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Towards a filtered density function approach for reactive transport in groundwater

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ABSTRACT

Evolution equations for probability density functions (PDFs) and filtered density functions (FDFs) of random species concentrations weighted by conserved scalars are formulated as Fokker–Planck equations describing stochastically equivalent processes in concentration-position spaces. This approach provides consistent numerical PDF/FDF solutions, given by the density in the concentration-position space of an ensemble of computational particles governed by the associated Itô equations. The solutions are obtained by a global random walk (GRW) algorithm, which is stable, free of numerical diffusion, and practically insensitive to the increase of the number of particles. The general FDF approach and the GRW numerical solution are illustrated for a reduced complexity problem consisting of the transport of a single scalar in groundwater. Randomness is induced by the stochastic parameterization of the hydraulic conductivity, characterized by short range correlations and small variance. The objective is to infer the statistics of the random concentration sampled at the plume center of mass, integrated over the transverse dimension of a two-dimensional spatial domain. The PDF/FDF problem can therefore be formulated in a two-dimensional domain as well, a spatial dimension and one in the concentration space. The upscaled drift and diffusion coefficients describing the PDF transport in the physical space are estimated on single-trajectories of diffusion in velocity fields with short-range correlations, owing to their self-averaging property. The mixing coefficients describing the PDF transport in concentration spaces are parameterized by the trend and the noise inferred from the statistical analysis of an ensemble of simulated concentration time series, as well as by classical mixing models. A Gaussian spatial filter applied to a Kraichnan velocity field generator is used to construct coarse-grained simulations (CGS) for FDF problems. The purposes of the CGS simulations are two-fold: first to understand the significance of the FDF approach from a practical point of view and its relation to the PDF approach; second to investigate the limits of the mixing models considered here and the desirable features of the mixing models for groundwater systems.

1. Introduction

Geological formations are heterogeneous and their properties are normally not measurable everywhere. This lack of knowledge implies uncertainty in aquifer parameters like hydraulic conductivity. As a consequence, quantifying the transport of solutes through these formations is also uncertain. This uncertainty is dealt with by using a probabilistic description of the involved processes. Going beyond mean values and also considering the variance is a good starting point to take the uncertainty into account. But in the case of risk assessments, the need to predict extreme values of contaminant concentration becomes important. However, such occurrences cannot be described by the variance alone. Hence, the need to describe the solute concentrations by their complete probability density function (PDF) arises. Another major advantage of the PDF approach is the easy way to handle reactive solutes if the reaction rates depend only on the set of species concentrations. Then, even highly nonlinear reactions can simply be included in the formulation without the need to approximate them.

PDF methods were originally developed in the context of modeling turbulent reacting flows as a powerful tool to close highly nonlinear terms arising from averaging chemical reaction rates [1–5]. The PDF approach is based on solving evolution equations

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for the one-dimensional (one-point one-time) Eulerian joint PDF of sets of variables describing the state of the system, for example velocity, chemical composition, turbulent frequency, temperature or enthalpy. PDF equations are derived by different methods [1,2] from local balance equations governing the flow and the evolution of the thermochemical state of the system. The PDF equations are unclosed because they contain terms that cannot be determined by the one-dimensional PDF alone [2]. But, irrespective of the complexity of the set of state variables, the chemical source terms in the PDF equations are closed, namely, they have the same functional form as the unaveraged reaction rates in the local balance equations [6]. The unclosed terms which require modeling are those describing the turbulent frequency, the scalar mixing by molecular diffusion, and, when the velocity is not included among the state variables, the effects of the turbulent velocity fluctuations [4,6].

Filtered density function (FDF) methods provide an alternative approach using spatially filtered quantities, that is, spatial averages, instead of stochastic averages on which the PDF methods are based [6–9]. The meaning of the FDF is that of a PDF of state variables at scales smaller than the filter width [10,11]. FDF evolution equations can be derived by similar procedures and have the same structure, with source terms in closed form, as the PDF equations [5–7]. With rare exceptions, the only state variables considered in FDF methods are the scalars describing the thermochemical composition [6]. Unlike the PDF, which is a deterministic function, the FDF is a random quantity. Its expectation tends to the PDF only in the limit of a small filter width [6]. However, since, by invoking an ergodic theorem, the ensemble averaging may be seen as a filtering in space with a filter width much smaller than the domain but larger than the characteristic length of large-scale motions, the FDF should approach the PDF in the limit of large filter width [10]. Even though this assertion has not been proved theoretically, in some cases the convergence may be demonstrated numerically, as we will see below in this paper.

In modeling transport through highly heterogeneous natural groundwater systems, the randomness is introduced by the stochastic parameterization of the hydraulic conductivity, which accounts for the parameter uncertainty due to a lack of measurements. This stochasticity implies the randomness of the Darcy flow velocity and of the dependent variables of the transport equations [12]. Randomness in modeling groundwater flows can be enhanced by considering uncertain sources in the flow equations (random recharge) [13] or, for transient flows, random parameterizations of storage coefficients [14]. However, the uncertainty of the hydraulic conductivity is an omnipresent source of randomness and we focus on it in this paper. While in earlier stochastic approaches the focus was mainly on the mean and in some cases the variance of the concentration, during the last decade the need to model the concentration PDF received an increased attention [12,15–21]. Concentration PDFs of conserved scalars may be inferred without solving PDF evolution equations in case of small or moderate fluctuations of the hydraulic conductivity, modeled as a lognormal random function with finite correlation lengths. Then, a Gaussian shape of the concentration may be assumed or inferred, which is completely determined by its first two moments. The statistics of these moments, specified under various assumptions in a first-order perturbation approach, is finally "mapped", using the Gaussian functional shape of the concentration, onto the concentration PDF via numerical [12] or analytical techniques [16,18]. Another favorable situation is that of stratified transport, when the Gaussian concentration can be expressed explicitly as a function of the hydraulic conductivity, with the only assumption of negligible transverse dispersion. This leads to an explicit functional dependence of the concentration PDF on the hydraulic conductivity [15]. This approach also provides the PDF of reacting chemical species if their concentrations are fully defined by monotonous functions of conserved scalars [15].

For advective–reactive transport, PDFs of reacting species can be computed by solving evolution equations similar to those used in turbulence [19]. Such PDF equations do not contain mixing terms, because molecular diffusion is neglected. The only closure problem concerns terms due to velocity fluctuations, which are modeled as effective, or upscaled, diffusion coefficients, leading to Fokker–Planck evolution equations [19,21]. By considering the velocity among the state variables, velocity–concentration PDF equations similar to turbulence problems can be derived and no closure for velocity fluctuations is necessary. Mixing is modeled similarly to turbulence approaches and the concentration PDF is obtained by integrating the joint velocity–concentration PDF over the velocity state space [17]. Evolution equations of the concentration PDF weighted by a conserved scalar, which generalizes the mass density function used in turbulence [2], can be formulated as Fokker–Planck equations [21]. Closures are provided by stochastically upscaled diffusion coefficients [20,21] and by mixing models, which are formulated as a diffusion in concentration space. The parameters for this diffusion process can be inferred from measured/simulated concentration time series [22].

While several PDF approaches are being developed [17,19,21], FDF methods were not yet used in modeling transport in groundwater systems. The purpose of this paper is to assess the feasibility and the utility of the FDF approach in stochastic subsurface hydrology. To begin, we notice the similarities between large eddy simulations (LES) in turbulence [7,8,11] and some approaches to coarse-scale, or coarse-grained simulations (CGS) in porous media [23–28]. In both cases, spatial averages of dependent variables are used to coarsen the grid, whereas subgrid effects are modeled. The objective is to obtain results comparable to fine grid simulations at reduced computational costs. In case of reactive transport, the upscaled equations obtained by spatial filtering contain unclosed averaged reaction terms. In LES, the closure problem is solved by coupling the filtered equations to FDF evolution equations [11]. In subsurface hydrology, effective reaction rates needed to close the problem can only be determined for specific problems under simplifying assumptions [29]. FDF approaches can be used to avoid the need to close the filtered reaction terms and CGS can be designed, based on numerical upscaling through volume averages [25,26], multiscale finite element methods [27] or, similarly to LES, by solving filtered equations [23,24].

Nevertheless, there are three major differences with respect to LES-FDF modeling for turbulent flows. The first one concerns the number of parameters. While only a few parameters are required to solve filtered LES equations [7,8], upscaling flow and transport processes in groundwater, by either spatial or stochastic averaging, requires fields of parameters: the hydraulic conductivity [24,26] or the velocity field [20,29]. The second difference, related to the first one, is the origin of the randomness. Turbulent flows are governed by the deterministic Navier–Stokes equations but, for large Reynolds numbers, the flow velocity behaves as a random variable due the sensible dependence of the solution on initial and boundary conditions. This mathematical aspect corresponds to an experimental lack of reproducibility of the measurements in turbulent systems [2]. In groundwater systems, the spatial variability of the hydraulic conductivity cannot be completely described. Therefore, stochastic parameterizations by random space functions are used to account for this uncertainty. The flow equations are thus solved in a probabilistic sense [18]. In this case, randomness is caused by the uncertainty of the parameter fields propagated through the flow and transport equations, which have to be modeled as stochastic equations. The third difference is given by the available experimental data. In turbulence, detailed velocity, temperature, or concentration profiles are available from