] respectively are transverse and longitudinal
 118 dispersivities, $\mathbf{I}$ is the identity matrix, and $\mathbf{v}^*$ [L T^{-1}] is fluid velocity defined as

$$119 \quad \mathbf{v}^* = \mathbf{q}^* / \phi. \quad (4)$$

120 We focus on a stable density-dependent problem within a heterogeneous porous domain, Ω , of
 121 width W^* [L], and height H^* [L], as depicted in Figure 1. In our set-up Darcy's flux $\mathbf{q}^*$ has
 122 components q_z^* and q_y^* , respectively along the vertical, z^* , and horizontal, y^* , direction. The
 123 column is initially filled by freshwater with density ρ_f^* . Seawater with density ρ_s^* at concentration
 124 C_s is continuously injected at time $t^* \geq 0$ at the bottom of the column, i.e., at $z^* = 0$. The remaining
 125 boundary conditions are: (1) no flow at $y^* = 0, W^*$; (2) constant pressure $p^* = p_{BC}^*$ at $z^* = 0$; (3)
 126 prescribed vertical flux, $q_z^* = q_{BC}^*$, and solute mass flux $(\mathbf{q}^* C^* - \mathbf{D}^* \cdot \nabla^* C^*) \cdot \mathbf{n}^* = q_{BC}^* C^*$ at $z^* = H^*$,
 127 with $\mathbf{n}^*$ being a unit vector pointing outwards (see Figure 1).

128 Closure of the system (1)-(4) is obtained by assuming viscosity to be constant and expressing

129 ρ^* as a linear function of C^* (e.g., [1])

$$130 \quad \rho^* = \rho_f^* + \beta^* C^*, \quad \text{with } \beta^* = (\rho_s^* - \rho_f^*) / C_s. \quad (5)$$

131 In the following we assume isotropic dispersivity values, $\alpha_T^* = \alpha_L^* = \alpha^*$ and disregard D_m^* since
 132 its contribution to the dispersion tensor can be usually neglected (e.g., [31]). Therefore, (3) reduces
 133 to

$$134 \quad \mathbf{D}^* = \alpha^* |\mathbf{v}^*| \mathbf{I}. \quad (6)$$

135 We adopt $\sqrt{\tilde{k}^*}$, $\tilde{k}^*$ being the geometric mean of the permeability field, as characteristic length
 136 scale. This leads to the introduction of the following dimensionless quantities

$$137 \quad y = \frac{y^*}{\sqrt{\tilde{k}^*}}, \quad z = \frac{z^*}{\sqrt{\tilde{k}^*}}, \quad \mathbf{k} = \frac{\mathbf{k}^*}{\tilde{k}^*}, \quad \mathbf{q} = \frac{\mathbf{q}^*}{v_{BC}^*}, \quad \mathbf{v} = \frac{\mathbf{v}^*}{v_{BC}^*}, \quad \nabla = \sqrt{\tilde{k}^*} \nabla^*, \quad (7)$$

$$\rho = \frac{\rho^*}{\Delta \rho^*}, \quad p = \frac{p^*}{\mu^* v_{BC}^* / \sqrt{\tilde{k}^*}}, \quad t = \frac{t^*}{\sqrt{\tilde{k}^*} / v_{BC}^*}, \quad C = \frac{C^*}{C_s},$$

138 with $\Delta \rho^* = \rho_s^* - \rho_f^*$, $v_{BC}^* = q_{BC}^* / \phi$. Using (7), Eqs. (1), (2), (5) and (6) can be written in
 139 dimensionless form as

$$140 \quad \frac{\partial \phi \rho}{\partial t} + \nabla \cdot (\rho \mathbf{q}) = 0, \quad \mathbf{q} = -\mathbf{k} \cdot (\nabla p + N_g \rho \nabla z), \quad \text{with } N_g = \frac{\Delta \rho^* \tilde{k}^* g^*}{\mu^* v_{BC}^*}, \quad (8)$$

$$141 \quad \frac{\partial (\rho \phi C)}{\partial t} + \nabla \cdot (\rho \mathbf{q} C) - \nabla \cdot (\rho \phi \mathbf{D} \cdot \nabla C) = 0, \quad (9)$$

$$142 \quad \rho = \rho_f + C \quad \text{with } \rho_f = \frac{\rho_f^*}{\Delta \rho^*}, \quad (10)$$

$$143 \quad \mathbf{D} = \frac{|\mathbf{v}|}{\text{Pe}} \mathbf{I} \quad \text{with } \text{Pe} = \frac{\sqrt{\tilde{k}^*}}{\alpha^*}. \quad (11)$$

144 Here, Pe is the Peclet number, expressing the ratio between advective and dispersive transport rate,
 145 and N_g is the gravity number, expressing the relative intensity of buoyancy and viscosity effects.

146 We solve (8)-(11) by means of the widely tested code SUTRA [58]. Domain heterogeneity is
 147 modeled by treating the dimensionless permeability $\mathbf{k}$ as an isotropic random field
 148 $\mathbf{k} = \exp(Y(y, z)) \mathbf{I}$, where $Y(y, z)$ is a zero-mean second order stationary random process of space
 149 characterized by an isotropic exponential covariance function

$$C_{YY}(\mathbf{r}^*) = \sigma_Y^2 \exp(-|\mathbf{r}^*|/l^*), \quad (12)$$

$\mathbf{r}^*$ [L], σ_Y^2 [-] and l^* [L] respectively being the separation vector (or lag) between two spatial locations, variance and correlation length (or **integral scale**) of Y . The heterogeneous Y fields are synthetically generated by the widely used and tested code SGSIM [20] on a uniform grid structured with five generation nodes per correlation length, l^* as detailed in Section 4.

3. Theoretical framework

We start by deriving the equation satisfied by section-averaged concentrations in the setting of Figure 1, which naturally leads to an effective one-dimensional model. This equation includes a term, called *single realization* dispersive flux, which enables us to embed in a simple one-dimensional (along the mean flow direction) mathematical formulation the effect of the spatial heterogeneity of $\mathbf{k}$. We then leverage on the above one-dimensional (section-averaged) model and average it in probability space. The ensuing mathematical formulation includes an *ensemble* dispersive flux that enables us to encapsulate the effect of the uncertainty in the spatial arrangement of $\mathbf{k}$ on the mean concentration distribution. **The main purpose of the analytical formulations presented in Sections 3.1-3.2 is to enrich our understanding of the interplay between permeability and stabilizing effects that is at the origin of the solute spread reduction taking place in stable flows within heterogeneous media. Our formulations facilitate grasping the impact of variations of N_g and σ_Y^2 on the system behavior in terms of flow structure and resulting solute spreading. Our study enables us to link the key moments of the flow and log-permeability fields to the solute spreading behavior.**

3.1 Section-Averaged Concentration

Point concentration values $C(y, z, t)$ are seldom available from laboratory or field experiments [7], section-averaged concentration profiles being typically monitored (e.g., [11, 23, 26, 31, 39,

52]). Therefore, we focus on cross-section (or horizontal) average concentration, $\bar{C}(z,t)$, where the overbar indicates the horizontal averaging operator defined as

$$\bar{\Theta} = \frac{1}{W} \int_0^W \Theta(y) dy, \quad (13)$$

where Θ denotes a generic quantity (a parameter or a state variable). In the following, we derive the equation satisfied by $\bar{C}(z,t)$ relying on the Boussinesq approximation (e.g., [18], [21]), i.e.,

assuming that $\nabla \cdot \mathbf{q} \gg \frac{\phi}{\rho} \frac{\partial \rho}{\partial t} + \mathbf{q} \cdot \frac{\nabla \rho}{\rho}$. Under this assumption (8) and (9) can be simplified as

$$\nabla \cdot \mathbf{v} = 0, \quad \mathbf{q} = -\mathbf{k} \cdot (\nabla p + N_g \rho \nabla z), \quad (14)$$

$$\frac{\partial C}{\partial t} + \mathbf{v} \cdot \nabla C - \nabla \cdot (\mathbf{D} \cdot \nabla C) = 0. \quad (15)$$

Note that $\mathbf{k}$ is spatially variable and all remaining quantities (i.e., $\mathbf{v}$, $\mathbf{q}$, p , ρ , C and $\mathbf{D}$) are both spatially and temporally variable. According to (14)-(15), steady state conditions never occur in the variable density problem considered in this work and sketched in Figure 1 because flow evolves in time due to the effects of the buoyancy term in (14). Therefore, (14) must be solved jointly with (10) and (15).

We decompose each variable in (14)-(15) as the sum of a horizontal spatial mean and a (spatial) zero-mean fluctuation (along the y -direction), i.e.,

$$\begin{aligned} p &= \bar{p} + p', & C &= \bar{C} + C', & \mathbf{k} &= \mathbf{I} + k' \mathbf{I}, \\ \rho &= \bar{\rho} + \rho', & \mathbf{v} &= \bar{\mathbf{v}} + \mathbf{v}', & \mathbf{D} &= \bar{\mathbf{D}} + \mathbf{D}', \end{aligned} \quad (16)$$

where horizontally-averaged quantities depend on (z, t) , while fluctuations depend on (y, z, t) .

According to (10), one can see that

$$\bar{\rho} = \rho_f + \bar{C}, \quad \rho' = C'. \quad (17)$$

Horizontally averaged concentrations, $\bar{C}(z,t)$, satisfy the following equation (see A.4 in Appendix

A)

$$\frac{\partial \bar{C}}{\partial t} + \frac{\partial \bar{C}}{\partial z} - \frac{1}{\text{Pe}} \frac{\partial^2 \bar{C}}{\partial z^2} + \frac{\partial \overline{v_z' C'}}{\partial z} = 0, \quad (18)$$

where $\overline{v_z' C'}$ is the *single realization* dispersive flux. At **second order**, the latter reads (see A.11a)

$$\overline{v_z' C'} = - \int_0^t \int_0^H \overline{v_z'(z,t) v_z'(\xi,\tau)} G_z^T(z,t,\xi,\tau) \frac{\partial \bar{C}(\xi,\tau)}{\partial \xi} d\xi d\tau. \quad (19)$$

Here, $G_z^T(z,t,\xi,\tau)$ is the deterministic Green's function introduced in (A.10) and $\overline{v_z'(z,t) v_z'(\xi,\tau)}$ (defined in (A.11b)) is the horizontal average of $v_z'(y,z,t) v_z'(\eta=y,\xi,\tau)$, i.e., of the product between vertical velocity fluctuations evaluated at the same horizontal coordinate and diverse vertical positions and times. The single realization dispersive flux, $\overline{v_z' C'}$, embeds in the upscaled one-dimensional (vertical) model (18) the effect of permeability fluctuations on the section-averaged concentration $\bar{C}(z,t)$ (e.g., [16]).

To highlight the effect of density variations on $C(y,z,t)$, we decompose the vector of velocity fluctuations as the sum of (a) a *stationary* component, $\mathbf{v}^{st}'(z,y)$, which corresponds to the solution of (14) with constant density ($\rho = \rho_f$), and (b) a *dynamic* component, $\mathbf{v}^{dy}'(z,y,t)$, accounting for the stabilizing buoyancy effects due to density variations, i.e.,

$$\mathbf{v}'(y,z,t) = \mathbf{v}^{st}'(y,z) + \mathbf{v}^{dy}'(y,z,t). \quad (20)$$

The equations satisfied by $\mathbf{v}^{st}'(y,z)$ and $\mathbf{v}^{dy}'(y,z,t)$ are derived in Appendix C and read (see (C.5))

$$v_z^{st}'(y,z) = Y(y,z) - \frac{1}{\phi} \frac{\partial p^{st}'(y,z)}{\partial z}, \quad v_z^{dy}'(y,z,t) = -\frac{1}{\phi} \left(\frac{\partial p^{dy}'(y,z,t)}{\partial z} + N_g \rho'(y,z,t) \right), \quad (21)$$

Here, similar to (20), we decompose pressure fluctuation $p'(y,z,t)$ as the sum of a *stationary*, $p^{st}'(y,z)$, and a *dynamic*, $p^{dy}'(y,z,t)$, component. A numerical analysis of (21) (details not

213 shown) reveals that, for the set up of Figure 1, $Y(y,z) \gg \partial p^{st}/\partial z / \phi$ and

214 $N_g \rho'(y,z,t) \gg \partial p^{dy}/\partial z$. Therefore, (21) can be approximated as

$$215 \quad v_z^{st}/\phi \approx Y(y,z) \quad , \quad v_z^{dy}/\phi \approx -\frac{N_g}{\phi} \rho'(y,z,t) \quad . \quad (22)$$

216 While (22) shows a clear dependence of v_z^{st}/ϕ on the fluctuation $Y(y,z)$ of the permeability

217 field, the behavior of v_z^{dy}/ϕ depends on that of $\rho'(y,z,t)$ through the multiplying factor

218 N_g/ϕ . Noticing that according to (17) $\rho'(y,z,t) = C'(y,z,t)$, and making use of (20) and (A.8), the

219 second of (22) can be rewritten as

$$220 \quad v_z^{dy}/\phi \approx -\frac{N_g}{\phi} C'(y,z,t) = \frac{N_g}{\phi} \int_0^t \int_{\Omega} \left[Y(\eta,\xi) - \frac{N_g}{\phi} \rho'(\eta,\xi,\tau) \right] \frac{\partial \bar{C}(\xi,\tau)}{\partial \xi} \widehat{G}^T(y,z,t;\xi,\eta,\tau) d\eta d\xi d\tau$$

221 (23)

222 Equation (23) allows recognizing that v_z^{dy}/ϕ is a non-local quantity embedding flow and

223 transport features. Two effects are encapsulated in (23): (i) permeability fluctuations are seen to

224 promote velocity fluctuations; (ii) the action of the stabilizing buoyant term contributes to reducing

225 velocity fluctuations, with an intensity proportional to N_g and to the density fluctuations. These two

226 competing mechanisms are weighted by the mean concentration (or density) gradient, the behavior

227 of which is a function of both N_g and permeability heterogeneity.

228 In the following, we briefly elucidate the benefit of decomposition (20) by focusing on a

229 simple heterogeneous domain formed by a single semi-circular inclusion of high permeability

230 $\mathbf{k}(y,z) = 10\mathbf{I}$ (depicted in red in Figures 2a-2c), within an otherwise uniform porous medium with

231 $\mathbf{k}(y,z) = \mathbf{I}$. The remaining relevant key dimensionless quantities are set to $N_g = 0.35$, $Pe = 0.25$,

232 $\phi = 0.35$, $\rho_f = 40$. The analysis for randomly heterogeneous porous media is offered in Section 4.

233 Figures 2a, 2b and 2c respectively depict v_z^{st}/ϕ , v_z^{dy}/ϕ and $\mathbf{v}(y,z,t)$ at time $t = 664$. The

234 total velocity field $\mathbf{v}(y,z,t)$ is shown in Figure 2d together with the concentration field. As an

235 additional term of comparison, Figure 2e depicts $v(y, z)$ and $C(y, z, t)$ computed at the same
 236 dimensionless time considering a solute with $\rho = \rho_f$ (i.e., corresponding to the transport of a
 237 passive scalar in a constant density setting). Figures 2d and 2e clearly show that solute spreading
 238 decreases when density effects are considered. This observed reduction of solute dispersion for the
 239 variable density stable scenario is strictly linked to the ensuing velocity distribution. Figure 2a
 240 indicates that vertical *stationary* flow fluctuations, v_z^{st} , are positive in the high permeability zone,
 241 favoring solute advancement within this area. This generates positive density fluctuations, ρ' ,
 242 which in turn trigger negative vertical *dynamic* flow fluctuations, v_z^{dy} , due to the effect of
 243 (reduction in) buoyancy (see Figure 2b). The opposite occurs in the portion of the domain adjacent
 244 to the high permeability inclusion. Since v_z^{st} and v_z^{dy} are associated with opposite signs, the total
 245 vertical flow fluctuation v_z' is smaller than its counterpart evaluated for the constant density
 246 scenario (compare Figure 2c and Figure 2a). Note that the stabilizing effect of v_z^{dy} tends to
 247 decrease the intensity of v_z' (with respect to the uniform density case) without altering its sign.
 248 This behavior is also observed in the heterogeneous field analyzed in Section 4 for the investigated
 249 values of N_g . Ultimately, the velocity field at the solute front is more uniform in the variable density
 250 than in the constant density case (compare the velocity fields in Figures 2d and 2e) causing a
 251 decreased solute dispersion in the former scenario, as compared against the latter. This observation
 252 is further supported by Figures 2f and 2g where the distribution along z of the variance of the
 253 vertical velocity, $\overline{\sigma_{v_z}^2}(z, t)$, and of $\overline{v_z' C'}$ (19) for the constant density case (black curves) are
 254 compared against their counterpart associated with variable density (red curves). One can clearly
 255 note the reduction of the magnitude of both $\overline{\sigma_{v_z}^2}(z, t)$ and $\overline{v_z' C'}$ under variable density conditions.
 256 The basic mechanisms highlighted here for this relatively simple heterogeneous configuration are at

the heart of the observed reduction in solute spreading observed for stable variable density flow within heterogeneous porous media, as further discussed in Section 4.

3.2 Ensemble Analysis

Here we treat $\mathbf{k}$ as a second order stationary random field and derive the equation satisfied by the ensemble mean of the section-averaged concentration, i.e., $\langle \bar{C}(z,t) \rangle$, $\langle \rangle$ indicating the ensemble mean operator. This analysis enables us to link the main features of $\langle \bar{C}(z,t) \rangle$ to key statistics of the permeability field. The equation satisfied by $\langle \bar{C}(z,t) \rangle$ results from ensemble averaging of (18), i.e.,

$$\frac{\partial \langle \bar{C} \rangle}{\partial t} + \frac{\partial \langle \bar{C} \rangle}{\partial z} - \frac{1}{\text{Pe}} \frac{\partial^2 \langle \bar{C} \rangle}{\partial z^2} + \frac{\partial \langle v_z' C' \rangle}{\partial z} = 0, \quad (24)$$

where $\langle \overline{v_z' C'} \rangle$ is the *ensemble* dispersive flux defined, at second order and after localization with respect to the mean concentration vertical gradient, as (see Appendix B, eq. (B.3))

$$\langle \overline{v_z' C'} \rangle = - \int_0^t \int_0^H \langle \overline{v_z'(z,t) v_z'(\xi,\tau)} \rangle G_z^T(z,t,\xi,\tau) d\xi d\tau \frac{\partial \langle \bar{C}(z,t) \rangle}{\partial z}. \quad (25)$$

Here $\langle \overline{v_z'(z,t) v_z'(\xi,\tau)} \rangle$ is the horizontal spatial mean of $\langle v_z'(y,z,t) v_z'(\eta=y,\xi,\tau) \rangle$ which is the covariance between the vertical velocity fluctuation at z at time t and its counterpart evaluated at ξ at time τ , both at the same horizontal coordinate y . The *ensemble* dispersive flux takes into account the effect of the spatial heterogeneity of $\mathbf{k}$ across the *ensemble* of realizations on the evaluation of the *ensemble* mean of the section-averaged concentration. As discussed by Morales-Casique et al. [41], [42], the *ensemble* dispersive flux is related to the effects of the loss of information associated with the random nature of $\mathbf{k}$, and therefore of the velocity field, on the prediction of C through $\langle \bar{C} \rangle$.

Equation (25) highlights the key role of the velocity field statistics in controlling the ensemble dispersive flux. We show in Appendix D that the velocity covariance, $\langle \overline{v_z'(z,t) v_z'(\xi,\tau)} \rangle$, can be

decomposed as the sum of four terms, involving stationary and dynamic velocity fluctuations, as
(see (D.12))

$$\begin{aligned} \langle v'_z(z, t) v'_z(\xi, \tau) \rangle &= \langle v_z^{st}(z) v_z^{st}(\xi) \rangle + \langle v_z^{st}(z) v_z^{dy}(\xi, \tau) \rangle + \langle v_z^{dy}(z, t) v_z^{st}(\xi) \rangle + \langle v_z^{dy}(z, t) v_z^{dy}(\xi, \tau) \rangle \\ &= \mathbf{A} \left\{ \overline{Y(z)Y(\xi)} \right\} + \frac{N_g}{\phi} \mathbf{B} \left\{ \overline{Y(z)C'(\xi, \tau)} \right\} + \frac{N_g}{\phi} \mathbf{C} \left\{ \overline{C'(z, t)Y(\xi)} \right\} + \frac{N_g^2}{\phi^2} \mathbf{D} \left\{ \overline{C'(z, t)C'(\xi, \tau)} \right\} \end{aligned} \quad (26)$$

where $\mathbf{A}$, $\mathbf{B}$, $\mathbf{C}$, $\mathbf{D}$ are operators defined (D.8)-(D.11). Equation (26) indicates that while the horizontal spatial mean of the *stationary-stationary* term, $\langle v_z^{st}(z) v_z^{st}(\xi) \rangle$, depends on the covariance of the log-permeability field, the *stationary-dynamic*, $\langle v_z^{st}(z) v_z^{dy}(\xi, \tau) \rangle$, and *dynamic-stationary*, $\langle v_z^{dy}(z, t) v_z^{st}(\xi) \rangle$, terms depend on the cross-covariance $\langle Y(z)C'(\xi, \tau) \rangle$ between log permeability and concentration and are characterized by a quasi-linear dependence on N_g , since $\langle Y(z)C'(\xi, \tau) \rangle$ in (26) depends on N_g as dictated by (E.5). Otherwise, the *dynamic-dynamic* component, $\langle v_z^{dy}(z, t) v_z^{dy}(\xi, \tau) \rangle$, is a function of the covariance of the concentration field, $\langle C'(z, t)C'(\xi, \tau) \rangle$, and exhibits a quasi-linear dependence on N_g^2 (note that also $\langle C'(z, t)C'(\xi, \tau) \rangle$ depends on N_g , as shown in (F.6)). The main features of covariances embedded in (26) and their impact on the solute transport behavior are analyzed and discussed in Section 4. Note that the constant flow boundary condition and the adoption of the Boussinesq approximation in our set-up ensure that the center of mass of the advancing solute front travels at the same speed for all realizations of Y , thus yielding no artificial spreading (see e.g., [16]).

4. Results and discussion

Numerical Monte Carlo (MC) based and semi-analytical results are presented here for the quantities introduced in Section 3, i.e., velocity covariance, cross-covariance between concentration and permeability, ensemble dispersive flux and concentration variance. Numerical and semi-analytical

299 results are depicted as solid and dashed curves, respectively, in the plots of this Section. We recall
 300 for convenience the main assumptions invoked in our semi-analytical formulations presented in
 301 Section 3, i.e.,: (i) all statistical moments are approximated at second order (in terms of the
 302 fluctuation Y of the permeability field), thus nominally limiting our semi-analytical solution to
 303 mildly heterogeneous media, $\sigma_Y^2 \ll 1$; (ii) flow and transport Green's functions are approximated
 304 by their one-dimensional formulations (A.10) and (D.4b). Additionally, to speed up the evaluation
 305 of our semi-analytical expressions and to make use of already existing analytical results, (iii) cross-
 306 covariance between log conductivity and hydraulic head in (E.5) is computed via an analytical
 307 solution available for mean uniform steady-state flow in an infinite two-dimensional domain [14];
 308 and (iv) the contribution of $\langle v_z^{st}(z)v_z^{st}(\xi) \rangle$ in (25)-(26) is set equal to $\sigma_Y^2 l$, which corresponds to
 309 the asymptotic value for macro dispersion derived in Gelhar and Axness [25]. Due to the coupled
 310 nature of the problem (e.g., $\langle v_z C \rangle$ depends on $\partial \langle C \rangle / \partial z$, as shown by (24)), a semi-analytical
 311 solution for the quantities presented in Section 3 requires an iterative procedure and therefore a
 312 CPU time which is comparable with the one associated with MC simulations. Hence, in the
 313 following we take advantage of the MC results to evaluate the mean concentration gradient,
 314 $\partial \langle C \rangle / \partial z$.

315 In our simulations we set $\phi = 0.35$, $\tilde{k}^* = 10^{-9} \text{ m}^2$, $H^* = 0.3 \text{ m}$, $W^* = 0.05 \text{ m}$, $\alpha^* = 1.25 \times 10^{-4} \text{ m}$,
 316 $\rho_f^* = 1000 \text{ kg/m}^3$, $\rho_s^* = 1025 \text{ kg/m}^3$, $l^* = 2.50 \times 10^{-3} \text{ m}$ (i.e., $H^*/W^* = 6$, $H = H^*/\sqrt{\tilde{k}^*} \approx 9486.8$,
 317 $W = W^*/\sqrt{\tilde{k}^*} \approx 1581.1$, $l = l^*/\sqrt{\tilde{k}^*} = 79$, $H^*/l^* = 120$, $W^*/l^* = 20$, $\text{Pe} = 0.25$, $l^*/\alpha^* = 20$,
 318 $\rho_f = 40$). The size of the domain has been selected to ensure that we frame our calculations on a
 319 large system, in terms of the correlation scale, l^* , of Y . The domain is discretized by a regular mesh
 320 of square elements. We performed a set of preliminary simulations aimed at testing the influence of
 321 space and time grid discretization on the quantities of interest (namely, horizontal mean

concentration, *single realization* and *ensemble* dispersive flux and velocity covariance). Accurate results at an affordable CPU time have been obtained with a spatial grid formed by 100×600 elements (i.e., $\Delta z^* = \Delta y^* = 5 \times 10^{-4}$ m) and dimensionless time step $\Delta t = 2.8$. Five rows of grid elements formed by a homogeneous porous medium with $k^* = \tilde{k}^*$ are added at the inlet boundary to regularize solute inflow. This procedure is commonly adopted in experimental set-ups with highly heterogeneous porous media (e.g., Kretz et al., 2003) to regularize the inflow conditions. We investigate four scenarios characterized by diverse levels of heterogeneity of the permeability field, i.e., $\sigma_Y^2 = 0.1, 0.5$, combined with diverse intensities of buoyancy effects, as quantified by $N_g = 0, 0.1, 1.0$. Our numerical results are grounded on 1000 MC simulations for each parameter set investigated.

4.1 Variance and Covariance of Vertical Velocity

The covariance of the vertical velocity components (26) plays a critical role in determining the mean solute transport behavior, as highlighted by (24)-(25). Here, we compare the results obtained for the constant and variable density scenarios to elucidate and quantify the origin of the reduced solute spreading documented in Figure 1.

Figures 3 depicts the velocity variance, $\langle \overline{v_z'^2} \rangle$, and the (negative) mean concentration gradient, $-\partial \langle \overline{C} \rangle / \partial z$, at three dimensionless times (i.e., $t_1 = 2.2 \times 10^3$, $t_2 = 4.4 \times 10^3$, and $t_3 = 6.6 \times 10^3$) for $N_g = 1$ (left) and $N_g = 0.1$ (right) and for $\sigma_Y^2 = 0.1$ (two top rows) and $\sigma_Y^2 = 0.5$ (two bottom rows). For illustration purposes, Figure 3 includes only the Monte Carlo based numerical results. The corresponding semi-analytical results are in good agreement with these. Velocity variance $\langle \overline{v_z'^2} \rangle$ varies with z . It is zero at the boundaries (recall that we maintain a constant permeability at the five extreme rows of elements), but immediately rises to its constant density stationary value, except at the region near the solute front, where it is reduced. This reduction reflects the role of the

345 space-time variability of fluid density in stabilizing the system, thus reducing $\langle \overline{v_z'^2} \rangle$ at the solute
346 front as N_g increases. The occurrence of local minima of $\langle \overline{v_z'^2} \rangle$ and $\partial \langle \overline{C} \rangle / \partial z$ is strikingly evident
347 for $N_g = 1$ and small heterogeneity ($\sigma_Y^2 = 0.1$). Decreasing the intensity of buoyancy effects (i.e.,
348 for $N_g = 0.1$) and/or increasing σ_Y^2 cause the local minima of $\langle \overline{v_z'^2} \rangle$ to be dampened and local
349 maxima of $-\partial \langle \overline{C} \rangle / \partial z$ to decrease. We remark that there is strong feedback between the behavior
350 of $\langle \overline{v_z'^2} \rangle$ and $\partial \langle \overline{C} \rangle / \partial z$. An increase of $\langle \overline{v_z'^2} \rangle$ and the corresponding increase of the vertical
351 velocity fluctuation $v_z'(y, z, t)$ is associated with an increase of the solute spreading. These are in
352 turn linked to a decrease of the mean concentration gradient, $\partial \langle \overline{C} \rangle / \partial z$, i.e., to a smoothing of mean
353 concentration profiles.

354 We have shown in (26) that the vertical velocity covariance, $\langle \overline{v_z'(z, t) v_z'(\xi, \tau)} \rangle$, can be
355 decomposed as the sum of four components. These are shown in Figure 4 for $\sigma_Y^2 = 0.1$, and $N_g = 1$
356 (left) and $N_g = 0.1$ (right). Both the overall covariance $\langle \overline{v_z'(z, t) v_z'(\xi, \tau)} \rangle$ (red curve Figure 4a) and
357 its stationary component $\langle \overline{v_z^{st}(z) v_z^{st}(\xi)} \rangle$ (black curve) attain their maximum value at the front (the
358 curves are plotted versus ξ for fixed $t = \tau = t_2$ and $z = z_2 = 4.27 \times 10^3$). Obviously, this maximum is
359 the variance, which was shown (Figure 3(a) and (b)) to display a minimum at the front for the
360 variable density case. The reduction in variance is now manifest (Figures 4(a) and (b)) in the
361 observation that the peak displayed by $\langle \overline{v_z'(z, t) v_z'(\xi, \tau)} \rangle$ at $\xi = z$ is smaller than that of
362 $\langle \overline{v_z^{st}(z) v_z^{st}(\xi)} \rangle$. A perhaps more interesting effect is the asymmetry introduced by variable density
363 on the velocity covariance, which is symmetric and positive for constant density. In fact,

364 $\langle \overline{v_z'(z,t)v_z'(\xi,\tau)} \rangle$ and $\langle \overline{v_z^{st}'(z)v_z^{st}'(\xi)} \rangle$ tend to coincide for $\xi > z$; but $\langle \overline{v_z'(z,t)v_z'(\xi,\tau)} \rangle$ becomes
 365 **significantly** smaller, even negative, than $\langle \overline{v_z^{st}'(z)v_z^{st}'(\xi)} \rangle$ for $\xi \leq z$. As expected, $\langle \overline{v_z'(z,t)v_z'(\xi,\tau)} \rangle$
 366 and $\langle \overline{v_z^{st}'(z)v_z^{st}'(\xi)} \rangle$ **tend** to coincide when N_g decrease (see Figure 4b).

367 **Negative values of the overall covariance** $\langle \overline{v_z'(z,t)v_z'(\xi,\tau)} \rangle$ **for** $\xi \leq z$ **are due to the effect of the**
 368 **dynamic and stationary velocity cross-covariances,** $\langle \overline{v_z^{dy}'(z,t)v_z^{st}'(\xi)} \rangle$ **and** $\langle \overline{v_z^{st}'(z)v_z^{dy}'(\xi,\tau)} \rangle$, **which**
 369 **are always negative.** This result is consistent with (23) and with our discussion in Section 3 arguing
 370 **that positive values of** $v_z^{st}'(\xi)$ **favor the solute to advance within the domain, thus originating**
 371 **negative dynamic velocity fluctuations,** $v_z^{dy}'(z \geq \xi, t)$, **through positive density fluctuations,**
 372 $\rho'(z \geq \xi, t)$. **The same holds when** $v_z^{st}'(\xi)$ **is negative, which promotes positive values of**
 373 $v_z^{dy}'(z \geq \xi, t)$. **We note that** $\langle \overline{v_z^{dy}'(z,t)v_z^{st}'(\xi)} \rangle$ **attains its largest (absolute) value for** $\xi < z$ **and**
 374 $\langle \overline{v_z^{st}'(z)v_z^{dy}'(\xi,\tau)} \rangle$ **for** $\xi > z$. These findings suggest that the stabilizing *dynamic* fluctuation
 375 $\langle \overline{v_z^{dy}'(z,t)v_z^{st}'(\xi)} \rangle$ arising at a given space-time location (z, t) displays a strong **negative correlation**
 376 **with the stationary velocity fluctuations (i.e., with variations of Y according to (22)), especially**
 377 **those occurring at points** $\xi < z$. **This is a result of (i) the coupling between flow and transport (as**
 378 **documented by the dependency of** v_z^{dy}' **on** ρ' **in (22) and (23)), and (ii) the nature of the transport**
 379 **setting according to which density fluctuations are mainly dictated by the velocity perturbations**
 380 **which have been already experienced. This aspect is clearly highlighted by our semi-analytical**
 381 **solution (23) since** the Green's function (A.10) of the transport problem is non zero mainly in the
 382 range of space-time coordinates $(\xi \leq z; \tau \leq t)$. As a consequence, fluctuations v_z^{dy}' , which are
 383 rooted in the coupled nature of flow and transport, are less correlated with the corresponding

384 stationary flow perturbation, v_z^{st} , which occur at forward (in space and time along the mean flow
385 direction) locations. The term $\left\langle \overline{v_z^{dy}(z,t)v_z^{st}(\xi)} \right\rangle$ depicted in Figure 4c-4d imbues information about
386 the extent of the region within which the dynamic velocity field at a given location (in space-time)
387 is influenced by variation of the stationary velocity field at all locations within the domain. At the
388 same time, $\left\langle \overline{v_z^{st}(z)v_z^{dy}(\xi,\tau)} \right\rangle$ depicted in Figures 4e-4f can be seen as an indication of the extent in
389 space and time of the region within which stationary velocity fluctuations promote dynamic
390 perturbations of the flow, an effect which is mainly felt at spatial locations $\xi > z$. In other words,
391 while Figures 4c-4d highlight the way density (or concentration) perturbations at a given location
392 are affected by the heterogeneity spanning the entire domain, Figures 4e-4f highlight the way a
393 local perturbation of the log permeability field contributes to the concentration variability across the
394 full domain. Note also that a change in N_g induces a quasi-linear variation of the magnitude of
395 $\left\langle \overline{v_z^{st}(z)v_z^{dy}(\xi,\tau)} \right\rangle$ and $\left\langle \overline{v_z^{dy}(z,t)v_z^{st}(\xi)} \right\rangle$, as also indicated by (26).

396 Figures 4g and 4h depict $\left\langle \overline{v_z^{dy}(z,t)v_z^{dy}(\xi,\tau)} \right\rangle$ versus ξ for $z = z_2$, $t = t_2$ and three values of τ
397 when $N_g = 1$ and 0.1, respectively. This quantity represents the covariance between the dynamic
398 velocity components, which is positive, as expected. It can then be noted that $\left\langle \overline{v_z^{dy}(z,t)v_z^{dy}(\xi,\tau)} \right\rangle$
399 exhibits a supra-linear dependence on N_g , with a power which is different from two (compare
400 Figures 4g and 4h) according to (26) and (F.6). Quantities $\left\langle \overline{v_z^{dy}(z,t)v_z^{st}(\xi)} \right\rangle$, $\left\langle \overline{v_z^{st}(z)v_z^{dy}(\xi,\tau)} \right\rangle$
401 and $\left\langle \overline{v_z^{dy}(z,t)v_z^{dy}(\xi,\tau)} \right\rangle$ display a behavior which is qualitatively similar to what illustrated above
402 at all space-time locations investigated (details not shown).

403 Figure 5 depicts results analogous to those of Figure 4 for $\sigma_Y^2 = 0.5$. For both Y variances, the
404 semi-analytical solution captures remarkably the behavior of the Monte Carlo results. As mentioned

405 earlier, $\left\langle \overline{v_z^{st}(z)v_z^{st}(\xi)} \right\rangle$ is linearly proportional to σ_Y^2 . Increasing the heterogeneity causes
 406 $\left\langle \overline{v_z(z,t)v_z(\xi,\tau)} \right\rangle$ to increase. What is more interesting, we find that, for a given N_g , the
 407 dampening effect of the buoyancy term on velocity fluctuations tends to decrease as σ_Y^2 **increases**,
 408 i.e., $\left\langle \overline{v_z(z,t)v_z(\xi,\tau)} \right\rangle$ is closer to $\left\langle \overline{v_z^{st}(z)v_z^{st}(\xi)} \right\rangle$ in Figure 5a than in Figure 4a. This behavior is
 409 essentially due to the diminished capability of the negative cross-covariances $\left\langle \overline{v_z^{dy}(z,t)v_z^{st}(\xi)} \right\rangle$ and
 410 $\left\langle \overline{v_z^{st}(z)v_z^{dy}(\xi,\tau)} \right\rangle$ to stabilize the flow, **i.e., the absolute value of $\left\langle \overline{v_z^{dy}(z,t)v_z^{st}(\xi)} \right\rangle$ and**
 411 **$\left\langle \overline{v_z^{st}(z)v_z^{dy}(\xi,\tau)} \right\rangle$ increases sub-linearly with σ_Y^2 .** To **further** clarify this aspect, we derive an
 412 analytical expression for $\left\langle \overline{v_z^{st}(z)v_z^{dy}(z,t)} \right\rangle$ based on the following set of rough approximations of
 413 (23): (i) the buoyancy term is neglected, and (ii) $\partial \overline{C}(\xi,\tau)/\partial \xi$ is approximated through a Gaussian
 414 function whose degree of spreading for the tracer case is governed by the macro-dispersion
 415 coefficient, D^{mt} , (Gelhar and Axness, [25]). Multiplying (23) by $v_z^{st}(z)$ (as in (22)), taking spatial
 416 and ensemble expectation **and making use of (12)** leads to (after some manipulations):

$$\begin{aligned}
 \left\langle \overline{v_z^{st}(z)v_z^{dy}(z,t)} \right\rangle &\approx \frac{N_g}{\phi} \int_0^t \int_{\Omega} \frac{C_Y}{\sqrt{4\pi D^{mt} \tau}} \exp\left(-\frac{(\xi-\tau)^2}{4D^{mt} \tau}\right) G_z^T(z,t;\xi,\tau) d\xi d\tau \\
 &\approx \frac{N_g}{\phi} \int_0^t \int_{\Omega} \frac{\sigma_Y}{\sqrt{4\pi l \tau}} \exp\left(-\frac{(\xi-\tau)^2}{l \sigma_Y^2 \tau}\right) \exp\left(-\frac{|z-\xi|}{l}\right) G_z^T(z,t;\xi,\tau) d\xi d\tau
 \end{aligned} \tag{27}$$

418 which highlights a sublinear grow of the cross-covariance of vertical velocity with σ_Y^2 , for a given
 419 N_g . Recalling (25) and (26), the sublinear dependence on σ_Y^2 of the cross-covariance between
 420 *dynamic* and *stationary* components, when viewed in the context of the linear scaling on σ_Y^2 of the
 421 *stationary* velocity covariance, suggests that the effect of stabilization of $\left\langle \overline{v_z^{dy}(z,t)v_z^{st}(\xi)} \right\rangle$ and

422 $\left\langle \overline{v_z^{st}(z)v_z^{dy}(\xi,\tau)} \right\rangle$ on the flow field, and therefore on the solute front, decreases as the
 423 heterogeneity of the porous medium increases. We emphasize that the purpose of (27) is not to
 424 provide an exact solution of $\left\langle \overline{v_z^{st}(z)v_z^{dy}(\xi,\tau)} \right\rangle$ but to help in clarifying the effects of σ_Y^2 on the
 425 *dynamic* and *stationary* velocity cross-covariance.

426 4.2 Cross Covariance between Concentration and Permeability

427 Figure 6 depicts the **Monte Carlo based** section-averaged cross-covariance between permeability
 428 and concentration, $\left\langle \overline{Y(z)C'(\xi,\tau)} \right\rangle$, versus ξ when $z=z_2$ (black curves) and versus z when
 429 $\xi=\xi_2=z_2$ (red curves) at time $\tau=t_2=4.4\times 10^3$ for $\sigma_Y^2=0.1$ when $N_g=1$ (Figure 6a) or 0.1
 430 (Figure 6b). Corresponding results for $\sigma_Y^2=0.5$ and $N_g=1$ or 0.1 are illustrated in Figures 6c and
 431 6d. **Corresponding semi-analytical results are in good agreement (details not shown for clarity).**
 432 Results obtained for $\left\langle \overline{Y(z=z_2)C'(\xi,\tau)} \right\rangle$ (blues curves) and $\left\langle \overline{Y(z)C'(\xi=\xi_2,\tau)} \right\rangle$ (green curves)
 433 when density is constant are also included.

434 The analysis of $\left\langle \overline{Y(z)C'(\xi,\tau)} \right\rangle$ versus ξ allows highlighting the way permeability fluctuations,
 435 $Y(z)$, are correlated with concentration fluctuation, $C'(\xi,\tau)$, at space-time coordinates (ξ,τ) .
 436 Otherwise, graphs of $\left\langle \overline{Y(z)C'(\xi,\tau)} \right\rangle$ **versus z** reveal how concentration fluctuations $C'(\xi,\tau)$ (at a
 437 given location) are affected by permeability fluctuations at a set of z spatial coordinates. It can be
 438 noted that: (a) peaks of $\left\langle \overline{Y(z)C'(\xi,t)} \right\rangle$ do not occur at the selected reference locations ξ_2 or z_2 ; (b)
 439 values of $\left\langle \overline{Y(z_2)C'(\xi,t)} \right\rangle$ at $\xi > z_2$ are larger than those at $\xi < z_2$, **thus implying that permeability**
 440 **fluctuations** influence $C'(\xi,t)$ at downstream (i.e., when $\xi > z_2$) locations along the direction of the
 441 mean flow more strongly than they do at $\xi < z_2$. Consistent with this mechanism, $C'(\xi_2,t)$ is highly
 442 correlated with $Y(z)$ when $z < \xi_2$ (see red curve in Figure 6). In summary, Figure 6 suggests that

443 (a) permeability at a given location highly influences concentration at downstream positions, and
 444 (b) concentration are highly correlated with permeability at upstream locations. These results also
 445 imply that permeability data are expected to be beneficial (on average) to reduce uncertainty on
 446 solute transport prediction at downstream (along the mean flow direction) locations, concentration
 447 measurements conveying relevant information to infer permeability within upstream zones.

448 One can note that values of $\langle \overline{Y(z)C'(\xi_2, t)} \rangle$ for a given z are larger for the constant density than
 449 in a variable density scenario (compare green and red curves in Figure 6). The mechanism
 450 associated with the reduction of $\langle \overline{Y(z)C'(\xi_2, t)} \rangle$ for the variable density scenario is elucidated by
 451 our semi-analytical solution (E.5). It is linked to the last term on the right hand side of (E.5), which
 452 depends linearly on N_g and vanishes in the uniform density case. From a physical point of view,
 453 the emerging behavior is due to the capability of the stabilizing buoyant effects of smoothing the
 454 perturbation in the solute concentration profiles induced by the spatial variability of permeability.
 455 As a consequence, an increase of buoyancy effects in a heterogeneous domain results in
 456 concentration profiles that are characterized by a regular shape, which is akin to the one observed in
 457 homogeneous media. Values of $\langle \overline{Y(z)C'(\xi, \tau)} \rangle$ in density-driven flows tend to coincide with those
 458 associated with uniform density scenarios for increasing σ_Y^2 and/or decreasing N_g , because the
 459 effect of the stabilizing buoyancy terms decreases.

460 Comparison of Figures 4 and 6 reveals that the shape of $\langle \overline{v_z^{dy}(z, t)v_z^{st}(\xi)} \rangle$ and
 461 $\langle \overline{v_z^{st}(z)v_z^{dy}(\xi, \tau)} \rangle$ is respectively similar to that of $\langle \overline{C'(z, t)Y(\xi)} \rangle$ and $\langle \overline{Y(z)C'(\xi, \tau)} \rangle$. This
 462 numerical result is in agreement with (D.8)-(D.9), according to which
 463 $\langle \overline{v_z^{dy}(z, t)v_z^{st}(\xi)} \rangle \propto -N_g \langle \overline{C'(z, t)Y(\xi)} \rangle / \phi$ and $\langle \overline{v_z^{st}(z)v_z^{dy}(\xi, \tau)} \rangle \propto -N_g \langle \overline{Y(z)C'(\xi, \tau)} \rangle / \phi$. We
 464 remark that our analysis of the cross-covariance between permeability and concentration leads to

conclusions which are consistent with those stemming from our discussion about velocity covariances because for the scenario here investigated the characteristics of Y mainly dictate the behavior of v_z^{st} , C being clearly linked to v_z^{dy} .

4.3 Ensemble Dispersive Flux and Concentration Variance

Here, we analyze the *ensemble* dispersive flux, $\langle \overline{v_z' C'} \rangle$ introduced in (24)-(25). We recall (25)-(26) and remark that $\langle \overline{v_z' C'} \rangle$ depends non-linearly on the mean concentration gradient, $\partial \langle \overline{C} \rangle / \partial z$. This observation is consistent with previous analytical and numerical results (see, e.g., [31, 32, 60]). The root of this non-linear dependency lies in the coupling between the flow and transport problems and in particular is due to the stabilizing buoyant term in the *dynamic* velocity fluctuations, which depends on the mean concentration gradient, **as emphasized by (23)**.

Figures 7a and 7b respectively depict $\langle \overline{v_z' C'} \rangle$ and the concentration variance, $\sigma_c^2 = \langle \overline{C'^2} \rangle$, versus z at three selected times, for $\sigma_Y^2 = 0.1$ and constant density (blue curves), or $N_g = 0.1$ (red curves), and 1 (black curves) computed via Monte Carlo simulations. The semi-analytical solutions of (25), for $\langle \overline{v_z' C'} \rangle$, and (F.6), for σ_c^2 , are also depicted. Corresponding results for $\sigma_Y^2 = 0.5$ are depicted in Figures 7c and 7d. One can note from Figure 7 that our semi-analytical solutions capture **the behavior of** the Monte Carlo numerical results. Values of $\langle \overline{v_z' C'} \rangle$ and σ_c^2 decrease in time and/or for increasing N_g suggesting that $\langle \overline{C(z,t)} \rangle$ can be considered as a good approximation of (random) concentrations $\overline{C}(z,t)$ and $C(y,z,t)$ (a) at late times when the solute front has sampled the overall log-permeability variability and concentration fluctuations along the transverse direction tend to be smoothed out due to mixing, and/or (b) for large values of N_g , because the solute front in heterogeneous domains tends to resemble the pattern documented in homogenous media. An increase of the heterogeneity of the permeability field causes both $\langle \overline{v_z' C'} \rangle$ and σ_c^2 to increase and

the regularizing ability of the stabilizing buoyancy effects to decrease for a given N_g . These findings are in agreement with the experimental results of Landman et al. [32]-[33]. Our semi-analytical solutions overestimate the MC based peaks of the dispersive flux and of the concentration variance. This type of behavior has already been detected in second-order analytical and semi-analytical methods developed for constant density scenarios (see, e.g., [28, 41, 42]) and it has been attributed to the observation that such approaches neglect third order moments (involving velocity and concentration fluctuations) that can play a significant role as aquifer heterogeneity, time and Péclet number increase.

The decrease of $\langle v_z' C' \rangle$ and σ_c^2 with N_g is described by our semi-analytical solution (25)-(26) and (F.1) and is linked to the negative cross-covariances, $\langle v_z^{dy}(z, t) v_z^{st}(\xi) \rangle$ and $\langle v_z^{st}(z) v_z^{dy}(\xi, \tau) \rangle$ analyzed in Section 4.1. Our results embedded in Figures 4 and 5 suggest that the main contribution to the reduction of $\langle v_z' C' \rangle$ and σ_c^2 in a stable variable density setting stems from $\langle v_z^{dy}(z, t) v_z^{st}(\xi) \rangle$. An interpretation of this result can be offered by considering the nature of the problem and noting that the evolution of mean concentration at (z, t) is affected by the perturbations that the velocity field have already experienced, i.e., at times $\tau \leq t$ and locations $\xi \leq z$. These effects are encapsulated in the Green's function $G_z^T(z, t; \xi, \tau)$ in (25) and (F.1). Note that $\langle v_z^{dy}(z, t) v_z^{st}(\xi) \rangle$ in Figures 4c-4d and 5c-5d exhibits long negative tails for $\xi \leq z$. The effect of such tails is opposite to the one given by $\langle v_z^{st}(z) v_z^{st}(\xi, \tau) \rangle$. One can see that the convolution between $G_z^T(z, t; \xi, \tau)$ and $\langle v_z^{dy}(z, t) v_z^{st}(\xi) \rangle$ in (25) yields a net reduction of the memory of concentration perturbation on the experienced velocity field due to the stabilizing effects of the density-driven scenario.

5 Conclusions

509 We analyze the reduction of solute spreading in randomly heterogeneous porous media driven by
 510 density variations in a stable flow setting. A key finding of this study is that the reduction of
 511 spreading and contraction of concentration profiles are due to a spatial reorganization of the
 512 velocity field, as compared to a constant density scenario. We observe that the velocity variance and
 513 the spatial extent of velocity correlation tend to decrease when density effects are considered. This
 514 reduction of variance and correlation scale of velocity causes a corresponding reduction of the
 515 strength of the dispersive flux.

516 We quantify the **reduction of the velocity fluctuations variance** and of solute
 517 spreading/dispersion for density-driven flows by decomposing the flow field fluctuations into a
 518 *stationary* component, v_z^{st} , associated with the solution of the flow problem for a constant density
 519 fluid, and a *dynamic* component, v_z^{dy} , which takes into account the coupling between transport and
 520 flow phenomena introduced by density variations. This decomposition enables us to **highlight** the
 521 interplay between permeability, velocity, and concentration, which is what triggers the reduction of
 522 spreading we observe when comparing constant and variable density settings.

523 The origin of the solute dispersion reduction is linked, both from a mathematical and a physical
 524 point of view, to the emergence of negative cross-covariances between stationary and dynamic
 525 velocity fluctuations, $\left\langle v_z^{dy} v_z^{st} \right\rangle$. These cross-covariances describe the interplay between
 526 permeability-induced *stationary* fluctuations (which promote solute spreading) and stabilizing
 527 buoyancy-induced *dynamic* fluctuations (which hinder solute spreading). We analyze **the way** these
 528 **cross-covariances** vary with the intensity of the ratio between buoyancy and viscous forces (as
 529 expressed by N_g) and with the heterogeneity of the permeability field (as quantified by σ_Y^2). The
 530 absolute values of $\left\langle v_z^{dy} v_z^{st} \right\rangle$ scale approximately linearly with N_g (i.e., with permeability and
 531 density contrast) and sub-linearly with σ_Y^2 . This is in contrast with the well-known linear scaling in

532 σ_Y^2 exhibited by the stationary (i.e., constant density) cross-covariance, $\left\langle \overline{v_z^{st} v_z^{st'}} \right\rangle$. Therefore, the
533 relative importance of the stabilizing components induced by density variations tends to decrease as
534 the heterogeneity of the porous medium increases. The reduction of the strength of the dispersive
535 flux as well as of the concentration variance which is observed for stable density-driven flow (in
536 comparison with the constant density setting) tends to be stronger for increasing values of N_g and
537 decreasing values of σ_Y^2 .

538 Our study allows investigating the factors controlling the dispersive flux, which is typically
539 interpreted as an upscaled parameter to be estimated through model calibration in field scale density
540 driven problems. A key result of our study is that such a parameter does not depend solely on the
541 aquifer properties (such as permeability) but also on flow and transport variables (such as velocity
542 and solute concentration). As such, dispersion coefficients used in a variable density flow and
543 transport models should vary not only in space but also in time to properly represent flow and
544 transport processes. Investigation of processes underpinning the spreading in unstable density-
545 driven flows as well as applications of the developed approaches to estimate the upscaled dispersive
546 flux in field scale scenarios are envisioned to be the focus of future studies.

547

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552

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- 739

740 Appendix A. Section averaged concentration and effective dispersive flux

741 Substituting (16)-(17) in (15), applying the spatial mean operator (13) and making use of the first of
742 (14), leads to the following equation

$$743 \quad \frac{\partial \bar{C}}{\partial t} + \bar{v}_z \frac{\partial \bar{C}}{\partial z} - \frac{\partial}{\partial z} \left(\overline{D_{zz} \frac{\partial \bar{C}}{\partial z}} \right) + \frac{\partial \overline{v_z' C'}}{\partial z} - \frac{\partial}{\partial z} \left(\overline{D_{zz}' \frac{\partial C'}{\partial z}} \right) = 0, \quad (\text{A.1})$$

744 satisfied by the spatially-averaged concentration, $\bar{C}(z, t)$, for the problem set up depicted in Figure
745 1. Here,

$$746 \quad \begin{aligned} \overline{D_{zz}} &= \frac{1}{W} \int_0^W D_{zz}(y, z, t) dy = \frac{1}{WPe} \int_0^W |\mathbf{v}(y, z, t)| dy = \frac{|\bar{\mathbf{v}}(z, t)|}{Pe} \approx \frac{\bar{v}_z}{Pe}, \\ \overline{v_z' C'} &= \frac{1}{W} \int_0^W v_z'(y, z, t) C'(y, z, t) dy, \quad \overline{D_{zz}' \frac{\partial C'}{\partial z}} = \frac{1}{W} \int_0^W D_{zz}'(y, z, t) \frac{\partial}{\partial z} C'(y, z, t) dy. \end{aligned} \quad (\text{A.2})$$

747 Applying (13) to the first of (14) and considering decomposition (16) yields

$$748 \quad \nabla \cdot (\bar{\mathbf{v}}(z, t) + \mathbf{v}'(z, t)) = \frac{\partial \bar{v}_z(z, t)}{\partial z} = 0, \quad \bar{v}_z(z, t) = v_{BC} = 1 \quad (\text{A.3})$$

749 i.e., the spatial mean of the vertical velocity is constant **and coincides with the prescribed vertical**
750 **velocity at $z = H$** . A numerical analysis allows recognizing that it is possible to neglect the last term
751 of (A.1) (details not shown). **Moreover, making use of (A.2) and (A.3), $\overline{D_{zz}}$ can be approximated as**
752 **$1/Pe$. This leads to**

$$753 \quad \frac{\partial \bar{C}}{\partial t} + \frac{\partial \bar{C}}{\partial z} - \frac{1}{Pe} \frac{\partial^2 \bar{C}}{\partial z^2} + \frac{\partial \overline{v_z' C'}}{\partial z} = 0, \quad (\text{A.4})$$

754 where $\overline{v_z' C'}$ is the *single realization* dispersive flux discussed in Section 3.1. Closure of (A.4) is
755 obtained upon writing $\overline{v_z' C'}$ as a function of $\bar{C}$, as detailed in the following. Subtracting (A.4)
756 from (15) and neglecting terms involving the product of fluctuations yields

$$757 \quad \frac{\partial C'}{\partial t} + \frac{\partial}{\partial z} C' - \frac{\nabla^2 C'}{Pe} = -\mathbf{v}' \cdot \nabla \bar{C} + \nabla \cdot (\mathbf{D}' \nabla \bar{C}(z, t)). \quad (\text{A.5})$$

758 Numerical simulations show that $-\mathbf{v}' \cdot \nabla \bar{C} \gg \nabla \cdot (\mathbf{D}' \nabla \bar{C})$ (details not shown) so that (A.5)
759 simplifies as

$$760 \quad \frac{\partial C'}{\partial t} + \frac{\partial}{\partial z} C' - \frac{\nabla^2 C'}{Pe} = -v_z' \frac{\partial \bar{C}}{\partial z}, \quad (\text{A.6})$$

761 subject to the following (homogeneous) boundary and initial conditions

$$\begin{aligned}
 & C'(y, z=0, t) = 0, \quad C'(y, z=H, t) - \frac{1}{\text{Pe}} \frac{\partial C'(y, z, t)}{\partial z} = 0, \\
 & \frac{\partial C'(y=0, z, t)}{\partial y} = \frac{\partial C'(y=W, z, t)}{\partial y} = 0, \quad C'(y, z, t=0) = 0.
 \end{aligned} \tag{A.7}$$

763 Solution of (A.6) can be expressed in terms of $\widehat{G}^T$, i.e., the Green's function that satisfies (A.6)-
 764 (A.7) with the source term replaced by $\delta(\eta-y)\delta(\xi-z)\delta(t-\tau)$, where δ is the Dirac' delta
 765 function, as

$$C'(y, z, t) = - \int_0^t \int_{\Omega} v_z'(y, \xi, \tau) \frac{\partial}{\partial \xi} \overline{C}(\xi, \tau) \widehat{G}^T(y, z, t; \xi, \eta, \tau) d\eta d\xi d\tau. \tag{A.8}$$

767 Multiplying (A.8) by $v_z'(y, z, t)$ and applying (13) leads to the following expression for the *single*
 768 *realization* dispersive flux

$$\overline{v_z'(z, t) C'(z, t)} = - \int_0^t \int_{\Omega} v_z'(z, t) \widehat{G}^T(z, t; \eta, \xi, \tau) v_z'(\eta, \xi, \tau) \frac{\partial \overline{C}(\xi, \tau)}{\partial \xi} d\eta d\xi d\tau, \tag{A.9a}$$

770 with

$$\overline{v_z'(z, t) \widehat{G}^T(z, t; \eta, \xi, \tau)} = \frac{1}{W} \int_0^W v_z'(y, z, t) \widehat{G}^T(y, z, t; \eta, \xi, \tau) dy. \tag{A.9b}$$

772 We further simplify our analysis and follow Bolster et al. [5], [6] by considering a one-dimensional
 773 (1D) problem (along the vertical z -direction) within a semi-infinite domain, $z \in [0, \infty)$. Under this
 774 setting, a closed-form analytical expression for $\widehat{G}^T$ is (see [33])

$$\begin{aligned}
 & \widehat{G}^T(y, z, t; \xi, \eta, \tau) = \delta(y - \eta) G_z^T(z, t; \xi, \tau) \\
 & \text{with } G_z^T(z, t; \xi, \tau) = \sqrt{\frac{\text{Pe}}{4\pi(t-\tau)}} \left[e^{-\text{Pe} \left(\frac{(z-\xi-t+\tau)^2}{4(t-\tau)} \right)} - e^{-\text{Pe} \left(\frac{(\xi+z+t-\tau)^2}{4(t-\tau)} \right)} \right].
 \end{aligned} \tag{A.10}$$

776 Making use of (A.10), (A.9a) simplifies as

$$\overline{v_z'(z, t) C'(z, t)} = - \int_0^t \int_0^H v_z'(z, t) v_z'(\xi, \tau) G_z^T(z, t; \xi, \tau) \frac{\partial \overline{C}(\xi, \tau)}{\partial \xi} d\xi d\tau, \tag{A.11a}$$

778 with

$$\overline{v_z'(z,t)v_z'(\xi,\tau)} = \frac{1}{W} \int_0^W v_z'(y,z,t)v_z'(y,\xi,\tau)dy. \quad (\text{A.11b})$$

Note that $\eta = y$ by virtue of (A.10), so that only the product of velocity fluctuations along the same vertical locations is retained in (A.11a) and (A.11.b).

Appendix B. Ensemble mean of section-averaged concentration and ensemble dispersive flux

Applying the ensemble mean operator to (A.4) yields

$$\frac{\partial \langle \bar{C} \rangle}{\partial t} + \frac{\partial \langle \bar{C} \rangle}{\partial z} - \frac{1}{\text{Pe}} \frac{\partial^2 \langle \bar{C} \rangle}{\partial z^2} = - \frac{\partial \langle v_z' C' \rangle}{\partial z} \quad (\text{B.1})$$

where $\langle v_z' C' \rangle$ is the ensemble dispersive flux discussed in Section 3.2. This term can be evaluated applying the ensemble averaging operator to (A.11a) and neglecting terms involving products of fluctuations of order higher than two, as

$$\langle v_z'(z,t)C'(z,t) \rangle = - \int_0^t \int_0^H \langle v_z'(z,t)v_z'(\xi,\tau) \rangle \frac{\partial \langle \bar{C}(\xi,\tau) \rangle}{\partial \xi} G_z^T(z,t,\xi,\tau) d\xi d\tau. \quad (\text{B.2})$$

Equation (B.2) clearly elucidates that the ensemble dispersive flux is a non-local quantity in time and space, as it depends on velocity cross-covariances and concentration gradients evaluated across the whole domain at all times $\tau \leq t$. A (partial) localization of (B.2) is offered by

$$\langle v_z'(z,t)C'(z,t) \rangle = - \int_0^t \int_0^H \langle v_z'(z,t)v_z'(\xi,\tau) \rangle G_z^T(z,t,\xi,\tau) d\xi d\tau \frac{\partial \langle \bar{C}(z,t) \rangle}{\partial z} \quad (\text{B.3})$$

Appendix C. Velocity and pressure Fluctuations

A first order approximation of velocity fluctuation components $v_y'(y,z,t)$ and $v_z'(y,z,t)$ defined in (16) are derived by subtracting the spatial mean of velocity from the second of (14), approximating the permeability fluctuation as $k'(y,z) = \exp(Y(y,z)) - 1 \approx Y(y,z)$ and disregarding terms involving the product of fluctuations, as

$$v_y'(y,z,t) = - \frac{1}{\phi} \frac{\partial p'(y,z,t)}{\partial y} \quad (\text{C.1})$$

$$v_z'(y,z,t) = Y(y,z) - \frac{1}{\phi} \left(\frac{\partial p'(y,z,t)}{\partial z} + N_g \rho'(y,z,t) \right). \quad (\text{C.2})$$

Note that (20) and (A.3) enable us to write

$$\bar{v}_z(z,t) = 1 = \bar{v}_z^{st}(z) + \bar{v}_z^{dy}(z,t) = \bar{v}_z^{st}, \quad \bar{v}_z^{dy}(z,t) = 0. \quad (\text{C.3})$$

802 Recalling the *stationary* and *dynamic* decomposition introduced in (20) and (C.3) allows rewriting
 803 (C.1)-(C.2) as

$$804 \quad v_y^{st} \prime(y, z) = -\frac{1}{\phi} \frac{\partial p^{st} \prime(y, z)}{\partial y}, \quad v_y^{dy} \prime(y, z, t) = -\frac{1}{\phi} \frac{\partial p^{dy} \prime(y, z, t)}{\partial y}, \quad (C.4)$$

$$805 \quad v_z^{st} \prime(y, z) = Y(y, z) - \frac{1}{\phi} \frac{\partial p^{st} \prime(y, z)}{\partial z}, \quad v_z^{dy} \prime(y, z, t) = -\frac{1}{\phi} \left(\frac{\partial p^{dy} \prime(y, z, t)}{\partial z} + N_g \rho \prime(y, z, t) \right), \quad (C.5)$$

806 where $p^{st} \prime(y, z)$ and $p^{dy} \prime(y, z, t)$ respectively are *stationary* and *dynamic* pressure fluctuations.

807 Using (C.4) and (C.5) and noting that $\mathbf{v}^{st}$ is divergence-free yields

$$808 \quad \nabla^2 p^{st} \prime(y, z) = \phi \frac{\partial Y(y, z)}{\partial z}. \quad (C.6)$$

809 Note that (C.6) coincides with the first order approximation of the equation of constant density flow
 810 through heterogeneous media. Considering the set-up of Figure 1, equation (C.6) is subject to the
 811 following boundary conditions

$$812 \quad p^{st} \prime(y, z = 0) = 0, \quad \frac{\partial p^{st} \prime(y, z = H)}{\partial z} = 0, \quad \frac{\partial p^{st} \prime(y = 0, z)}{\partial y} = \frac{\partial p^{st} \prime(y = W, z)}{\partial y} = 0. \quad (C.7)$$

813 Solution of (C.6)-(C.7) is

$$814 \quad p^{st} \prime(y, z) = \phi \int_{\Omega} \frac{\partial}{\partial \xi} Y(\eta, \xi) \widehat{G}^F(y, z; \eta, \xi) d\eta d\xi \quad (C.8)$$

815 where $\widehat{G}^F(y, z; \eta, \xi)$ is a Green's function, i.e., it satisfies (C.6)-(C.7) with the source term replaced
 816 by $\delta(\eta - y)\delta(\xi - z)$.

817 A similar procedure yields the following equation satisfied by pressure perturbations

$$818 \quad \nabla^2 p \prime(y, z, t) = \phi \frac{\partial Y(y, z)}{\partial z} - N_g \frac{\partial \rho \prime(y, z, t)}{\partial z}. \quad (C.9)$$

819 Subtracting (C.6) from (C.9) and making use of (17) yields the following equation satisfied by the
 820 dynamic pressure fluctuation

$$821 \quad \nabla^2 p^{dy} \prime(y, z, t) = -N_g \frac{\partial C \prime(y, z, t)}{\partial z}, \quad (C.10)$$

822 subject to the boundary **and initial** conditions

$$823 \quad p^{dy} \prime(y, z = 0, t) = 0, \quad \frac{\partial p^{dy} \prime(y, z = H, t)}{\partial z} = 0, \quad \frac{\partial p^{dy} \prime(y = 0, z, t)}{\partial y} = \frac{\partial p^{dy} \prime(y = W, z, t)}{\partial y} = 0, \quad (C.11)$$

$$p^{dy} \prime(y, z, t = 0) = 0.$$

824 Solution of (C.10)-(C.11) is given by

$$p^{dy}(y, z, t) = -N_g \int_{\Omega} \frac{\partial C'(\eta, \xi, t)}{\partial \xi} \widehat{G}^F(y, z; \eta, \xi) d\eta d\xi. \quad (C.12)$$

Appendix D. Covariance of Vertical Velocity

Making use of (20), the horizontal spatial mean of the covariance of vertical velocity components can be written as

$$\langle v_z'(z, t) v_z'(\xi, \tau) \rangle = \langle v_z^{st}(z) v_z^{st}(\xi) \rangle + \langle v_z^{st}(z) v_z^{dy}(\xi, \tau) \rangle + \langle v_z^{dy}(z, t) v_z^{st}(\xi) \rangle + \langle v_z^{dy}(z, t) v_z^{dy}(\xi, \tau) \rangle \quad (D.1)$$

Considering (C.5) and (17), enables us to write $\langle v_z^{st}(z) v_z^{dy}(\xi, \tau) \rangle$ as

$$\begin{aligned} \langle v_z^{st}(z) v_z^{dy}(\xi, \tau) \rangle &= -\frac{1}{\phi} \left[\frac{\partial}{\partial \xi} \langle Y(z) p^{dy}(\xi, \tau) \rangle - \frac{1}{\phi} \frac{\partial^2}{\partial z \partial \xi} \langle p^{st}(z) p^{dy}(\xi, \tau) \rangle \right] \\ &\quad - \frac{N_g}{\phi} \left[\langle Y(z) C'(\xi, \tau) \rangle - \frac{1}{\phi} \left\langle \frac{\partial p^{st}(z)}{\partial z} C'(\xi, \tau) \right\rangle \right]. \end{aligned} \quad (D.2)$$

Recalling (C.12), the first term on right hand side of (D.2) can be computed as

$$\begin{aligned} -\frac{1}{\phi} \frac{\partial}{\partial \xi} \langle Y(z) p^{dy}(\xi, \tau) \rangle &= -\frac{1}{\phi W} \int_0^W \frac{\partial}{\partial \xi} \langle Y(z, y) p^{dy}(\xi, y, \tau) \rangle dy \\ &= \frac{N_g}{\phi W} \int_0^W \int_0^H \int_0^W \frac{\partial}{\partial \xi} \langle Y(z, y) C'(\hat{\eta}, \hat{\xi}, \tau) \rangle \frac{\partial}{\partial \xi} \widehat{G}^F(y, \xi; \hat{\eta}, \hat{\xi}) d\hat{\eta} d\hat{\xi} dy. \end{aligned} \quad (D.3)$$

We then approximate $\widehat{G}^F(y, \xi; \hat{\eta}, \hat{\xi})$ as

$$\widehat{G}^F(y, \xi; \hat{\eta}, \hat{\xi}) = \delta(y - \hat{\eta}) G^F(\xi; \hat{\xi}), \quad (D.4a)$$

with [10]

$$G^F(\xi; \hat{\xi}) = \begin{cases} \ln \frac{H}{H - \xi} - 2 \sum_{n=1}^{\infty} I_0(\lambda_n, \hat{\xi}) \left[\frac{K_0(\lambda_n, H)}{I_0(\lambda_n, H)} I_0(\lambda_n, H - \xi) - K_0(\lambda_n, H - \xi) \right] & \text{if } 0 \leq \xi \leq H - \hat{\xi} \\ \ln \frac{H}{\hat{\xi}} - 2 \sum_{n=1}^{\infty} I_0(\lambda_n, H - \xi) \left[\frac{K_0(\lambda_n, H)}{I_0(\lambda_n, H)} I_0(\lambda_n, \hat{\xi}) - K_0(\lambda_n, \hat{\xi}) \right] & \text{if } H - \hat{\xi} \leq \xi \leq H \end{cases} \quad (D.4b)$$

Here, I_0 and K_0 respectively are the modified Bessel function of first and second kind and

$\lambda_n = n\pi / H$. Making use of (D.4a), (D.3) becomes

$$\begin{aligned}
& -\frac{1}{\phi} \frac{\partial}{\partial \xi} \left\langle \overline{Y(z) p^{dy} '(\xi, \tau)} \right\rangle \approx \frac{N_g}{\phi W} \int_0^H \int_0^W \frac{\partial}{\partial \hat{\xi}} \left\langle Y(z, y) C'(\hat{\eta} = y, \hat{\xi}, \tau) \right\rangle \frac{\partial}{\partial \xi} G^F(\xi; \hat{\xi}) dy d\hat{\xi} \\
& = \frac{N_g}{\phi} \int_0^H \frac{\partial}{\partial \hat{\xi}} \left\langle \overline{Y(z) C'(\hat{\xi}, \tau)} \right\rangle \frac{\partial}{\partial \xi} G^F(\xi; \hat{\xi}) d\hat{\xi}.
\end{aligned} \tag{D.5}$$

A similar procedure yields the following expressions for the remaining terms of (D.2)

$$\frac{1}{\phi^2} \frac{\partial^2}{\partial z \partial \xi} \left\langle \overline{p^{st} '(z) p^{dy} '(\xi, \tau)} \right\rangle \approx -\frac{N_g}{\phi} \int_0^H \int_0^H \frac{\partial^2}{\partial \tilde{\xi} \partial \hat{\xi}} \left\langle \overline{Y(\tilde{\xi}) C'(\hat{\xi}, \tau)} \right\rangle \frac{\partial}{\partial \xi} G^F(\xi; \hat{\xi}) \frac{\partial}{\partial z} G^F(z; \tilde{\xi}) d\tilde{\xi} d\hat{\xi} \tag{D.6}$$

$$\frac{N_g}{\phi^2} \frac{\partial}{\partial z} \left\langle \overline{p^{st} '(z) C'(\xi, \tau)} \right\rangle \approx \frac{N_g}{\phi} \int_0^H \frac{\partial}{\partial \tilde{\xi}} \left\langle \overline{Y(\tilde{\xi}) C'(\xi, \tau)} \right\rangle \frac{\partial}{\partial z} G^F(z; \tilde{\xi}) d\tilde{\xi} \tag{D.7}$$

Substituting (D.5)-(D.7) into (D.2) leads to

$$\left\langle \overline{v_z^{st} '(z) v_z^{dy} '(\xi, \tau)} \right\rangle \approx \frac{N_g}{\phi} \left\{ \begin{aligned} & -\left\langle \overline{Y(z) C'(\xi, \tau)} \right\rangle \\ & -\int_0^H \int_0^H \frac{\partial^2}{\partial \tilde{\xi} \partial \hat{\xi}} \left\langle \overline{Y(\tilde{\xi}) C'(\hat{\xi}, \tau)} \right\rangle \frac{\partial}{\partial z} G^F(z; \tilde{\xi}) \frac{\partial}{\partial \xi} G^F(\xi; \hat{\xi}) d\tilde{\xi} d\hat{\xi} \\ & + \int_0^H \frac{\partial}{\partial \tilde{\xi}} \left\langle \overline{Y(\tilde{\xi}) C'(\xi, \tau)} \right\rangle \frac{\partial}{\partial z} G^F(z; \tilde{\xi}) d\tilde{\xi} + \\ & + \int_0^H \frac{\partial}{\partial \hat{\xi}} \left\langle \overline{Y(z) C'(\hat{\xi}, \tau)} \right\rangle \frac{\partial}{\partial \xi} G^F(\xi; \hat{\xi}) d\hat{\xi} \end{aligned} \right\}. \tag{D.8}$$

Equation (D.8) indicates that $\left\langle \overline{v_z^{st} '(z) v_z^{dy} '(\xi, \tau)} \right\rangle$ depends **sub-linearly** on N_g / ϕ and is a function of the cross-covariance $\left\langle \overline{Y(z) C'(\xi, \tau)} \right\rangle$.

Following a similar procedure, one can then write $\left\langle \overline{v_z^{dy} '(z, t) v_z^{st} '(\xi)} \right\rangle$, $\left\langle \overline{v_z^{dy} '(z, t) v_z^{dy} '(\xi, \tau)} \right\rangle$ and

$\left\langle \overline{v_z^{st} '(z) v_z^{st} '(\xi)} \right\rangle$ respectively as

$$\left\langle \overline{v_z^{dy} '(z, t) v_z^{st} '(\xi)} \right\rangle \approx \frac{N_g}{\phi} \left\{ \begin{aligned} & -\left\langle \overline{C'(z, t) Y(\xi)} \right\rangle \\ & -\int_0^H \int_0^H \frac{\partial^2}{\partial \tilde{\xi} \partial \hat{\xi}} \left\langle \overline{Y(\tilde{\xi}) C'(\hat{\xi}, t)} \right\rangle \frac{\partial}{\partial z} G^F(z; \tilde{\xi}) \frac{\partial}{\partial \xi} G^F(\xi; \hat{\xi}) d\tilde{\xi} d\hat{\xi} \\ & + \int_0^H \frac{\partial}{\partial \hat{\xi}} \left\langle \overline{C'(z, t) Y(\hat{\xi})} \right\rangle \frac{\partial}{\partial \xi} G^F(\xi; \hat{\xi}) d\hat{\xi} \\ & + \int_0^H \frac{\partial}{\partial \tilde{\xi}} \left\langle \overline{Y(\tilde{\xi}) C'(\hat{\xi}, t)} \right\rangle \frac{\partial}{\partial z} G^F(z; \tilde{\xi}) d\tilde{\xi} \end{aligned} \right\} \tag{D.9}$$

$$\begin{aligned}
855 \quad \left\langle v_z^{dy}(z,t) v_z^{dy}(\xi,\tau) \right\rangle &\approx \left(\frac{N_g}{\phi} \right)^2 \left\{ \begin{aligned} &\left\langle C'(z,t) C'(\xi,\tau) \right\rangle \\ &+ \int_0^H \int_0^H \frac{\partial^2}{\partial \tilde{\xi} \partial \xi} \left\langle C'(\hat{\xi},t) C'(\tilde{\xi},\tau) \right\rangle \frac{\partial}{\partial z} G^F(z;\hat{\xi}) \frac{\partial}{\partial \xi} G^F(\xi;\tilde{\xi}) d\tilde{\xi} d\xi \\ &- \int_0^H \frac{\partial}{\partial \hat{\xi}} \left\langle C'(\hat{\xi},t) C'(\xi,\tau) \right\rangle \frac{\partial}{\partial z} G^F(z;\hat{\xi}) d\hat{\xi} \\ &- \int_0^H \frac{\partial}{\partial \tilde{\xi}} \left\langle C'(\hat{\xi},\tau) C'(z,t) \right\rangle \frac{\partial}{\partial \xi} G^F(\xi;\tilde{\xi}) d\tilde{\xi} \end{aligned} \right\}
\end{aligned}$$

856 (D.10)

$$\begin{aligned}
857 \quad \left\langle v_z^{st}(z) v_z^{st}(\xi) \right\rangle &\approx \left\{ \begin{aligned} &\left\langle Y(z) Y(\xi) \right\rangle \\ &+ \int_0^H \int_0^H \frac{\partial^2}{\partial \hat{\xi} \partial \tilde{\xi}} \left\langle Y(\hat{\xi}) Y(\tilde{\xi}) \right\rangle \frac{\partial}{\partial \xi} G^F(\xi;\hat{\xi}) \frac{\partial}{\partial z} G^F(z;\tilde{\xi}) d\hat{\xi} d\tilde{\xi} - \\ &- \int_0^H \frac{\partial}{\partial \hat{\xi}} \left\langle Y(z) Y(\hat{\xi}) \right\rangle \frac{\partial}{\partial \xi} G^F(\xi;\hat{\xi}) d\hat{\xi} - \\ &- \int_0^H \frac{\partial}{\partial \tilde{\xi}} \left\langle Y(\xi) Y(\tilde{\xi}) \right\rangle \frac{\partial}{\partial z} G^F(z;\tilde{\xi}) d\tilde{\xi} \end{aligned} \right\} \quad (D.11)
\end{aligned}$$

858 Equation (D.10) shows that $\left\langle v_z^{dy}(z,t) v_z^{dy}(\xi,\tau) \right\rangle$ depends **sub-linearly** on $(N_g/\phi)^2$ and is a
859 function of $\left\langle C'(z,t) C'(\xi,\tau) \right\rangle$, i.e., the horizontal spatial mean of concentration covariance. It is also
860 seen that $\left\langle v_z^{st}(z) v_z^{st}(\xi) \right\rangle$ is a function of the covariance of Y .

861 We now introduce the following notation

$$\begin{aligned}
862 \quad \left\langle v'_z(z,t) v'_z(\xi,\tau) \right\rangle &= \mathbf{A} \left\{ \left\langle Y(z) Y(\xi) \right\rangle \right\} + \frac{N_g}{\phi} \mathbf{B} \left\{ \left\langle Y(z) C'(\xi,\tau) \right\rangle \right\} + \\
&+ \frac{N_g}{\phi} \mathbf{C} \left\{ \left\langle C'(z,t) Y(\xi) \right\rangle \right\} + \frac{N_g^2}{\phi^2} \mathbf{D} \left\{ \left\langle C'(z,t) C'(\xi,\tau) \right\rangle \right\} \quad (D.12)
\end{aligned}$$

863 where operators $\mathbf{A}\{ \}$, $\mathbf{B}\{ \}$, $\mathbf{C}\{ \}$, $\mathbf{D}\{ \}$ are respectively defined by the terms in parenthesis
864 appearing at the right hand side of (D.11), (D.8), (D.9) and (D.10). As highlighted by (D.12), one
865 needs to derive the equations satisfied by the cross-covariances $\left\langle C'(z,t) Y(\xi) \right\rangle$ and
866 $\left\langle C'(z,t) C'(\xi,\tau) \right\rangle$ to determine $\left\langle v'_z(z,t) v'_z(\xi,\tau) \right\rangle$. These expressions are respectively derived in
867 Appendices E and F.

868 **Appendix E. Cross covariance between permeability and concentration**

869 Here we derive the horizontal spatial mean of the cross-covariance between the log-permeability
 870 and concentration, i.e., $\langle \overline{Y(z)C'(\xi, \tau)} \rangle$. Multiplying (A.8) by $Y(y, z)$, applying (13) and the
 871 ensemble averaging operator, making use of (A.10) and disregarding products of fluctuations larger
 872 than two yields

$$\begin{aligned}
 \langle \overline{Y(z)C'(\xi, \tau)} \rangle &= \frac{1}{W} \int_0^W \langle Y(z, y) C'(\eta = y, \xi, \tau) \rangle dy \\
 873 \quad &= -\frac{1}{W} \int_0^t \int_0^H \int_0^W \langle Y(z, y) v_z'(y, \hat{\xi}, \hat{\tau}) \rangle \frac{\partial}{\partial \hat{\xi}} \langle \overline{C}(\hat{\xi}, \hat{\tau}) \rangle G_z^T(\xi, \tau, \hat{\xi}, \hat{\tau}) dy d\hat{\xi} d\hat{\tau} \quad (E.1) \\
 &= -\int_0^t \int_0^H \langle \overline{Y(z) v_z'(\hat{\xi}, \hat{\tau})} \rangle G_z^T(\xi, \tau, \hat{\xi}, \hat{\tau}) \frac{\partial}{\partial \hat{\xi}} \langle \overline{C}(\hat{\xi}, \hat{\tau}) \rangle d\hat{\xi} d\hat{\tau}.
 \end{aligned}$$

874 Recalling (C.5), equation (E.1) becomes

$$\begin{aligned}
 \langle \overline{Y(z)C'(\xi, \tau)} \rangle &= -\int_0^t \int_0^H \frac{\partial}{\partial \hat{\xi}} \langle \overline{C}(\hat{\xi}, \hat{\tau}) \rangle G_z^T(\xi, \tau, \hat{\xi}, \hat{\tau}) \\
 875 \quad &\left\{ \langle \overline{Y(z)Y(\hat{\xi})} \rangle - \frac{1}{\phi} \frac{\partial}{\partial \hat{\xi}} \langle \overline{Y(z)p^{st}(\hat{\xi})} \rangle - \frac{N_g}{\phi} \langle \overline{Y(z)C'(\hat{\xi}, \hat{\tau})} \rangle - \frac{1}{\phi} \frac{\partial}{\partial \hat{\xi}} \langle \overline{Y(z)p^{dy}(\hat{\xi}, \hat{\tau})} \rangle \right\} d\hat{\xi} d\hat{\tau} \quad (E.2)
 \end{aligned}$$

876 A numerically analysis of (E.2) (details not shown) allows recognizing that it is possible to
 877 disregard the last term on the right hand side of (E.2) for the test cases considered in this work.
 878 Introducing the operator $\mathbf{F}\{ \}$ defined as

$$879 \quad \mathbf{F}\{\Theta(z; \hat{\xi}, \hat{\tau})\} = -\int_0^t \int_0^H \Theta(z; \hat{\xi}, \hat{\tau}) G_z^T(\xi, \tau, \hat{\xi}, \hat{\tau}) \frac{\partial}{\partial \hat{\xi}} \langle \overline{C}(\hat{\xi}, \hat{\tau}) \rangle d\hat{\xi} d\hat{\tau}, \quad (E.3)$$

880 allows rewriting (E.2) as

$$881 \quad \langle \overline{Y(z)C'(\xi, \tau)} \rangle + \frac{N_g}{\phi} \mathbf{F}\left\{ \langle \overline{Y(z)C'(\hat{\xi}, \hat{\tau})} \rangle \right\} = \mathbf{F}\left\{ \langle \overline{Y(z)Y(\hat{\xi})} \rangle \right\} - \frac{1}{\phi} \mathbf{F}\left\{ \frac{\partial}{\partial \hat{\xi}} \langle \overline{Y(z)p^{st}(\hat{\xi})} \rangle \right\}. \quad (E.4)$$

882 Equation (E.4) provides the link amongst $\langle \overline{Y(z)C'(\xi, \tau)} \rangle$ and (i) the covariance of Y , i.e.,
 883 $\langle \overline{Y(z)Y(\xi)} \rangle$, and (ii) the cross-covariance between *stationary* pressure and permeability, i.e.
 884 $\langle \overline{Y(z)p^{st}(\xi)} \rangle$. Introducing the dimensionless hydraulic head $h = p^{st} / g\rho_f + z$ (v_{BC}^*
 885 $g = \sqrt{k^*} g^* / v_{BC}^{*2}$ being the dimensionless gravity), allows rewriting (E.4) as

$$\langle Y(z)C'(\xi, \tau) \rangle = \mathbf{F} \left\{ \langle \overline{Y(z)Y(\hat{\xi})} \rangle \right\} - \frac{g\rho_f}{\phi} \mathbf{F} \left\{ \frac{\partial}{\partial \hat{\xi}} \langle \overline{Y(z)h'(\hat{\xi})} \rangle \right\} - \frac{N_g}{\phi} \mathbf{F} \left\{ \langle \overline{Y(z)C'(\hat{\xi}, \hat{\tau})} \rangle \right\}, \quad (\text{E.5})$$

where $\langle \overline{Y(z)h'(\hat{\xi})} \rangle$ is the cross-covariance between log-permeability and hydraulic head for the constant density case. The last term on right hand side of (E.5) embeds the stabilizing buoyancy effects which reduce the correlation between permeability and concentration.

Appendix F. Concentration Covariance

Here we derive the equation satisfied by the horizontal spatial mean of concentration covariance, $\langle \overline{C'(z, t)C'(\xi, \tau)} \rangle$. Multiplying (A.8) by $C'(\eta, \xi, \tau)$, taking the ensemble mean operator, making use of (A.10) and neglecting terms involving power of fluctuations larger than two yields

$$\langle C'(z, y, t)C'(\xi, \eta, \tau) \rangle = - \int_0^H \int_0^t \langle v_z'(\hat{\eta} = \eta, \hat{\xi}, \hat{\tau}) C'(z, y, t) \rangle \frac{\partial}{\partial \hat{\xi}} \langle \overline{C(\hat{\xi}, \hat{\tau})} \rangle G_z^T(\xi, \tau, \hat{\xi}, \hat{\tau}) d\hat{\xi} d\hat{\tau}. \quad (\text{F.1})$$

The term $\langle \overline{C'(z, t)C'(\xi, \tau)} \rangle$ is defined as

$$\langle \overline{C'(z, t)C'(\xi, \tau)} \rangle = \frac{1}{W} \int_0^W \langle C'(\eta = y, \xi, \tau) C'(z, y, t) \rangle dy. \quad (\text{F.2})$$

Recalling (C.5), (F.2) becomes

$$\begin{aligned} \langle \overline{C'(z, t)C'(\xi, \tau)} \rangle &= - \int_0^H \int_0^t \frac{\partial}{\partial \hat{\xi}} \langle \overline{C(\hat{\xi}, \hat{\tau})} \rangle G_z^T(\xi, \tau, \hat{\xi}, \hat{\tau}) \\ &\left(\langle \overline{Y(\hat{\xi})C'(z, t)} \rangle - \frac{1}{\phi} \frac{\partial}{\partial \hat{\xi}} \langle \overline{C'(z, t)p^{st}(\hat{\xi})} \rangle - \frac{N_g}{\phi} \langle \overline{C'(z, t)C'(\hat{\xi}, \hat{\tau})} \rangle - \frac{1}{\phi} \frac{\partial}{\partial \hat{\xi}} \langle \overline{C'(z, t)p^{dy}(\hat{\xi}, \hat{\tau})} \rangle \right) d\hat{\xi} d\hat{\tau} \end{aligned} \quad (\text{F.3})$$

A numerically analysis of (F.3) (details not shown) allows recognizing that it is possible to disregard the last term on the right hand side of (F.3) for the test cases considered in this work.

Introducing the operator $\mathbf{H}\{ \}$ defined as

$$\mathbf{H}\{ \Theta(z, t; \hat{\xi}, \hat{\tau}) \} = - \int_0^H \int_0^t \Theta(z, t; \hat{\xi}, \hat{\tau}) G_z^T(\xi, \tau, \hat{\xi}, \hat{\tau}) \frac{\partial}{\partial \hat{\xi}} \langle \overline{C(\hat{\xi}, \hat{\tau})} \rangle d\hat{\xi} d\hat{\tau}, \quad (\text{F.4})$$

allows rewriting (F.3) as

$$\langle \overline{C'(z, t)C'(\xi, \tau)} \rangle = \mathbf{H} \left\{ \langle \overline{Y(\hat{\xi})C'(z, t)} \rangle \right\} - \frac{1}{\phi} \mathbf{H} \left\{ \frac{\partial}{\partial \hat{\xi}} \langle \overline{C'(z, t)p^{st}(\hat{\xi})} \rangle \right\} - \frac{N_g}{\phi} \mathbf{H} \left\{ \langle \overline{C'(z, t)C'(\hat{\xi}, \hat{\tau})} \rangle \right\} \quad (\text{F.5})$$

The last term in (F.5) represents the impact of the stabilizing buoyancy effects and leads to a reduction in the concentration variance as discussed in Section 4.3. Finally, making use of (C.8) allows rewriting (F.5) as

$$\begin{aligned}
& \left\langle \overline{C'(z,t)C'(\xi,\tau)} \right\rangle + \frac{N_g}{\phi} \mathbf{H} \left\{ \left\langle \overline{C'(z,t)C'(\hat{\xi},\hat{\tau})} \right\rangle \right\} = \\
& \mathbf{H} \left\{ \left\langle \overline{Y(\hat{\xi})C'(z,t)} \right\rangle \right\} - \mathbf{H} \left\{ \frac{\partial}{\partial \hat{\xi}} \int_0^H \frac{\partial}{\partial \tilde{\xi}} \left\langle \overline{C'(z,t)Y(\tilde{\xi})} \right\rangle G^F(\hat{\xi},\tilde{\xi}) d\tilde{\xi} \right\} .
\end{aligned}
\tag{F.6}$$

Fig. 1. (a) Flow and transport problem set-up and colorized image of the permeability field for $\sigma_Y^2 = 0.5$. Sample contour plots of normalized concentration for (b) constant density, (c) $N_g = 1$ and (d) $N_g = 10$, with $N_g = \Delta \rho^* \tilde{k}^* g^* / (\mu^* v_{BC}^*)$. Magnification at solute front of the spatial variance of (e) vertical velocity, i.e., $\overline{\sigma_{v_z}^2}$, and (f) of concentration, i.e., $\overline{\sigma_c^2}$.

927

928 Fig. 2. Examples of (a) $\mathbf{v}^{st}$, (b) $\mathbf{v}^{dy}$ and (c) $\mathbf{v}'$ for the variable density flow problem. The
 929 underlying $\mathbf{k}$ field is also depicted. Velocity, $\mathbf{v}$, and concentration, C (depicted as color scale), for
 930 (d) variable and (e) constant density. Vertical distribution of (f) variance of vertical velocity, $\sigma_{v_z}^2$,
 931 and (g) *single realization* dispersive flux (19), i.e., $\overline{v_z' C'}$, for the tracer (red curve) and variable
 932 density (black curve) case. All of the results have been evaluated at $t = 664$.

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Fig. 3. Section-averaged vertical velocity variance, $\langle v_z'^2 \rangle$, and (negative) gradient of ensemble concentration $(-\partial \langle \bar{C} \rangle / \partial z)$ for (a), (c) $\sigma_Y^2 = 0.1$ and $N_g = 1$; (b), (d) $\sigma_Y^2 = 0.1$ and $N_g = 0.1$; (e), (g) $\sigma_Y^2 = 0.5$ and $N_g = 1$; (f), (h) $\sigma_Y^2 = 0.5$ and $N_g = 0.1$. Results are evaluated at dimensionless times $t_1 < t_2 < t_3$.

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942 Fig. 4. Section-averaged vertical velocity covariance components **for** $z = z_2 = 4.27 \times 10^3$: (a, b)
 943 $\langle v_z^{st}(z, t) v_z^{st}(\xi, \tau) \rangle$ (red curve) and $\langle v_z^{st}(z) v_z^{st}(\xi) \rangle$ (black curve); (c, d) $\langle v_z^{dy}(z, t) v_z^{st}(\xi) \rangle$ (insets
 944 show the entire vertical domain); (e, f) $\langle v_z^{st}(z) v_z^{dy}(\xi, \tau) \rangle$; (g, h) $\langle v_z^{dy}(z, t) v_z^{dy}(\xi, \tau) \rangle$ versus ξ for
 945 $\sigma_Y^2 = 0.1$ and $N_g = 1$ (left column) or $N_g = 0.1$ (right column). In (a)-(d) and (g)-(h)
 946 $t = t_2 = 4.4 \times 10^3$; in (e)-(h) $\tau_1 = 4.4 \times 10^3$, $\tau_2 = 3.9 \times 10^3$, $\tau_3 = 3.3 \times 10^3$. Numerical Monte Carlo and
 947 semi-analytical solutions are depicted as solid and dashed curves, respectively.

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Fig. 5. Section-averaged vertical velocity covariance components for $z = z_2 = 4.27 \times 10^3$: (a, b) $\langle v'_z(z, t) v'_z(\xi, \tau) \rangle$ (red curve) and, $\langle v_z^{st}(z) v_z^{st}(\xi) \rangle$ (black curve); (c, d) $\langle v_z^{dy}(z, t) v_z^{st}(\xi) \rangle$ (insets show the entire vertical domain); (e, f) $\langle v_z^{st}(z) v_z^{dy}(\xi, \tau) \rangle$; (g, h) $\langle v_z^{dy}(z, t) v_z^{dy}(\xi, \tau) \rangle$ versus ξ for $\sigma_Y^2 = 0.5$ and $N_g = 1$ (left column) or $N_g = 0.1$ (right column). In (a)-(d) and (g)-(h) $t = t_2 = 4.4 \times 10^3$; in (e)-(h) $\tau_1 = 4.4 \times 10^3$, $\tau_2 = 3.9 \times 10^3$, $\tau_3 = 3.3 \times 10^3$. Numerical Monte Carlo and semi-analytical solutions are depicted as solid and dashed curves, respectively.

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961 Fig. 6. Section-averaged log-permeability and concentration cross-covariance, $\langle Y(z)C'(\xi, \tau) \rangle$,
 962 versus ξ when $z = z_2 = 4.27 \times 10^3$ (black curves) or versus z when $\xi = \xi_2 = 4.27 \times 10^3$ (red curves) at
 963 time $\tau = t_2 = 4.4 \times 10^3$ for $\sigma_Y^2 = 0.1, 0.5$ and (a,c) $N_g = 1$, or (b,d) $N_g = 0.1$. The results obtained for
 964 the tracer case (i.e., $\rho = \text{const}$) are also depicted for $\langle Y(z_2)C'(\xi, \tau) \rangle$ (blue curves) and
 965 $\langle Y(z)C'(\xi_2, \tau) \rangle$ (green curves).

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969 Fig. 7. Section-averaged ensemble dispersive flux, $\langle v_z'(z,t)C'(z,t) \rangle$, and concentration variance,
 970 $\sigma_c^2(z,t) = \langle C'(z,t)^2 \rangle$, for $N_g = 1$, (black curves), $N_g = 0.1$, (red curves), $\rho = const$, (blue curves),
 971 (a)-(b) for $\sigma_Y^2 = 0.1$ and (c)-(d) for $\sigma_Y^2 = 0.5$. Numerical Monte Carlo and semi-analytical solutions
 972 of (25) (for $\langle v_z'C' \rangle$) and (F.6) (for $\sigma_c^2(z,t)$) are depicted as solid and dashed curves, respectively.
 973 Results are evaluated at dimensionless times $t_1 < t_2 < t_3$.