

# Asymptotic properties of the Eulerian truncation approximation: Analysis of the perfectly stratified transport problem

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[1] It is well known that stochastic groundwater problems are difficult to solve without making approximations of one kind or another. A popular approximation used in both subsurface flow and transport applications is the so-called “Eulerian truncation.” This approximation, which relies on a perturbation expansion of the governing equation, neglects certain terms involving products of small fluctuations. Several recent publications [e.g., *Dagan and Neuman*, 1991; *Neuman and Orr*, 1993; *Neuman*, 1993] question the validity of Eulerian truncation, arguing that it is inconsistent because neglected terms are of the same order as those retained. In this paper we analyze the asymptotic properties of the Eulerian truncation solution, using a Taylor series approach which clearly reveals the properties of different order approximations. We show that the Eulerian truncation does, in fact, generate an asymptotic ensemble mean expansion for the perfectly stratified example considered by *Dagan and Neuman* [1991]. Moreover, we show that the Eulerian truncation solution to this problem is identical to the well-known cumulant expansion approximation, which is also based on an asymptotic expansion. *INDEX TERMS*: 1832 Hydrology: Groundwater transport; 1829 Hydrology: Groundwater hydrology; 1869 Hydrology: Stochastic processes; *KEYWORDS*: stochastic methods, Eulerian truncation, perturbation methods, field-scale transport, stratified transport, Eulerian methods

## 1. Introduction

[2] Stochastic methods provide a convenient way to deal with the effects of geological variability on subsurface flow and transport. These methods do not try to resolve detailed fluctuations in the variables of interest but focus instead on mean quantities which describe large-scale trends when certain conditions (ergodicity requirements) are satisfied. One of the fundamental problems in stochastic subsurface hydrology is derivation of the mean concentration of a solute moving in a heterogeneous velocity field [*Dagan*, 1987; *Gelhar and Axness*, 1983; *Neuman et al.*, 1987]. In this case the goal is to relate the concentration mean (or perhaps the spatial moments of this mean) to the ensemble moments of random velocity fluctuations and relevant large-scale variables such as the mean velocity. In certain special cases it is possible to derive exact expressions for the concentration mean. More often, approximations must be introduced in order to make the problem tractable. A number of approximation methods have been proposed in the literature [see, e.g., *Bakr et al.*, 1978; *Gelhar and Axness*, 1983; *Dagan*, 1987; *Neuman et al.*, 1987; *Koch and Brady*, 1988; *Van Lent and Kitaniadis*, 1989; *Graham and McLaughlin*, 1989; *Li and McLaughlin*, 1991, 1995; *Cushman et al.*, 1995; *Hu et al.*, 1995; *Yeh et al.*, 1996]. Many of these rely on truncated

expansions of the solution or of particular terms in the governing stochastic transport equations.

[3] Here we focus on an Eulerian approximation method (Eulerian truncation) which has been widely used in both subsurface flow and transport applications [*Bakr et al.*, 1978; *Gelhar and Axness*, 1983; *Koch and Brady*, 1988; *Graham and McLaughlin*, 1989; *Li and McLaughlin*, 1991, 1995; *Cushman et al.*, 1995; *Hu et al.*, 1995; *Zhang*, 2001]. *Dagan and Neuman* [1991] (DN) provide a brief summary and critique of this approach. In particular, they conclude (1) that Eulerian truncation provides a low-order mean approximation which differs from the result obtained from asymptotic expansions generated with Lagrangian and cumulant expansion methods and (2) that the Eulerian truncation leads to a nonasymptotic expansion which is inconsistent in the sense that it neglects terms of the same order of terms that are retained. These conclusions are based largely on a particular example which is convenient because it illustrates the issues very clearly and it has an exact solution which can serve as a basis for comparison. The DN example is the well known case of solute transport in a perfectly stratified porous medium with no local dispersion [e.g., *Dagan and Neuman*, 1991; *Gelhar*, 1993; *Shvidler*, 1993; *Warran and Skiba*, 1964; *Mercado*, 1967; *Simmons*, 1982]. In this paper we review the key elements of the DN analysis and show that Eulerian truncation provides an approximation for the perfectly stratified problem which

is, in fact, asymptotic. Moreover, we show that the Eulerian truncation solution to this problem is identical to the well-known cumulant expansion approximation, which is also based on an asymptotic expansion. The results obtained for the perfectly stratified case indicate that the Eulerian truncation approach provides a reasonable and consistent way to describe the large-scale features of solute transport through heterogeneous porous media.

## 2. Exact Lagrangian Solution to the Perfectly Stratified Transport Problem

[4] The perfectly stratified transport problem considered in DN makes several highly simplified but analytically convenient assumptions about transport of a conservative solute in a heterogeneous porous medium. This medium is assumed to be in a vertical plane that extends infinitely far in the horizontal ( $x_1$ ) direction and consists of an infinite number of geologically uniform strata layered in the vertical ( $x_2$ ) direction. The layer boundaries and velocity vectors are aligned with the  $x_1$  coordinate. The horizontal velocity  $u_1(x_2)$  depends only on  $x_2$  and is uniform in the  $x_1$  direction. The vertical velocity  $u_2$  is zero. The  $u_1$  velocity values in different layers are independent random variables. The concentration profile in each layer is random by virtue of its dependence on  $u$ . Molecular diffusion and local dispersion are ignored. Given these assumptions, the relevant solute transport equation is (with  $x_1$  denoted by  $x$  and  $u$  by  $u_1(x_2)$ ), for any given  $x_2$ :

$$\frac{\partial c(x, t)}{\partial t} + u \frac{\partial c(x, t)}{\partial x} = 0 \quad (1)$$

The initial concentration distribution is  $c(x, 0) = c_{in}(x)$ . Following DN, we assume that the function  $c_{in}(x)$  is infinitely differentiable (an example is a Gaussian function). From a mathematical viewpoint, this is a one-dimensional transport equation with a scalar random coefficient  $u$ .

[5] Our goal is to solve for the ensemble mean concentration, given information on the statistical moments of the random velocity  $u$ . The velocity and concentration may be decomposed into their respective means  $\bar{u}$  and  $\bar{c}(x, t)$  and zero-mean random fluctuations  $u'$  and  $c'(x, t)$  about these means:

$$u = \bar{u} + u' \quad (2)$$

$$c(x, t) = \bar{c}(x, t) + c'(x, t) \quad (3)$$

Note that we write the mathematical expectation operation with an overbar (e.g.  $\bar{c}$ ) while DN use brackets (e.g.  $\langle c \rangle$ ). Also, we write the total velocity, the mean velocity, and the fluctuation of the velocity about its mean as  $u$ ,  $\bar{u}$  and  $u'$  while DN adopt the symbols  $V$ ,  $U$  and  $u$  for the same quantities. Otherwise, we use essentially the same notation.

[6] Following DN, we introduce the moving coordinate  $\mu = x - \bar{u}t$  and write (1) as:

$$\frac{\partial c(\mu, t)}{\partial t} + u' \frac{\partial c(x, t)}{\partial \mu} = 0 \quad (4)$$

Using the terminology of the cumulant expansion approach, the right-hand side of the moving coordinate transport equation can be divided into terms containing a non-random

(“sure”) spatial differential operator  $A_0$  and a random spatial differential operator  $A_1$ :

$$\frac{\partial c(\mu, t)}{\partial t} = [A_0 + A_1]c(\mu, t) \quad (5)$$

where:

$$A_0 = 0$$

$$A_1 = -u' \frac{\partial}{\partial \mu}$$

Note that, for the perfectly stratified problem, the transformation to moving coordinates yields a zero sure operator and a random operator that is linear in the random constant  $u'$ .

[7] The perfectly stratified transport problem has the advantage of having a known mean concentration solution when  $u$  has a Gaussian distribution. This solution can be derived with a Lagrangian approach based on an asymptotic expansion of the random function  $u(X)$  in DN equation (7), where  $X(t)$  is the random location (displacement) of a solute particle and  $u(X)$  is the velocity at that location. For the perfectly stratified problem the  $u(X)$  expansion yields a series of exact expressions for the moments of  $X$  (DN equation (14)), even when  $u$  is not Gaussian. If  $u$  is Gaussian the mean concentration obeys the following (moving coordinate) “pseudo-Fickian” transport equation, which can be used to recover the exact spatial moments:

$$\begin{aligned} \frac{\partial \bar{c}(\mu, t)}{\partial t} &= \left[ \int_0^t \sigma_u^2 dt \right] \frac{\partial^2 \bar{c}(x, t)}{\partial \mu^2} \\ &= [\sigma_u^2 t] \frac{\partial^2 \bar{c}(x, t)}{\partial \mu^2} \\ &= D(t) \frac{\partial^2 \bar{c}(x, t)}{\partial \mu^2} \end{aligned} \quad (6)$$

The particular expression obtained for  $\bar{c}(\mu, t)$  depends on the initial condition.

[8] The exact solution to the perfectly stratified transport problem can also be expressed as an infinite series which is obtained by noting that the exact (random) solution to (4) is:

$$c(\mu, t) = c_{in}(\mu - u't) = c_{in}(x - \bar{u}t - u't) \quad (7)$$

That is, the true concentration in each layer (each random replicate) has a spatial profile identical (for all time) to  $c_{in}(x)$  but translated forward at the velocity  $\bar{u} + u't$ . Since  $c_{in}$  is differentiable we can expand  $c_{in}(\mu - u't)$  in a Taylor series to obtain:

$$c(\mu, t) = c_{in}(\mu) + \sum_{n=1}^{\infty} \frac{(-u't)^n}{n!} \frac{\partial^n c_{in}}{\partial \mu^n} \quad (8)$$

The mean of  $c(\mu)$  is then:

$$\bar{c}(\mu, t) = c_{in}(\mu) + \sum_{n=1}^{\infty} \frac{\overline{(-u')^n} t^n}{n!} \frac{\partial^n c_{in}}{\partial \mu^n} \quad (9)$$

This series can also be obtained by applying the asymptotic perturbation expansion method of [e.g., *Van Dyke*, 1964; *Dagan*, 1989; *Hu et al.*, 1999] to the perfectly stratified transport problem.

[9] If  $u'$  is a Gaussian random variable, as is assumed here and in DN, the ensemble moments of odd powers of  $u'$  are zero while the even powers are related to the velocity variance  $\partial/\sigma_u^2$  [*Kendall and Stuart*, 1977]. In this case, the Taylor series for  $\bar{c}$  becomes:

$$\begin{aligned}\bar{c}(\mu, t) &= c_{in}(\mu) + \sum_{m=1}^{\infty} \frac{\sigma_u^{2m} t^{2m}}{2^m m!} \frac{\partial^{2m} c_{in}(\mu)}{\partial \mu^{2m}} \\ &= c_{in}(\mu) + \frac{1}{2} \frac{\partial}{\sigma_u^2} t^2 \frac{\partial^2 c_{in}(\mu)}{\partial \mu^2} + \frac{1}{8} \sigma_u^4 t^4 \frac{\partial^4 c_{in}(\mu)}{\partial \mu^4} \\ &\quad + \text{terms of order } \sigma_u^6 \text{ and higher}\end{aligned}\quad (10)$$

DN note that the large-time exact mean solution to the perfectly stratified transport problem with Gaussian  $u'$  has a Gaussian spatial profile with the spatial moments given in DN equation (15). This implies that the series in (10) converges to a Gaussian function for large  $t$ . We will use (10) in our analysis of the asymptotic properties of the Eulerian truncation approximation.

### 3. Approximate Eulerian Solutions to the Perfectly Stratified Transport Problem

[10] Although the perfectly stratified transport problem has an exact solution, we can also derive an approximate solution using either of two Eulerian methods: (1) the cumulant expansion approach [*Sposito and Barry*, 1987; *Sposito et al.*, 1991; *van Kampen*, 1992] or (2) the Eulerian truncation approach [*Gelhar and Axness*, 1983; *Koch and Brady*, 1988; *Graham and McLaughlin*, 1989; *Li and McLaughlin*, 1991, 1995; *Dagan and Neuman*, 1991]. DN state that the asymptotic cumulant expansion approach reproduces the exact Lagrangian result of (6) for the perfectly stratified transport problem while the Eulerian truncation does not. They also state that the Eulerian truncation expansion is not asymptotic because it is based on an inconsistent approximation. By contrast, we show below that the cumulant expansion and Eulerian truncation approaches give identical asymptotic approximations for the mean concentration.

[11] The cumulant expansion approximation is based on the following formal (operator) solution to (5) [*Sposito et al.*, 1991; *van Kampen*, 1992]:

$$c(\mu, t) = c(\mu, 0) \left[ \int_0^t \exp(A_1(t')) dt' \right] \quad (11)$$

where  $A_1 = -u' \partial/\partial \mu$  ( $A_0$  is omitted because it is zero when the problem is posed in moving coordinates). An approximate mean concentration solution can be obtained from a formal second-order MacLaurin expansion of the exponential operator in this equation [*Sposito and Barry*, 1987]. This solution satisfies the following differential equation [*Sposito et al.*, 1991]:

$$\frac{\partial \bar{c}(\mu, t)}{\partial t} = \left[ \int_0^t \overline{A_1(t') A_1(t'')} dt' \right] \bar{c}(\mu, t) \quad (12)$$

When the appropriate expression is substituted for  $A_1(t)$  and the assumptions of the perfectly stratified transport problem are applied, the mean equation becomes:

$$\begin{aligned}\frac{\partial \bar{c}(\mu, t)}{\partial t} &= \left[ \int_0^t \overline{u' u'} \frac{\partial}{\partial \mu} \frac{\partial}{\partial \mu} \right] \bar{c}(\mu, t) dt \\ &= \sigma_u^2 \int_0^t \frac{\partial^2 \bar{c}(x, t)}{\partial \mu^2} dt\end{aligned}\quad (13)$$

This cumulant expansion approximation is identical to DN equation (26). It may also be obtained by replacing  $x$  with  $\mu + \bar{u}t$  and applying the assumptions of the perfectly stratified transport problem in the derivation given in equations (3) through (9) of *Sposito and Barry* [1987].

[12] The Eulerian truncation approach replaces the random concentration in (4) by the moving coordinate version of (3). The result is the following random equation:

$$\frac{\partial \bar{c}(\mu, t)}{\partial t} + \frac{\partial c'(\mu, t)}{\partial t} + u' \frac{\partial \bar{c}(x, t)}{\partial \mu} + u' \frac{\partial c'(x, t)}{\partial \mu} = 0 \quad (14)$$

The mean of this equation is:

$$\frac{\partial \bar{c}(\mu, t)}{\partial t} = -\frac{\partial}{\partial \mu} \overline{[u' c'(\mu, t)]} \quad (15)$$

If (15) is subtracted from (14) we obtain the following equation for the concentration perturbation  $c'$ :

$$\frac{\partial c'(\mu, t)}{\partial t} = -u' \frac{\partial \bar{c}(\mu, t)}{\partial \mu} - \frac{\partial}{\partial \mu} [u' c'(\mu, t) - \overline{u' c'(\mu, t)}] \quad (16)$$

Both (15) and (16) are exact, but the mean equation depends on the unknown closure term  $\overline{u' c'(\mu, t)}$ . This closure term may be derived from the perturbation equation if certain approximations are introduced. The Eulerian truncation approximation of interest here assumes that, if  $\sigma_u$  is sufficiently small, the second (bracketed) term on the right-hand side of (16) may be dropped. This approximation is based on the argument that the truncated term should be of higher-order in  $\sigma_u$  than the retained term because it depends on the product of the perturbations  $u'$  and  $c'$ .

[13] Once the Eulerian truncation approximation is introduced the unknown closure term  $\overline{u' c'(\mu, t)}$  may be derived by multiplying the truncated perturbation equation by  $u'$ , taking the expectation, and integrating over time. The resulting approximate mean and perturbation equations are:

$$\frac{\partial \bar{c}_1(\mu, t)}{\partial t} = \frac{\partial}{\sigma_u^2} \int_0^t \frac{\partial^2 \bar{c}_1}{\partial \mu^2} dt' \quad (17)$$

$$\frac{\partial c'_1(\mu, t)}{\partial t} = -u' \frac{\partial \bar{c}_1}{\partial \mu} \quad (18)$$

Since the initial condition is presumed to be known perfectly, we use  $c'(\mu, 0) = 0$  in the temporal integration of

(16). The subscript on  $\bar{c}_1$  indicates that the mean concentration obtained from this equation is an approximation (see below).

[14] The Eulerian truncation mean equation (17) is identical to DN equation (26) (also derived from an Eulerian truncation approximation) and to the result (13) derived earlier from the cumulant expansion approach. The equivalence of the Eulerian truncation and cumulant expansion results demonstrated here contradicts the DN assertion that the cumulant expansion and Eulerian truncation approaches give different results for the perfectly stratified transport problem (see the second column of page 3250 of DN). It should be noted that the approximate cumulant expansion and Eulerian truncation mean equations differ from the exact mean concentration expression of (5). In the terminology of DN the approximations describe ‘‘convolutive non-Fickian’’ while the exact equation describes ‘‘pseudo-Fickian’’ behavior. The dispersion term of convolutive non-Fickian approximation integrates the second spatial derivative of the mean concentration over time while the exact pseudo-Fickian result does not.

[15] Both DN (in their equation (29)) and *Koch and Brady* [1988] note that (17) can be written as a wave equation:

$$\frac{\partial^2 \bar{c}_1}{\partial t^2} = \frac{\partial}{\sigma_u^2} \frac{\partial^2 \bar{c}_1}{\partial \mu^2} \quad (19)$$

A solution which satisfies the initial condition is:

$$\bar{c}_1(\mu, t) = \frac{1}{2} c_{in}(\mu - \sigma_u t) + \frac{1}{2} c_{in}(\mu + \sigma_u t) \quad (20)$$

This approximate solution, which consists of two gradually diverging but non-dispersive pulses, has the correct first and second spatial moments. But the quality of the approximation deteriorates with time as the mean concentration becomes bimodal and eventually splits into two parts. This is clearly incompatible with the exact solution, which yields a unimodal Gaussian spatial profile for large time. The non-physical large-time behavior of (20) led DN to question whether the series expansion generated by the Eulerian truncation approach is asymptotic. This topic is addressed in the next subsection.

#### 4. Asymptotic Properties of the Eulerian Truncation Approximation

[16] In their analysis, DN point out that the Eulerian truncation approach may be applied repeatedly, with each perturbation expression used to derive the closure term for a new mean equation. This process generates a series of corrections to the original approximate mean and perturbation expressions ( $\bar{c}_1$  and  $c'_1$ ) which may be assembled in the following infinite series:

$$\bar{c} \approx \bar{c}_1 + \bar{c}_2 + \bar{c}_3 + \dots ; \quad (21)$$

$$\bar{c}_1(\mu, 0) = c_{in}(\mu) , \quad \bar{c}_n(\mu, 0) = 0 \text{ for } n > 1$$

$$c' \approx c'_1 + c'_2 + c'_3 + \dots ; \quad c'_n(\mu, 0) = 0 \quad \forall n \quad (22)$$

If the Eulerian truncation is valid the  $\bar{c}$  expansion of (21) should be asymptotic and should converge to the exact mean.

[17] The terms in (21) and (22) obey the following recursions (DN equations (32) and (33)):

$$\frac{\partial \bar{c}_n}{\partial t} = - \frac{\partial}{\partial \mu} \overline{uc'_n} \quad (23)$$

$$\frac{\partial c'_n}{\partial t} = -u' \frac{\partial \bar{c}_n}{\partial \mu} - \frac{\partial}{\partial \mu} (u' c'_{n-1}) + \frac{\partial}{\partial \mu} \overline{u' c'_{n-1}} ; \quad c'_0 \equiv 0 \quad (24)$$

When  $n=1$  these expressions correspond to the mean and perturbation equations (17) and (18) obtained from the standard Eulerian truncation procedure described above and their solutions are the Eulerian truncation approximations  $\bar{c}_1$  and  $c'_1$  defined in (17) and (18).

[18] We can identify the form of the  $\bar{c}_1$  term in (21) by expanding the approximate solution (20) in a Taylor series:

$$\begin{aligned} \bar{c}_1(\mu, t) &= c_{in}(\mu, t) + \sum_{m=1}^{\infty} \frac{\sigma_u^{2m} t^{2m}}{(2m)!} \frac{\partial^{2m} c_{in}(\mu)}{\partial \mu^{2m}} \\ &= c_{in}(\mu, t) + \frac{1}{2} \frac{\partial}{\sigma_u^2} t^2 \frac{\partial^2 c_{in}(\mu)}{\partial \mu^2} + \frac{1}{24} \sigma_u^4 t^4 \frac{\partial^4 c_{in}(\mu)}{\partial \mu^4} \\ &\quad + \text{terms of order } \sigma_u^6 \text{ and higher} \end{aligned} \quad (25)$$

Note that the leading two terms of this approximate solution and the exact solution given in (10) are the same. Also, these series converge to the same result as  $\sigma_u \rightarrow 0$ . At orders higher than  $\partial/\sigma_u^2$  the approximate series has the same general form as the exact series but the constant coefficients differ.

[19] It is difficult to analyze the asymptotic properties of the mean expansion in (21) directly since each of the terms in this expansion depends on  $\sigma_u$  in a complex way. For example, we have already seen that the Taylor series version of  $\bar{c}_1$ , which forms part of the  $\bar{c}$  expansion, depends on all even powers of  $\sigma_u$ . In order to explore the asymptotic behavior of (21) we follow the indirect analysis of DN, which focuses on implicit equations for  $\bar{c}_2$ ,  $\bar{c}_3$ , etc.

[20] DN use (23) and (24) to derive an implicit integral expression for the Eulerian truncation approximation  $\bar{c}_1$ , which is shown here together with the explicit solution from (20):

$$\bar{c}_1(\mu, t) = \frac{\partial}{\sigma_u^2} \int_0^t \int_0^{t'} \frac{\partial^2 \bar{c}_1(\mu, t'')}{\partial \mu^2} dt'' dt' + c_{in}(\mu) \quad (26)$$

$$= \frac{1}{2} c_{in}(\mu - \sigma_u t) + \frac{1}{2} c_{in}(\mu + \sigma_u t) \quad (27)$$

They use a similar approach to derive the following explicit expression for the correction term  $c_2$  (see the discussion following DN equation (38)):

$$\bar{c}_2(\mu, t) = 0 \quad (28)$$

Although DN do not provide an explicit expression for the next correction term  $\bar{c}_3$ , they derive the following

differential equation for the case of a Gaussian  $u'$  (DN equation (44)):

$$\begin{aligned} \frac{\partial \bar{c}_3(\mu, t)}{\partial t} &= \frac{\partial}{\sigma_\mu^2} \int_0^t \frac{\partial^2 \bar{c}_3(\mu, t')}{\partial \mu^2} dt' \\ &+ 2 \frac{\partial}{\sigma_\mu^2} \int_0^t \frac{\partial^2 \bar{c}_1(\mu, t')}{\partial \mu^2} dt' \\ &- 2 \frac{\partial}{\sigma_\mu^2} t \frac{\partial^2 c_{in}(\mu)}{\partial \mu^2} \end{aligned} \quad (29)$$

DN then use (29) to derive a differential equation for  $\bar{c} \approx \bar{c}_1 + \bar{c}_2 + \bar{c}_3$ , the next higher-order mean approximation which is different from  $\bar{c}_1$ :

$$\begin{aligned} \frac{\partial \bar{c}(\mu, t)}{\partial t} &= \frac{\partial}{\sigma_\mu^2} \int_0^t \frac{\partial^2 \bar{c}(\mu, t')}{\partial \mu^2} dt' \\ &+ 2 \frac{\partial}{\sigma_\mu^2} \int_0^t \frac{\partial^2 \bar{c}_1(\mu, t')}{\partial \mu^2} dt' \\ &- 2 \frac{\partial}{\sigma_\mu^2} t \frac{\partial^2 c_{in}(\mu)}{\partial \mu^2} \end{aligned} \quad (30)$$

DN assert that the last two terms on the right-hand side of this equation are of order  $O(\partial/\sigma_\mu^2)$ , the same order as the single term kept in the differential equation (17) derived from the Eulerian truncation. They suggest that this proves that the  $\bar{c}$  expansion (21) resulting from the Eulerian truncation is non-asymptotic.

[21] A different interpretation results if we note that (30) can be written as:

$$\frac{\partial \bar{c}(\mu, t)}{\partial t} = \frac{\partial}{\sigma_\mu^2} \int_0^t \frac{\partial^2 \bar{c}(\mu, t')}{\partial \mu^2} dt' + 2 \frac{\partial}{\sigma_\mu^2} \int_0^t \frac{\partial^2 [\bar{c}_1(\mu, t') - c_{in}(\mu)]}{\partial \mu^2} dt' \quad (31)$$

The bracketed expression inside the second integral on the right-hand side depends on  $\partial/\sigma_\mu^2$ . In fact, this expression goes to zero as  $\partial/\sigma_\mu^2$  goes to zero since  $\bar{c}_1$  approaches  $c_{in}$  in this case. If we replace  $[\bar{c}_1(\mu, t') - c_{in}(\mu)]$  with the integral term in (26) we obtain the following alternative differential equation for  $\bar{c}(\mu, t)$ :

$$\begin{aligned} \frac{\partial \bar{c}(\mu, t)}{\partial t} &= \frac{\partial}{\sigma_\mu^2} \int_0^t \frac{\partial^2 \bar{c}(\mu, t')}{\partial \mu^2} dt' \\ &+ 2\sigma_u^4 \int_0^t \int_0^{t'} \int_0^{t''} \frac{\partial^4 \bar{c}_1(\mu, t''')}{\partial \mu^4} dt''' dt'' dt' \end{aligned} \quad (32)$$

If we expand  $\bar{c}_1$  in the second term on the right-hand side in a Taylor series and carry out the temporal integrations we obtain:

$$\begin{aligned} \frac{\partial \bar{c}(\mu, t)}{\partial t} &= \frac{\partial}{\sigma_\mu^2} \int_0^t \frac{\partial^2 \bar{c}(\mu, t')}{\partial \mu^2} dt' + 2 \sum_{m=1}^{\infty} \frac{\sigma_u^{2m+2} t^{2m+1}}{(2m+1)!} \frac{\partial^{2m+2} c_{in}(\mu)}{\partial \mu^{2m+2}} \\ &= \frac{\partial}{\sigma_\mu^2} \int_0^t \frac{\partial^2 \bar{c}(\mu, t')}{\partial \mu^2} dt' + \frac{\sigma_u^4 t^3}{3} \frac{\partial^4 c_{in}(\mu)}{\partial \mu^4} \\ &+ \text{terms of order } \sigma_u^6 \text{ and higher} \end{aligned} \quad (33)$$

It is apparent from this analysis that the correction given by the last two terms on the right-hand side of (30) has a leading term proportional to  $\sigma_u^4$  rather than  $\frac{\partial}{\sigma_\mu}$ , in contrast to the analysis given in DN.

[22] Additional insight can be obtained by examining the explicit solution for the correction term  $\bar{c}_3$ . We start by noting that the same method used to derive (32) can be used to write (29) as:

$$\begin{aligned} \frac{\partial \bar{c}_3(\mu, t)}{\partial t} &= \frac{\partial}{\sigma_\mu^2} \int_0^t \frac{\partial^2 \bar{c}_3(\mu, t')}{\partial \mu^2} dt' + 2\sigma_u^4 \int_0^t \int_0^{t'} \int_0^{t''} \\ &\cdot \frac{\partial^4 \bar{c}_1(\mu, t''')}{\partial \mu^4} dt''' dt'' dt' \end{aligned} \quad (34)$$

If we differentiate (34) with respect to time we get a wave equation of the same form as (19), but with a forcing term that depends on the known function  $\bar{c}_1(\mu, t)$ :

$$\frac{\partial^2 \bar{c}_3(\mu, t)}{\partial t^2} = \frac{\partial}{\sigma_\mu^2} \frac{\partial^2 \bar{c}_3(\mu, t)}{\partial \mu^2} + 2\sigma_u^4 \int_0^t \int_0^{t'} \frac{\partial^4 \bar{c}_1(\mu, t'')}{\partial \mu^4} dt'' dt' \quad (35)$$

This equation has the following series solution:

$$\begin{aligned} \bar{c}_3(\mu, t) &= \sum_{m=1}^{\infty} \frac{2m}{(2m+2)!} \sigma_u^{2m+2} t^{2m+2} \frac{\partial^{2m} c_{in}(\mu)}{\partial \mu^{2m}} \\ &= \frac{\sigma_u^4 t^4}{12} \frac{\partial^4 c_{in}(\mu)}{\partial \mu^4} + \text{terms of order } \sigma_u^6 \text{ and higher} \end{aligned} \quad (36)$$

Note that the  $\bar{c}_3$  series is asymptotic, with a leading term of order  $\sigma_u^4$ .

[23] We can combine the series expressions for  $\bar{c}_1$  and  $\bar{c}_3$  to obtain a series expression for the higher-order mean approximation  $\bar{c} \approx \bar{c}_1 + \bar{c}_2 + \bar{c}_3 = \bar{c}_1 + \bar{c}_3$  considered in (30) and in DN equation (45):

$$\begin{aligned} \bar{c}(\mu, t) &\approx c_{in}(\mu) + \sum_{m=1}^{\infty} \frac{(2m-1)}{(2m)!} \sigma_u^{2m} t^{2m} \frac{\partial^{2m} c_{in}(\mu)}{\partial \mu^{2m}} \\ &= c_{in}(\mu) + \frac{\sigma_u^2 t^2}{2} \frac{\partial^2 c_{in}(\mu)}{\partial \mu^2} + \frac{\sigma_u^4 t^4}{8} \frac{\partial^4 c_{in}(\mu)}{\partial \mu^4} \\ &+ \text{terms of order } \sigma_u^6 \text{ and higher} \end{aligned} \quad (37)$$

This higher-order Eulerian truncation approximation to the concentration mean is an asymptotic series which matches the true solution  $\bar{c}$  of (10) through terms of order  $\sigma_u^4$ . At higher orders the approximate series has the same general form as the exact series but the constant coefficients differ.

[24] Comparing (37) with the lower-order mean approximation in (25), and both with (10) it appears that each iteration of the Eulerian expansion recursion yields an approximate series that matches the exact solution at one higher order. That is,  $\bar{c}_1$  matches  $\bar{c}$  through order  $\partial/\sigma_\mu^2$ ,  $\bar{c}_1 + \bar{c}_3$  matches  $\bar{c}$  through order  $\sigma_u^4$ , etc. This behavior is reasonable and consistent. There is no reason to conclude from an analysis of the relevant series expansions that the Eulerian truncation is any less accurate than alternative approximation methods. In fact, we have already seen that the Eulerian truncation gives results for the perfectly stratified problem

which are identical to the cumulant expansion method, which is also based on an asymptotic expansion. This is consistent with the requirement that asymptotic expansions be unique [van Dyke, 1964; Dagan and Neuman, 1991].

## 5. Discussion and Conclusions

[25] The analysis presented above clarifies some of the features of exact and approximate solutions to the perfectly stratified transport problem. First, an exact (pseudo-Fickian) solution to this problem can be obtained from a Lagrangian analysis based on a series expansion of the kinematic transformation relating fluid velocity to particle displacement [Dagan and Neuman, 1991]. Although the Lagrangian expansion is exact for this particular problem, it will generally not give exact results for other more complex problems. Second, the cumulant expansion and Eulerian truncation methods give identical approximate (convolutive non-Fickian) solutions to the perfectly stratified transport problem. These methods are based on series expansions of the transformation relating velocity to the time derivative of concentration (rather than particle displacement). Third, the mean concentration expansions generated by the cumulant expansion and Eulerian truncation methods are asymptotic, in the sense that the ratios of successive terms vanish as the velocity variance approaches zero.

[26] The question remains as to why the cumulant expansion and Eulerian truncation solutions to the perfectly stratified problem differs so significantly from the exact solution at large time. We believe that the poor large-time performance of these Eulerian approximations is a direct result of the zero local dispersion assumption. In the absence of local dispersion the plume in any given random realization retains the same shape forever, deviating more and more from the Gaussian ensemble mean. As a result, fluctuations of the actual concentration from the mean become large compared to the mean value (i.e. the coefficient of variation becomes unbounded) and the assumption that the product  $u'c'(\mu, t)$  is small breaks down.

[27] The analyses of the perfectly stratified problem presented here and in DN should be viewed in a larger context. A number of investigators have used numerical simulation methods to show that perturbation methods based on Eulerian truncation and similar approximations can work well in groundwater applications [e.g., Ababou et al., 1989; Tompson and Gelhar, 1990; Hassan et al., 1997, 1998]. But the validity of a linearized perturbation analysis appears to be problem-dependent [Van Lent and Kitanidis, 1996; Li and McLaughlin, 1991]. Kapoor and Gelhar [1994] stress the importance of local dispersion in stochastic analyses of solute transport and point out that it is local dispersion that causes the variance to dissipate for conservative solute transport. Their analysis confirms that the concentration coefficient of variation increases without bound if local dispersion is neglected. Consequently, as time increases the role of the terms truncated becomes increasingly important and the accuracy of the Eulerian small perturbation assumption deteriorates.

[28] Our analysis confirms that the Eulerian truncation approximation yields an asymptotic expansion which converges to the true mean, even under the somewhat unrealistic assumptions made in the non-dispersive perfectly stratified transport problem. But, as we have seen in this

example, the fact that an approximation has desirable asymptotic properties does not necessarily mean that it will be accurate in practice. The practical validity of the Lagrangian and Eulerian methods considered here is problem-dependent. For this reason, it is risky to generalize from particular examples.

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