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Abstract

The process of filtration of non-charged, submicron particles is analyzed using the method of volume averaging. The particle continuity equation is represented in terms of the *first correction* to the Smoluchowski equation that takes into account particle inertia effects for small Stokes numbers. This leads to a cellular efficiency that contains a minimum in the efficiency as a function of the particle size, and this allows us to identify the *most penetrating particle size*. Comparison of the theory with results from Brownian dynamics indicates that the first correction to the Smoluchowski equation gives reasonable results in terms of both the cellular efficiency and the most penetrating particle size. However, the results for larger particles clearly indicate the need to extend the Smoluchowski equation to include higher order corrections. Comparison of the theory with laboratory experiments, in the absence of adjustable parameters, provides interesting agreement for particle diameters that are equal to or less than the diameter of the most penetrating particle.

1. **Introduction**

The process of filtration takes place in an hierarchical porous media (Cushman, 1990) and we have illustrated this in Figure 1. In order to design a filter, one needs a particle transport equation in which the porosity heterogeneities have been *spatially smoothed*. This suggests the use of the first averaging volume shown in Figure 1 along with the method of large-scale averaging (Quintard and Whitaker, 1987, 1988, 1990; Plumb and Whitaker, 1988, 1990). Large-scale averaging requires the use of local volume averaged equations that are associated with the second averaging volume shown in Figure 1. These equations are sometimes referred to as the *Darcy-scale* transport equations and they represent the point in the hierarchical process at which the governing differential equations and boundary conditions are *joined*. These boundary conditions are imposed at the γ-σ interface which is illustrated in the third volume contained in Figure 1 where we have identified the fibers as the σ-phase and the fluid as the γ-phase. The

Figure 1
Hierarchical View of the Filtration Process

governing equation for the fluid velocity in the γ-phase will be taken to be Stokes' equations, while the governing equation for the particle concentration is represented by a Fokker-Planck equation for the probability density function. This idea is suggested in the last volume illustrated in Figure 1 where we have identified the particles as the κ-phase and the pure fluid as the β-phase. The fact that we are going to use Stokes' equations to described the velocity of the γ-phase indicates that the volume fraction of the particles is much, much less than one (Russell, 1981).

Macroscopic transport equations for filtration are often introduced heuristically in the form of a convective-dispersion equation with a source term accounting for the particle deposition. This source term requires knowledge of the *filter collection efficiency* that can be determined by experiments. The starting point for a theoretical derivation of the filter collection efficiency is a pore-scale description of the particle transport which must be subjected to both local volume averaging and large-scale averaging in order to obtain a filter transport equation. The particle transport equation must account for the various mechanisms that affect the particle deposition process such as Brownian diffusion, inertial deposition, electrostatic effects, etc. Results published in the literature [see Ramarao and Tien (1991) for an extensive review] can be classified according to the various assumptions made in describing the particle transport, as well as the methodology used in proceeding from the pore-scale equations to the macroscopic description.

If one assumes pure Brownian diffusion, particle transport can be viewed as equivalent to the process of convective-diffusion in a porous medium with reaction (Friedlander, 1977; Shapiro and Brenner, 1990). However, it has been recognized for some time that the existence of a *most penetrating particle size* is the result of a complex interaction between Brownian diffusion and inertial effects. Because of this, it is important to make use of a more complete description of the particle transport process that accounts for particle inertia effects.

Several approximate methods have been proposed to determine the particle velocity field. Deterministic particle trajectory calculations can be used in the convective-diffusion equation (Fuchs, 1964; Yuu and Jota, 1978; Fernández de la Mora and Rosner, 1981). More precise solutions of this stochastic process can be obtained by a continuous approach (de la Mora and Rosner, 1982; Yeh and Liu 1974; Banks and Kurowski 1983) which will be described later in this paper. More recently, direct solutions of the Langevin equations (the so-called Brownian dynamics calculations) have been used to simulate particle motions around single fibers (Kanaoka et al., 1983; Gupta and Peters. 1985, 1986; Ramarao and Tien, 1992, 1994).

Brownian dynamics represents the most reliable method of analyzing the particle transport process since the physics can be incorporated directly into the calculations for a single fiber (or multi-fiber) efficiency. However, the design of a filter requires an analysis that faithfully transmits the physics from the particle scale illustrated in Figure 1 to the filter scale, and this is difficult to accomplish via Brownian dynamics. This suggests the development of continuum equations that can accurately simulate the phenomena described by Brownian dynamics. Our approach will be to derive both the macroscopic equation and the macroscopic coefficients from the pore-scale description. This approach has been used successfully in dealing with several problems of transport phenomena in porous media. In particular, valuable results have been obtained for the process of passive dispersion (Lee, 1979; Brenner. 1980; Carbonell and Whitaker, 1983; Eidsath et al., 1983; Rubinstein and Mauri 1986), active dispersion (Zanotti and Carbonell, 1994; Quintard and Whitaker. 1994a), and dispersion with chemical reaction (Mauri, 1989; Edwards et al., 1993). In this paper, the macroscopic forms of the pore-scale equations are obtained by the method of volume averaging and effective transport properties are determined by two independent closure problems that are solved for periodic arrays of cylinders.

PARTICLE MOTION

Our description of the motion of the particles begins with the single particle Langevin equation

$$m_p \frac{d\mathbf{v}_p}{dt} = -\frac{3\pi\mu d_p}{c_s}(\mathbf{v}_p - \mathbf{v}) + \mathbf{F}_r(t) \tag{1.1}$$

in which m_p is the mass of the particle and $\mathbf{v}_p$ is the velocity of the particle. The first term on the right hand side of Eq. 1.1 describes the Stokes' drag on a single isolated particle with c_s representing the Cunningham correction factor (Tien, 1989). The use of this form for the force indicates that we are ignoring particle-particle interactions and the fluid mechanical complications that arise when a particle approaches a solid surface (Peters and Ying, 1991). These effects should certainly be taken into account; however, in this initial study it is our attention to keep the analysis as simple as possible.

It is convenient to express Eq. 1.1 as

$$\frac{d\mathbf{v}_p}{dt} = -\gamma(\mathbf{v}_p - \mathbf{v}) + \Gamma(t) \tag{1.2}$$

in which γ is inversely proportional to the Stokes' number

$$\gamma = \frac{3\pi\mu d_p}{m_p c_s} \sim St^{-1} \tag{1.3}$$

Here St represents the Stokes' number to be defined later. There are many particle transport processes for which the Stokes' number is small compared to one and this so-called *high friction limit* represents an important case for filtration. The Fokker-Planck equation associated with the stochastic process described by Eq. 1.2 takes the form (Risken, Sec. 10.1, 1989)

$$\frac{\partial W_p}{\partial t} + \frac{\partial}{\partial \mathbf{r}}\left(\mathbf{v}_p W_p\right) - \frac{\partial}{\partial \mathbf{v}_p}\left[\gamma(\mathbf{v}_p - \mathbf{v})\right] = \left(\frac{\gamma\, k\, T}{m_p}\right)\frac{\partial}{\partial \mathbf{v}_p}\cdot\frac{\partial}{\partial \mathbf{v}_p}\left(W_p\right) \tag{1.4}$$

and this result is often known as Kramer's (1940) equation. The dependent variable represents a probability density function

$$W_p = W_p(t,\mathbf{r},\mathbf{v}_p) \tag{1.5}$$

and the particle number density is given by

$$n_p = \int_{-\infty}^{+\infty} W_p(t,\mathbf{r},\mathbf{v}_p)\, d\mathbf{v}_p \tag{1.6}$$

The solution of Eq. 1.4 for the high friction limit is based on matrix continued fraction methods (Risken, Sec 10.4, 1989) which yield

$$\frac{\partial n_p}{\partial t} = \nabla\cdot\left\{\left[-\mathbf{v} + D_p\nabla\right]n_p\right\} + \gamma^{-1}\,\nabla\cdot\left\{\left[\mathbf{v}\cdot\nabla\mathbf{v} - (\nabla\cdot\mathbf{v})D_p\nabla\right]n_p\right\} + \tag{1.7}$$

$$+ \gamma^{-3}\,\nabla\cdot\left\{[\ldots\ldots.]n_p\right\} + \mathbf{O}(\gamma^{-5}) + \ldots\ldots$$

in which D_p is the Brownian diffusivity. It is important to keep in mind that this result *does not take into account* the complex fluid mechanics that occur when a particle approaches a solid surface. The first two terms on the right hand side of Eq. 1.7 were obtained by Titulaer (1978) and by Skinner and Wolynes (1979) using different methods. If only the first term on the right hand side of Eq. 1.7 is retained we have the Smoluchowski equation (Gardiner, Sec. 6.4, 1985) and under these circumstances the mean motion of the particles follows the fluid streamlines. As we shall see in Sec. 3, this leads to a situation that can not predict a crucial characteristic of many filtration processes, thus we will retain the second term on the right hand side of Eq. 1.7 so that our particle transport equation takes the form

$$\frac{\partial n_p}{\partial t} + \nabla\cdot\left\{\left[\mathbf{v} - \gamma^{-1}\mathbf{v}\cdot\nabla\mathbf{v}\right]n_p\right\} = \nabla\cdot(D_p\nabla n_p), \quad \textit{in the } \gamma\textit{-phase} \tag{1.8}$$

This correction to the Smoluchowski equation has been used by Yeh and Liu (1974), Banks and Kurowski (1983), and others for the study of particle transport in the filtration process, and derivations are available from de la Mora and Rosner (1981, 1982) and from

Peters and Ying (1991) in addition to the references cited above. Equation 1.8 represents the first smoothing process in the hierarchy of averaging processes illustrated in Figure 1. The next step in this process requires that we form the local volume average of Eq. 1.8.

2. **Volume Averaging**

At this point we are ready to express the complete transport problem under consideration, and we list the governing differential equations and boundary conditions as

$$\frac{\partial n_p}{\partial t} + \nabla\cdot\left\{\left[\mathbf{v} - \gamma^{-1}\mathbf{v}\cdot\nabla\mathbf{v}\right]n_p\right\} = \nabla\cdot(D_p\nabla n_p) \tag{2.1}$$

B.C. 1 $$n_p = 0, \quad \textit{at the } \gamma\text{-}\sigma \textit{ interface} \tag{2.2}$$

$$0 = -\nabla p + \rho\mathbf{g} + \mu\nabla^2\mathbf{v} \tag{2.3}$$

B.C. 2 $$\mathbf{v} = 0, \quad \textit{at the } \gamma\text{-}\sigma \textit{ interface} \tag{2.4}$$

$$\nabla\cdot\mathbf{v} = 0 \tag{2.5}$$

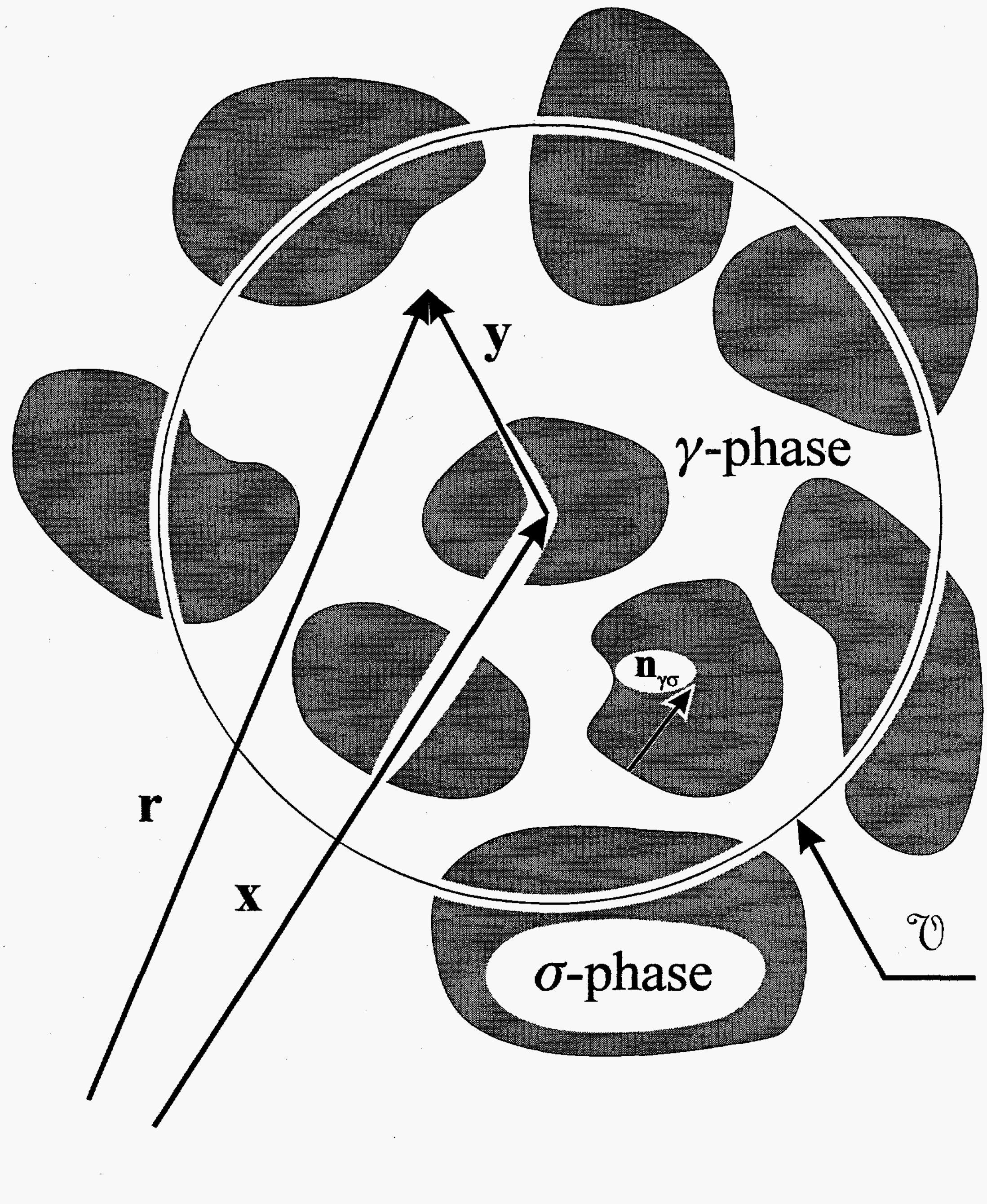
It should be clear that we have already used Eq. 2.5 with Eq. 1.7 in order to simplify that result to Eq. 1.8, and we will need to use Eq. 2.5 again in our analysis of the convective transport term in Eq. 2.1. The boundary condition represented by Eq. 2.2 must be thought of as a limiting case which will create an upperbound for the filtration efficiency. This boundary condition has been used by Ruckenstein and Prieve (1973), Shapiro and Brenner (1990), and others.

The method of volume averaging (Anderson and Jackson, 1967; Marle, 1967, Slattery, 1967; Whitaker, 1967) begins by associating with every point in space (in both the γ-phase and the σ-phase) an averaging volume that we denote by . Such a volume is illustrated in Figure 2 where we have located the centroid of the averaging volume by the position vector $\mathbf{x}$, the radius of the averaging volume by r_0, and the characteristic length of the γ-phase by ℓ_γ. We will make use of two averages in our analysis of Eq. 2.1 and the first of these is the *superficial* volume average which can be expressed as

$$\langle\psi_\gamma\rangle = \frac{1}{\mathscr{V}}\int_{V_\gamma}\psi_\gamma\, dV \tag{2.6}$$

Here ψ_γ is any function associated with the γ-phase and V_γ is the volume of the γ-phase contained within the averaging volume, . In addition to the superficial average, we will also make use of the *intrinsic* volume average that is defined by

Figure 2
Positions Vectors Associated with the Averaging Volume



$$\langle\psi_\gamma\rangle^\gamma \;=\; \frac{1}{V_\gamma}\int_{V_\gamma}\psi_\gamma dV \qquad (2.7)$$

These two averages are related by

$$\langle\psi_\gamma\rangle \;=\; \varepsilon_\gamma\langle\psi_\gamma\rangle^\gamma \qquad (2.8)$$

The average velocity is often represented in terms of the *superficial* velocity, while the average particle concentration is typically represented in terms of an *intrinsic* average. To avoid confusion between these two averages we will always make use of the nomenclature indicated in Eqs. 2.6 through 2.8.

When we form the volume average of Eq. 2.1 we will encounter averages of gradients and we will need to convert these to gradients of averages by means of the spatial averaging theorem (Howes and Whitaker, 1985) which we represent as

$$\langle\nabla\psi_\gamma\rangle \;=\; \nabla\langle\psi_\gamma\rangle + \frac{1}{\mathscr{V}}\int_{A_{\gamma\sigma}}\mathbf{n}_{\gamma\sigma}\psi_\gamma dA \qquad (2.8)$$

We begin the analysis of the particle transport process by expressing the superficial average of Eq. 2.1 as

$$\left\langle\frac{\partial n_p}{\partial t}\right\rangle + \left\langle\nabla\cdot\left\{[\mathbf{v}-\gamma^{-1}\mathbf{v}\cdot\nabla\mathbf{v}]n_p\right\}\right\rangle \;=\; \left\langle\nabla\cdot D_p\nabla n_p\right\rangle \qquad (2.9)$$

and note that the first term can be written as

$$\left\langle\frac{\partial n_p}{\partial t}\right\rangle \;=\; \frac{\partial\langle n_p\rangle}{\partial t} \qquad (2.10)$$

since V_γ is independent of time. The convective transport term in Eq. 2.10 requires the use of the averaging theorem which leads to

$$\left\langle\nabla\cdot[\mathbf{v}-\gamma^{-1}\mathbf{v}\cdot\nabla\mathbf{v}]n_p\right\rangle \;=\; \nabla\cdot\langle[\mathbf{v}-\gamma^{-1}\mathbf{v}\cdot\nabla\mathbf{v}]n_p\rangle \qquad (2.11)$$

on the basis of the no-slip condition given by Eq. 2.4. The diffusive term on the right hand side of Eq. 2.10 provides us with

$$\langle \nabla \cdot D_p \nabla n_p \rangle = \nabla \cdot \langle D_p \nabla n_p \rangle + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla n_p dA \tag{2.12}$$

Use of Eqs. 2.11 through 2.13 along with Eq. 2.8 in the form

$$\langle n_p \rangle = \varepsilon_\gamma \langle n_p \rangle^\gamma \tag{2.13}$$

allows us to express the superficial average particle transport equation as

$$\underbrace{\varepsilon_\gamma \frac{\partial \langle n_p \rangle^\gamma}{\partial t}}_{accumulation} + \underbrace{\nabla \cdot \langle \mathbf{v} n_p \rangle}_{convection} - \underbrace{\gamma^{-1} \nabla \cdot \langle [\mathbf{v} \cdot \nabla \mathbf{v}] n_p \rangle}_{\substack{particle \\ inertia\ convection}}$$

$$\underbrace{\nabla \cdot \left\{ D_p \left[\nabla \langle n_p \rangle + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} n_p dA \right] \right\}}_{diffusion} + \underbrace{\frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla n_p dA}_{particle\ capture} \tag{2.15}$$

Here we have identified the correction to the Smoluchowski equation as the *particle inertia convection*, and it is this term that plays a key role in the determination of the "most penetrating particle size" in the filtration process.

In order to eliminate the point value of the particle concentration in the diffusion term, we use the following decomposition (Gray, 1975) for the particle density

$$n_p = \langle n_p \rangle^\gamma + \tilde{n}_p \tag{2.14}$$

and later we will use the velocity decomposition given by

$$\mathbf{v} = \langle \mathbf{v} \rangle^\gamma + \tilde{\mathbf{v}} \tag{2.17}$$

One can use Eq. 2.16 in the diffusion term and follow the type of analysis given by Whitaker (Sec. 2, 1986a) or Quintard and Whitaker (Sec. II, 1993) to obtain

$$\nabla \cdot \left\{ D_p \left[\nabla \langle n_p \rangle + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} n_p dA \right] \right\} = \nabla \cdot \left\{ D_p \left[\nabla \langle n_p \rangle^\gamma + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \tilde{n}_p dA \right] \right\} \tag{2.18}$$

This development makes use of the lemma extracted from Eq. 2.9

$$\nabla \varepsilon_\gamma = -\frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} dA \tag{2.19}$$

along with the two length-scale constraints given by

$$\ell_\gamma \ll r_o \tag{2.20}$$

$$\frac{r_o^2}{L^2} \ll 1 \tag{2.21}$$

Here one should think of L as the smallest characteristic length associated with a macroscopic quantity such as ε_γ, $\langle n_p \rangle^\gamma$, $\nabla \langle n_p \rangle^\gamma$, etc. These length-scale constraints were originally developed by Carbonell and Whitaker (Sec. 2, 1984) and a more thorough discussion is available in the more recent work of Quintard and Whitaker (1994b). One can also use the decomposition given by Eq. 2.16 in order to express the particle capture term as

$$\frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla n_p dA = -(\nabla \varepsilon_\gamma) \cdot D_p \nabla \langle n_p \rangle^\gamma + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p dA \tag{2.22}$$

Use of Eqs. 2.17 and 2.21 in Eq. 2.15 leads to

$$\varepsilon_\gamma \frac{\partial \langle n_p \rangle^\gamma}{\partial t} + \nabla \cdot \langle \mathbf{v} n_p \rangle - \gamma^{-1} \nabla \cdot \langle [\mathbf{v} \cdot \nabla \mathbf{v}] n_p \rangle = \tag{2.23}$$

$$\nabla \cdot \left\{ \varepsilon_\gamma D_p \left[\nabla \langle n_p \rangle^\gamma + \frac{1}{V_\gamma} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \tilde{n}_p dA \right] \right\} - (\nabla \varepsilon_\gamma) \cdot D_p \nabla \langle n_p \rangle^\gamma + \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p dA$$

and in order to complete the averaging procedure we would like to express the convective transport in terms of $\langle n_p \rangle^\gamma$ and $\tilde{n}_p$. One can follow Carbonell and Whitaker (1983, 1984) in order to represent the traditional convective transport as

$$\langle \mathbf{v} n_p \rangle = \underbrace{\varepsilon_\gamma \langle \mathbf{v} \rangle^\gamma \langle n_p \rangle^\gamma}_{\substack{\textit{traditional} \\ \textit{convective transport}}} + \underbrace{\langle \tilde{\mathbf{v}} \tilde{n}_p \rangle}_{\substack{\textit{traditional} \\ \textit{dispersive transport}}} \tag{2.24}$$

The so-called inertial contribution to the convective transport is algebraically more complex, and it is convenient to use only the decomposition given by Eq. 2.16 to obtain

$$\langle(\mathbf{v}\cdot\nabla\mathbf{v})n_p\rangle = \underbrace{\langle\mathbf{v}\cdot\nabla\mathbf{v}\rangle\langle n_p\rangle^\gamma}_{\substack{\textit{particle}\\ \textit{inertia convection}}} + \underbrace{\langle(\mathbf{v}\cdot\nabla\mathbf{v})\tilde{n}_p\rangle}_{\substack{\textit{particle}\\ \textit{inertia dispersion}}} \tag{2.25}$$

On the basis of Eqs. 2.24 and 2.25 we can express the volume averaged particle transport equation as

$$\underbrace{\varepsilon_\gamma \frac{\partial\langle n_p\rangle^\gamma}{\partial t}}_{\textit{accumulation}} + \nabla\cdot\underbrace{\left(\varepsilon_\gamma\langle\mathbf{v}\rangle^\gamma\langle n_p\rangle^\gamma - \gamma^{-1}\langle\mathbf{v}\cdot\nabla\mathbf{v}\rangle\langle n_p\rangle^\gamma\right)}_{\textit{convection}} + \nabla\cdot\underbrace{\left(\langle\tilde{\mathbf{v}}\tilde{n}_p\rangle - \gamma^{-1}\langle(\mathbf{v}\cdot\nabla\mathbf{v})\tilde{n}_p\rangle\right)}_{\textit{dispersion}}$$

$$= \underbrace{\nabla\cdot\left\{\varepsilon_\gamma D_p\left[\nabla\langle n_p\rangle^\gamma + \frac{1}{V_\gamma}\int_{A_{\gamma\sigma}}\mathbf{n}_{\gamma\sigma}\tilde{n}_p\,dA\right]\right\}}_{\textit{diffusion}} + \underbrace{\frac{1}{\mathscr{V}}\int_{A_{\gamma\sigma}}\mathbf{n}_{\gamma\sigma}\cdot D_p\nabla\tilde{n}_p\,dA}_{\textit{particle capture}} \tag{2.26}$$

Here we have imposed the simplification

$$(\nabla\varepsilon_\gamma)\cdot D_p\nabla\langle n_p\rangle^\gamma \ll \frac{1}{\mathscr{V}}\int_{A_{\gamma\sigma}}\mathbf{n}_{\gamma\sigma}\cdot D_p\nabla\tilde{n}_p\,dA \tag{2.27}$$

and the length-scale constraint associated with this restriction is given by

$$\frac{\ell_\gamma^3\ell_{bl}}{\ell_\sigma^2 L_\varepsilon L_n} \ll 1 \tag{2.28}$$

The derivation of this result requires an estimate of $\tilde{n}_p$ and definitions of the various length scales, and the analysis is given in Appendix A.

Before moving on to the closure problem we should list the volume average forms of the Stokes' equation given by Eq. 2.3 and the continuity equation that was presented earlier as Eq. 2.5. The volume averaged forms of these two equations have been developed in detail elsewhere (Whitaker, 1986b; Quintard and Whitaker, 1994b) and we simple list the volume averaged form of Eq. 2.3 as

$$\langle \mathbf{v} \rangle = -\frac{\mathbf{K}}{\mu} \cdot \left(\nabla \langle p \rangle^{\gamma} - \rho \mathbf{g} \right), \quad \textit{Darcy's law} \tag{2.29}$$

in which $\langle \mathbf{v} \rangle$ represents the *superficial* volume averaged velocity and $\langle p \rangle^{\gamma}$ represents the *intrinsic* volume averaged pressure. The volume averaged continuity equation can be expressed either in terms of the superficial average velocity

$$\nabla \cdot \langle \mathbf{v} \rangle = 0 \tag{2.30}$$

or in terms of the intrinsic average velocity

$$\nabla \cdot (\varepsilon_{\gamma} \langle \mathbf{v} \rangle^{\gamma}) = 0 \tag{2.31}$$

Here we have made use of the nomenclature given by Eqs. 2.7 and 2.8 and the relation between the two averages indicated by Eq. 2.9.

In order to obtain a closed form of Eq. 2.26, we need to develop the boundary value problem for $\tilde{n}_p$ and we need to show how $\langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle$ can be determined by Darcy's law. In the *general* analysis of the filtration process we definitely need to take porosity variations into account via the method of large-scale averaging (Quintard and Whitaker, 1987); however, in the development of the closure problem it is permissible to consider a local homogeneous region in which variations of ε_{γ} can be ignored. Under these circumstances we can divide Eq. 2.26 by ε_{γ} to obtain

$$\frac{\partial \langle n_p \rangle^{\gamma}}{\partial t} + \nabla \cdot \left(\langle \mathbf{v} \rangle^{\gamma} \langle n_p \rangle^{\gamma} - \gamma^{-1} \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^{\gamma} \langle n_p \rangle^{\gamma} \right) + \nabla \cdot \left(\langle \tilde{\mathbf{v}} \tilde{n}_p \rangle^{\gamma} - \gamma^{-1} \langle (\mathbf{v} \cdot \nabla \mathbf{v}) \tilde{n}_p \rangle^{\gamma} \right)$$

$$= \nabla \cdot \left\{ D_p \left[\nabla \langle n_p \rangle^{\gamma} + \frac{1}{V_{\gamma}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \tilde{n}_p \, dA \right] \right\} + \frac{\varepsilon_{\gamma}^{-1}}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p \, dA \tag{2.32}$$

With this intrinsic form of the particle transport equation we are ready to begin the derivation of the closure problem.

3. Closure Problem

In order to develop the governing differential equation for $\tilde{n}_p$ we recall Eq. 1.8

$$\frac{\partial n_p}{\partial t} + \nabla \cdot \left\{ \left[\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right] n_p \right\} = \nabla \cdot (D_p \nabla n_p) \tag{3.1}$$

and remember Eq. 2.16 so that Eq. 2.32 can be subtracted from Eq. 3.1 to obtain

$$\frac{\partial \tilde{n}_p}{\partial t} + \nabla \cdot \left\{ \left[\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right] n_p - \left[\langle \mathbf{v} \rangle^\gamma - \gamma^{-1} \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma \right] \langle n_p \rangle^\gamma \right\}$$

$$- \nabla \cdot \langle \tilde{\mathbf{v}} \tilde{n}_p \rangle^\gamma + \gamma^{-1} \nabla \cdot \langle (\mathbf{v} \cdot \nabla \mathbf{v}) \tilde{n}_p \rangle^\gamma = \nabla \cdot (D_p \nabla \tilde{n}_p) -$$

$$- \nabla \cdot \left[\frac{D_p}{V_\gamma} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \tilde{n}_p dA \right] - \frac{\varepsilon_\gamma^{-1}}{\mathcal{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p dA \tag{3.2}$$

The second and third terms in this result can be arranged as

$$\left(\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right) n_p - \left(\langle \mathbf{v} \rangle^\gamma - \gamma^{-1} \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma \right) \langle n_p \rangle^\gamma =$$

$$\left(\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right) \tilde{n}_p - \left[\left(\langle \mathbf{v} \rangle^\gamma - \mathbf{v} \right) - \gamma^{-1} \left(\langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma - \mathbf{v} \cdot \nabla \mathbf{v} \right) \right] \langle n_p \rangle^\gamma \tag{3.3}$$

If we neglect variations of the porosity in the closure problem, we can use the various forms of the continuity equation to obtain

$$\nabla \cdot \left(\langle \mathbf{v} \rangle^\gamma - \mathbf{v} \right) = 0 \tag{3.4}$$

and this allows us to substitute Eq. 3.3 into Eq. 3.2 and obtain the following transport equation for $\tilde{n}_p$

$$\frac{\partial \tilde{n}_p}{\partial t} + \nabla \cdot \left[\left(\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right) \tilde{n}_p \right] + \underbrace{\left[\tilde{\mathbf{v}} - \gamma^{-1} \left(\mathbf{v} \cdot \nabla \mathbf{v} - \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma \right) \right] \cdot \nabla \langle n_p \rangle^\gamma}_{source} - \underbrace{\left\{ \nabla \cdot \left[\gamma^{-1} \left(\mathbf{v} \cdot \nabla \mathbf{v} - \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma \right) \right] \right\} \langle n_p \rangle^\gamma}_{source}$$

$$- \underbrace{\nabla \cdot \langle \tilde{\mathbf{v}} \tilde{n}_p \rangle^\gamma + \gamma^{-1} \nabla \cdot \langle (\mathbf{v} \cdot \nabla \mathbf{v}) \tilde{n}_p \rangle^\gamma}_{\text{non-local convective transport}} = \nabla \cdot \left(D_p \nabla \tilde{n}_p \right) - \underbrace{\nabla \cdot \left[\frac{D_p}{V_\gamma} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \tilde{n}_p dA \right]}_{\text{non-local diffusive transport}}$$

$$- \underbrace{\frac{\varepsilon_\gamma^{-1}}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p dA}_{particle\ capture} \tag{3.5}$$

Here we see terms representing

1. The classic effects of accumulation, local convection, and local diffusion.
2. Non-local convection and non-local diffusion.
3. Sources proportional to $\nabla\langle n_p\rangle^\gamma$ and $\langle n_p\rangle^\gamma$.
4. Particle capture.

We use the word *non-local* to describe those terms which involve integrals of $\tilde{n}_p$, and one can draw upon previous studies (Carbonell and Whitaker, 1984; Whitaker, 1986a; Quintard and Whitaker, 1993; Quintard and Whitaker, 1994a) to argue that these terms are negligible when length-scale constraints such as those indicated by Eqs. 2.20 and 2.21 are valid. The analysis consists of comparing the non-local terms with the associated local terms and demonstrating that the former are smaller than the latter by a factor of ℓ / L. This occurs because the non-local terms involve the derivatives of *average quantities* while the local terms always contain the derivatives of *point quantities*. Because ℓ / L is always small compared to one, the non-local terms can be neglected and Eq. 3.5 simplifies to

$$\frac{\partial \tilde{n}_p}{\partial t} + \nabla \cdot \left[\left(\mathbf{v} - \gamma^{-1}\mathbf{v} \cdot \nabla \mathbf{v}\right)\tilde{n}_p\right] + \underbrace{\left[\tilde{\mathbf{v}} - \gamma^{-1}(\mathbf{v} \cdot \nabla \mathbf{v} - \langle \mathbf{v} \cdot \nabla \mathbf{v}\rangle)\right] \cdot \nabla\langle n_p\rangle^\gamma}_{source} - \underbrace{\left\{\nabla \cdot \left[\gamma^{-1}\left(\mathbf{v} \cdot \nabla \mathbf{v} - \langle \mathbf{v} \cdot \nabla \mathbf{v}\rangle^\gamma\right)\right]\right\}\langle n_p\rangle^\gamma}_{source}$$

$$= \nabla \cdot \left(D_p \nabla \tilde{n}_p\right) - \frac{\varepsilon_\gamma^{-1}}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p dA \tag{3.6}$$

The last term in this result is also a non-local term; however, it is not negligible since it represents the rate at which particles are captured per unit volume.

Use of the boundary condition given by Eq. 2.2 and the decomposition represented by Eq. 2.16 leads to the following boundary condition

$$\text{B.C. 1} \qquad \tilde{n}_p = -\underbrace{\langle n_p\rangle^\gamma}_{source}, \quad \textit{at the } \gamma\text{-}\sigma \textit{ interface} \tag{3.7}$$

and in order to determine the $\tilde{n}_p$-field in some local, representative region, we are forced to accept the spatially periodic model of a porous medium and impose the following periodicity condition.

Periodicity: $$\tilde{n}_p(\mathbf{r}+\ell_i) = \tilde{n}_p(\mathbf{r}), \qquad i = 1,2,3 \tag{3.8}$$

In addition, when the length-scale constraints indicated by Eqs. 2.20 and 2.21 are valid, Carbonell and Whitaker (1984) have shown that the average of the spatial deviation can be set equal to zero and we express this idea as

Average: $$\langle \tilde{n}_p \rangle^\gamma = 0 \tag{3.9}$$

Strictly speaking, we need an initial condition for $\tilde{n}_p$ to complete our problem statement; however, both the volume averaged equation given by Eq. 2.26 and the closure equation represented by Eq. 3.6 can be treated as quasi-steady, thus the initial condition for both $\langle n_p \rangle^\gamma$ and $\tilde{n}_p$ can be ignored. This means that the closure problem takes the quasi-steady form given by

QUASI-STEADY CLOSURE PROBLEM

$$\nabla \cdot \left[\left(\mathbf{v} - \gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right) \tilde{n}_p \right] + \underbrace{\left[\tilde{\mathbf{v}} - \gamma^{-1} \left(\mathbf{v} \cdot \nabla \mathbf{v} - \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle \right) \right] \cdot \nabla \langle n_p \rangle^\gamma}_{source} - \underbrace{\left\{ \nabla \cdot \left[\gamma^{-1} \left(\mathbf{v} \cdot \nabla \mathbf{v} - \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma \right) \right] \right\} \langle n_p \rangle^\gamma}_{source}$$

$$= \nabla \cdot \left(D_p \nabla \tilde{n}_p \right) - \frac{\varepsilon_\gamma^{-1}}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p \, dA \tag{3.10a}$$

B.C. 1 $$\tilde{n}_p = -\underbrace{\langle n_p \rangle^\gamma}_{source}, \quad \textit{at the } \gamma\text{-}\sigma \textit{ interface} \tag{3.10b}$$

Periodicity: $$\tilde{n}_p(\mathbf{r}+\ell_i) = \tilde{n}_p(\mathbf{r}), \qquad i = 1,2,3 \tag{3.10c}$$

Average: $$\langle \tilde{n}_p \rangle^\gamma = 0 \tag{3.10d}$$

The form of this boundary value problem suggests a representation for $\tilde{n}_p$ given by

$$\tilde{n}_p = \mathbf{b} \cdot \nabla \langle n_p \rangle^\gamma - s \langle n_p \rangle^\gamma \tag{3.11}$$

in which $\mathbf{b}$ and s are referred to as the *closure variables* or the *mapping variables* since they map the sources onto the spatial deviation concentration. One can draw upon a series of studies associated with the closure problem (Eidsath et al., 1983; Nozad et al., 1985; Crapiste et al., 1986; Ochoa et al, 1986; Quintard and Whitaker, 1993 and 1994a) to conclude that the vector $\mathbf{b}$ and the scalar s are determined by two boundary value problems that are analogous to the problem given by Eqs. 3.10. The first of these problems determines the vector, $\mathbf{b}$, and this problem is given by

PROBLEM I

$$\nabla\cdot\left[\left(\mathbf{v}-\gamma^{-1}\mathbf{v}\cdot\nabla\mathbf{v}\right)\mathbf{b}\right]+\left[\tilde{\mathbf{v}}-\gamma^{-1}\left(\mathbf{v}\cdot\nabla\mathbf{v}-\langle\mathbf{v}\cdot\nabla\mathbf{v}\rangle^{\gamma}\right)\right] =$$

$$\nabla\cdot\left(D_p\nabla\mathbf{b}\right)+\frac{\varepsilon_\gamma^{-1}}{\mathscr{V}}\int_{A_{\gamma\sigma}}\mathbf{n}_{\gamma\sigma}\cdot D_p\nabla\mathbf{b}dA \tag{3.12a}$$

B.C. 1 $\quad \mathbf{b} = 0, \quad$ *at the* γ *-* σ *interface* (3.12b)

Periodicity: $\quad \mathbf{b}(\mathbf{r}+\ell_i) = \mathbf{b}(\mathbf{r}), \quad i = 1,2,3$ (3.12c)

Average: $\quad \langle\mathbf{b}\rangle^{\gamma} = 0$ (3.12d)

For the process of filtration, the second problem is much more important than the first since it is used to determine the capture coefficient, k_{eff}. This problem is given by

PROBLEM II

$$\nabla\cdot\left[\left(\mathbf{v}-\gamma^{-1}\mathbf{v}\cdot\nabla\mathbf{v}\right)s\right] + \nabla\cdot\left[\gamma^{-1}\left(\mathbf{v}\cdot\nabla\mathbf{v}-\langle\mathbf{v}\cdot\nabla\mathbf{v}\rangle^{\gamma}\right)\right] =$$

$$= \nabla\cdot\left(D_p\nabla s\right) - \frac{\varepsilon_\gamma^{-1}}{\mathscr{V}}\int_{A_{\gamma\sigma}}\mathbf{n}_{\gamma\sigma}\cdot D_p\nabla s dA. \tag{3.13a}$$

B.C. 1 $\quad s = 1, \quad$ *at the* γ *-* σ *interface* (3.13b)

Periodicity: $\quad s(\mathbf{r}+\ell_i) = s(\mathbf{r}), \quad i=1,2,3$ (3.13c)

Average: $\quad \langle s\rangle^{\gamma} = 0$ (3.13d)

These two closure problems can be solved using the numerical methods described by Quintard and Whitaker (1993), and their solution allows us to determine the mapping variables for the *spatial deviation* particle concentration represented by Eq. 3.11.

Substitution of Eq. 3.11 into the volume averaged transport equation given by Eq. 2.26 leads to the closed form of that equation which contains effective transport and capture coefficients which are determined by Problems I and II.

CLOSED FORM

The closed form of Eq. 2.26 can be expressed as

$$\varepsilon_\gamma \frac{\partial \langle n_p \rangle^\gamma}{\partial t} + \nabla \cdot \left[\left(\langle \mathbf{v} \rangle - \gamma^{-1} \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle \right) \langle n_p \rangle^\gamma \right] - (\mathbf{d} + \mathbf{u}) \cdot \nabla \langle n_p \rangle^\gamma \quad = \quad \nabla \cdot \left(\mathbf{D}^* \cdot \nabla \langle n_p \rangle^\gamma \right) - k_{eff} \langle n_p \rangle^\gamma \tag{3.14}$$

and it is important to recognize that this is a *superficial average* transport equation. This means that each term represents a certain quantity *per unit volume of the porous medium* and not per unit volume of the fluid phase. This is obvious for the accumulation term; however, it can be confusing for the particle capture term. The various coefficients that appear in Eq. 3.14 are defined as

$$\mathbf{u} \quad = \quad \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \mathbf{b} \, dA \tag{3.15}$$

$$\mathbf{d} \quad = \quad - D_p \left[\frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} s \, dA \right] \quad + \quad \left[\langle \tilde{\mathbf{v}} s \rangle - \gamma^{-1} \langle (\mathbf{v} \cdot \nabla \mathbf{v}) s \rangle \right] \tag{3.16}$$

$$\mathbf{D}^* \quad = \quad \varepsilon_\gamma D_p \left(\mathbf{I} + \frac{1}{V_\gamma} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \mathbf{b} \, dA \right) \quad - \quad \langle \tilde{\mathbf{v}} \mathbf{b} \rangle \quad - \quad \gamma^{-1} \langle (\mathbf{v} \cdot \nabla \mathbf{v}) \mathbf{b} \rangle \tag{3.17}$$

$$k_{eff} \quad = \quad \frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla s \, dA \tag{3.18}$$

The last of these coefficients represents the principle objective of this study, thus it is Problem II that will provide results that can be compared with Brownian dynamics and laboratory experiments. In order to determine k_{eff} it is convenient to transform Eqs. 3.13 by means of the following representation

$$s = 1 + k_{eff} S \tag{3.19}$$

Here the quantity S has units of time and the boundary problem for this new variable is given by

PROBLEM II

$$\nabla \cdot (\mathbf{v} S) - \nabla \cdot \left[\left(\gamma^{-1} \mathbf{v} \cdot \nabla \mathbf{v} \right) S \right] = \nabla \cdot \left(D_p \nabla S \right) - \varepsilon_\gamma^{-1} \tag{3.20a}$$

B.C. 1 $$S = 0, \quad \textit{at the } \gamma\text{-}\sigma \textit{ interface} \tag{3.20b}$$

Periodicity: $$S(\mathbf{r} + \ell_i) = S(\mathbf{r}), \quad i = 1,2,3 \tag{3.20c}$$

Average: $$k_{eff} = - \left(\langle S \rangle^\gamma \right)^{-1} \tag{3.20d}$$

In deriving this result from Eqs. 3.13 we have used the continuity equation given by Eq. 2.5, and we have also made use of the simplification indicated by

$$\nabla \cdot \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle^\gamma = 0 \tag{3.21}$$

This is consistent with the simplification used in the closure problem that variations in the porosity can be neglected.

In order to complete the closure of Eq. 3.14 we must represent the term $\langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle$ in a form that can be determined by the use of Darcy's law as given by Eq. 2.29. From the closure problem for Darcy's law (Whitaker, Sec. 4, 1986) we know that the point velocity is given in terms of the intrinsic average velocity by (Barrère et al., 1992)

$$\mathbf{v} = -\left(\mathbf{D} \cdot \mathbf{K}^{-1} \right) \cdot \langle \mathbf{v} \rangle \tag{3.22}$$

Here $\mathbf{D}$ is a second order mapping tensor that is determined by a modified closure problem that has been solved by a variety of authors (Sangani and Acrivos, 1982; Snyder and Stewart, 1966; Zick and Homsy, 1982), and $\mathbf{K}$ is the Darcy's law permeability tensor that appears in Eq. 2.29. In order to use this result with the convective transport term in Eq. 3.14, we make use of the continuity equation, the averaging theorem, and the no-slip condition to obtain

$$\langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle = \langle \nabla \cdot (\mathbf{v}\mathbf{v}) \rangle = \nabla \cdot \langle \mathbf{v}\mathbf{v} \rangle \tag{3.23}$$

Use of Eq. 3.22 in Eq. 3.23 leads to

$$\langle \mathbf{v}\cdot\nabla\mathbf{v}\rangle = \nabla\cdot\left[\langle \mathbf{DD}\rangle:\left(\mathbf{K}^{-1}\mathbf{K}^{-1}\right):\langle \mathbf{v}\rangle\langle \mathbf{v}\rangle\right] \tag{3.24}$$

in which the proper dyadic multiplication can be inferred from Eq. 3.22. When this result is used in Eq. 3.14 we obtain the *completely closed form* of the volume averaged particle transport equation given by

$$\varepsilon_\gamma \frac{\partial\langle n_p\rangle^\gamma}{\partial t} + \nabla\cdot\left\{\left(\langle \mathbf{v}\rangle - \gamma^{-1}\nabla\cdot\left[\langle \mathbf{DD}\rangle:\left(\mathbf{K}^{-1}\mathbf{K}^{-1}\right):\langle \mathbf{v}\rangle\langle \mathbf{v}\rangle\right]\right)\langle n_p\rangle^\gamma\right\} - (\mathbf{d}+\mathbf{u})\cdot\nabla\langle n_p\rangle^\gamma =$$

$$\nabla\cdot\left(\mathbf{D}^*\cdot\nabla\langle n_p\rangle^\gamma\right) - k_{eff}\langle n_p\rangle^\gamma \tag{3.25}$$

For simplicity we define a volume averaged particle velocity according to

$$\langle \mathbf{v}_p\rangle = \langle \mathbf{v}\rangle - \gamma^{-1}\nabla\cdot\left(\langle \mathbf{DD}\rangle:\left(\mathbf{K}^{-1}\mathbf{K}^{-1}\right):\langle \mathbf{v}\rangle\langle \mathbf{v}\rangle\right) \tag{3.26}$$

so that Eq. 3.25 takes the form

$$\varepsilon_\gamma \frac{\partial\langle n_p\rangle^\gamma}{\partial t} + \nabla\cdot\left(\langle \mathbf{v}_p\rangle\langle n_p\rangle^\gamma\right) - (\mathbf{d}+\mathbf{u})\cdot\nabla\langle n_p\rangle^\gamma = \nabla\cdot\left(\mathbf{D}^*\cdot\nabla\langle n_p\rangle^\gamma\right) - k_{eff}\langle n_p\rangle^\gamma \tag{3.27}$$

In order to understand the filtration process, we need to examine the effective coefficients in Eq. 3.27; however, it is the capture coefficient, k_{eff}, that dominates the filtration process, thus the discussion of $\mathbf{d}$, $\mathbf{u}$, and $\mathbf{D}^*$ will be relegated to Appendix B. Solution of the closure problem to determine k_{eff} requires a knowledge of the point velocity field and our results for the velocity and the capture coefficient will be presented in the next section.

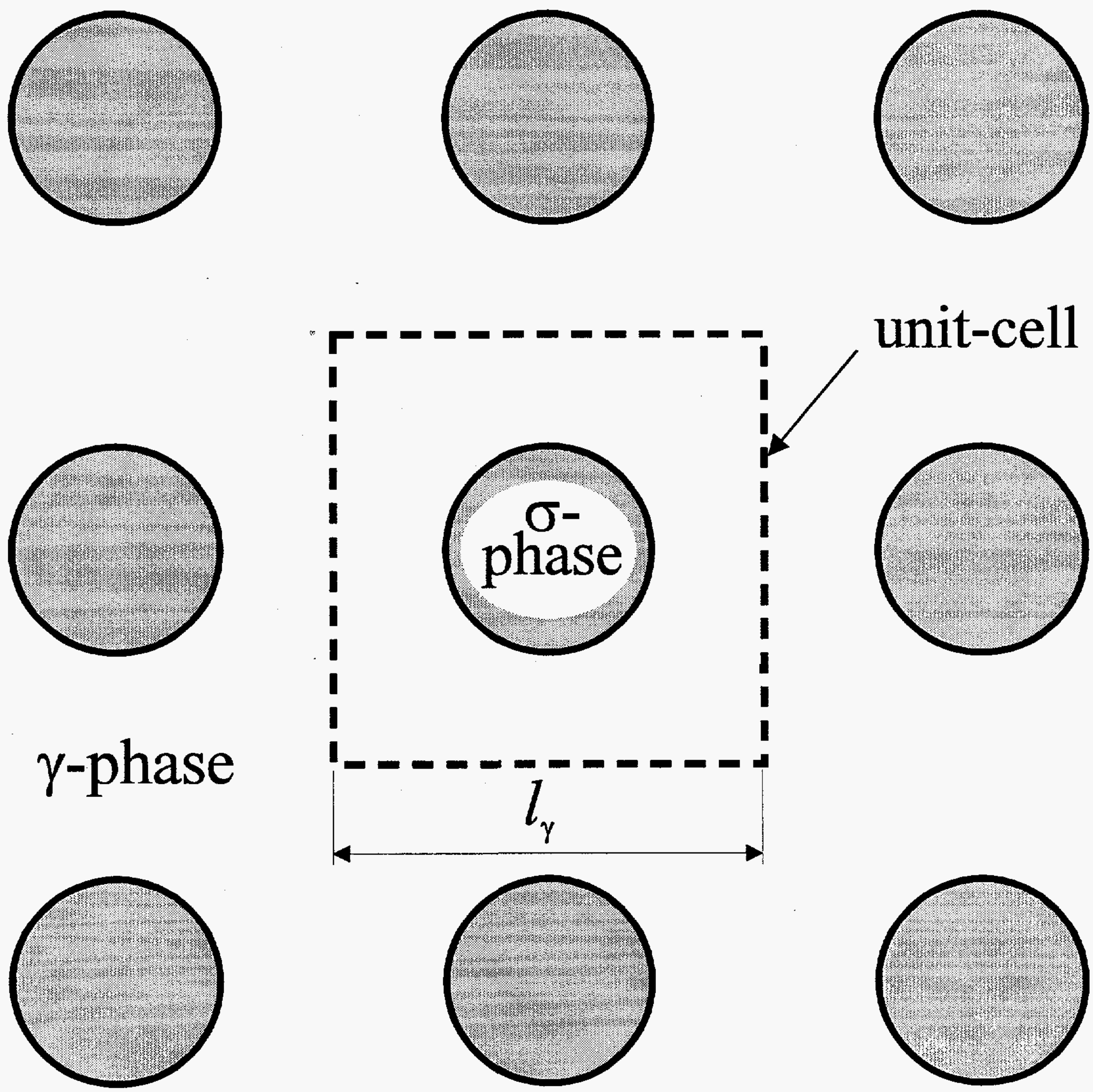
4. Determination of the Velocity Field and the Capture Coefficient

The simplest model of a fibrous porous medium is a regular array of cylinders such as that shown in Figure 3. For most practical cases, the Reynolds number for flow in fibrous filters is less than one, thus we can use Stokes' equations to determine the fluid velocity field. For the case of a *macroscopically uniform flow* one can justify the use of spatially periodic boundary conditions (Sanchez-Palencia, 1980), and the fluid mechanical problem to be solved is given by (Barrère et al., 1992)

$$\nabla\cdot\mathbf{v} = 0 \tag{4.1}$$

$$0 = -\nabla p + \rho\mathbf{g} + \mu\nabla^2\mathbf{v} \tag{4.2}$$

Figure 3
Spatially Periodic Array of Cylinders



B.C. 1 $\mathbf{v} = 0$, *at the* $\gamma-\sigma$ *interface* (4.3)

Periodicity: $\mathbf{v}(\mathbf{r}+\ell_i)=\mathbf{v}(\mathbf{r})$, $i=1$, 2, and 3 (4.4)

Here we have imposed the no-slip condition with the thought that slip will be unimportant when the fiber diameter is larger than several micrometers. The dimensionless velocity profile $(\mathrm{v}_x/\langle \mathrm{v}_x\rangle^\gamma$ as a function of y) at the entrance of a unit cell is illustrated in Figure 4, and there we see that the velocity profile is not uniform as is sometimes assumed in particle tracking calculations associated with the determination of single fiber efficiencies. The center of the cell associated with the profile shown in Figure 4 is at $y/\ell_\gamma = 0.5$, thus we see that the *entrance velocity* at the *edge* of the cell is about twice as large as the *entrance velocity* at the *center* of the cell. While the model shown in Figure 3 is clearly an oversimplification, it does contain some of the general features of a fibrous filter.

It is well known that the entrance length for the Stokes' flow problem is on the order of the characteristic length, ℓ_γ, thus the velocity profile illustrated in Figure 4 is representative of the conditions essentially everywhere in the uniform array shown in Figure 3. However, the *particle velocity* field need not have the same entrance region as the *fluid velocity* field, and it is of some interest to know how many unit cells are required in order to develop a spatially periodic particle velocity since this is required in order that the periodicity conditions represented by Eqs. 3.10c, 3.12c, and 3.13c be valid. The entrance region associated with the particle concentration field can be determined by the solution of Eq. 1.8; however, it is much easier to examine the velocity field on the basis of Eq. 1.2 with the Brownian or random force set equal to zero. This deterministic particle trajectory can be easily calculated on the basis of the special form of Eq. 1.2 given by

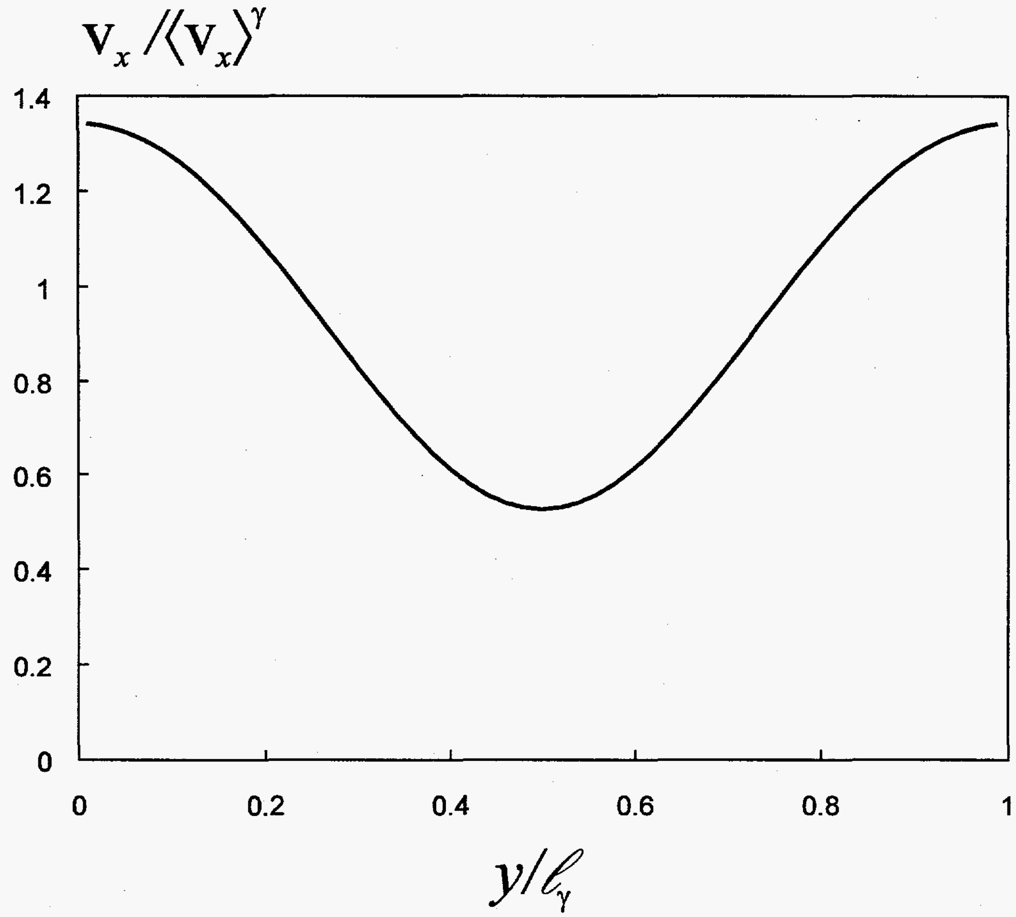
$$\frac{d\overline{\mathbf{v}}_p}{dt} = -\gamma^{-1}\left(\overline{\mathbf{v}}_p-\mathbf{v}\right), \qquad \textit{particle tracking equation} \tag{4.5}$$

and it is interesting to examine $\overline{\mathbf{v}}_p$ for several Stokes numbers where the Stokes number is defined as

$$St = \frac{\langle \mathrm{v}_x\rangle^\gamma \rho_p d_p^2 c_s}{18\mu\ell_\gamma} \tag{4.6}$$

Here ρ_p is the particle density and d_p is the particle diameter. This definition of the Stokes number involves the characteristic length of the unit cell, ℓ_γ, and is therefore different from other definitions that are found in the literature. To be precise about the

Figure 4
Velocity Profile at the Entrance of a Unit Cell



particle tracking calculations that were carried out, we summarize the algorithm as follows:

1. The porous medium is discretized by using a regular grid of blocks assigned to either the fluid phase of the solid phase.

2 Stokes' equations are solved by using a classical Uzawa algorithm.

3. The fluid velocity field required in Eq. 4.5 is obtained from step 2 by linear interpolation.

4. Equation 4.5 is solved by using a sixth order Runge-Kutta algorithm available in the IMSL library.

For the first unit cell shown in Figure 5 there is a wake region that contains no particle trajectories, and for subsequent unit cells there is an upstream region that is also devoid of trajectories. This is to be expected because of the deterministic nature of Eq. 4.5. The characteristic of the particle trajectories that is most interesting is the evolution of the $\overline{\mathbf{v}}_p$-field which is illustrated in Figure 6. There the dimensionless x-component of $\overline{\mathbf{v}}_p$ is shown for several Stokes numbers, and for a Stokes number of 0.155 we see that approximately ten cells are required before a stationary state is reach. In general, ten unit cells represents a small portion of any fibrous filter, thus the entrance region can be neglected and we can make use of spatially periodic conditions (Shapiro and Brenner, 1990) in order to solve the two closure problems given by Eqs 3.12 and 3.20. This will provide us with theoretical values of the effective coefficients in the volume averaged particle transport equation which we list here as

$$\varepsilon_\gamma \frac{\partial \langle n_p \rangle^\gamma}{\partial t} + \nabla \cdot \left(\langle \mathbf{v}_p \rangle \langle n_p \rangle^\gamma \right) - (\mathbf{d} + \mathbf{u}) \cdot \nabla \langle n_p \rangle^\gamma = \nabla \cdot \left(\mathbf{D}^* \cdot \nabla \langle n_p \rangle^\gamma \right) - k_{eff} \langle n_p \rangle^\gamma \tag{4.6}$$

One must remember that the velocity, $\langle \mathbf{v}_p \rangle$, *does not* represent the volume average of the particle velocity that appears in the Langevin equation, but instead it represents the inertia corrected average velocity defined by Eq. 3.21 which can also be expressed as

$$\langle \mathbf{v}_p \rangle = \langle \mathbf{v} \rangle - \gamma^{-1} \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle \tag{4.7}$$

For the special case of a *macroscopically uniform flow*, Eq. (3.24) requires that;

$$\langle \mathbf{v}_p \rangle = \langle \mathbf{v} \rangle \tag{4.8}$$

however, this need not be the case for heterogeneous filters and in future studies we will

Figure 5
Particle Trajectories in a Spatially Periodic Porous Medium

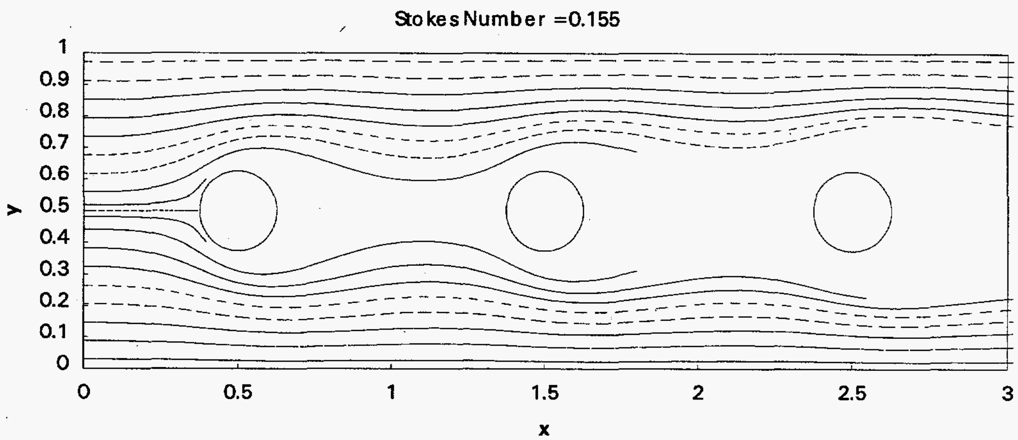
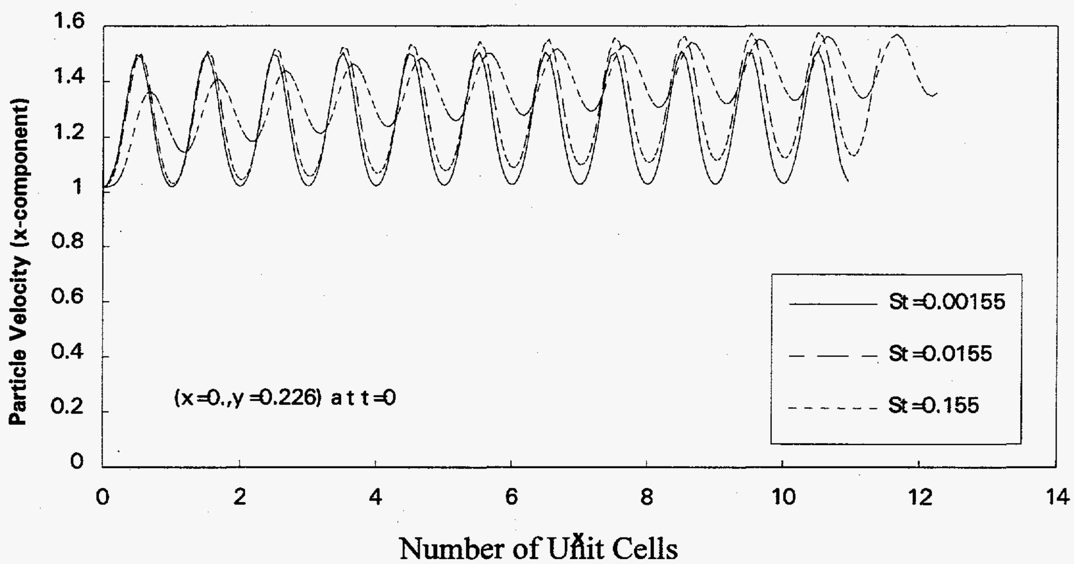


Figure 6
Evolution of Particle Trajectories



examine this matter more carefully. The non-traditional contributions to the convective transport represented by the "velocities", $\mathbf{d}$ and $\mathbf{u}$, are the result of the particle capture process, and this effect has been thoroughly documented by Paine et al. (1983) for the case of adsorption and chemical reaction in capillary tubes. The values of $\mathbf{d}$ and $\mathbf{u}$ are determined by Eqs. 3.15 and 3.16 and it is important to know the magnitude of these two terms relative to $\langle \mathbf{v}_p \rangle$. Calculated values of the x-components of $\mathbf{d}$ and $\mathbf{u}$ are presented in Appendix B where it is shown that these terms may contribute as much as 10% to the convective transport in Eq. 4.6. While this is not negligible, it is not a key issue in terms of the comparison of the theory with calculations based on Brownian dynamics or with laboratory experiments. Thus we will discard these terms, along with the and the dispersive transport so that our superficial volume averaged transport equation takes the form

$$\varepsilon_\gamma \frac{\partial \langle n_p \rangle^\gamma}{\partial t} + \nabla \cdot \left(\langle \mathbf{v} \rangle \langle n_p \rangle^\gamma \right) = - k_{eff} \langle n_p \rangle^\gamma \tag{4.9}$$

This result will be quasi-steady when the following constraint is satisfied

$$k_{eff}\, t \gg 1 \tag{4.10}$$

and for incompressible flows one can use the continuity equation given by Eq. 2.30 in order the express Eq. 4.9 as

$$\langle \mathbf{v} \rangle \cdot \nabla \cdot \langle n_p \rangle^\gamma = - k_{eff} \langle n_p \rangle^\gamma \tag{4.11}$$

One should think of this result as being a reasonable approximation for homogeneous filters; however, real filters are heterogeneous and the terms that have been discarded in going from Eq. 4.6 to Eq. 4.11 will be retained in future studies of heterogeneous porous media.

CELLULAR EFFICIENCY

In order to present our results for k_{eff} in a traditional form, we will write Eq. 4.11 as

$$\langle \mathrm{v}_x \rangle \frac{d \langle n_p \rangle^\gamma}{dx} = - k_{eff} \langle n_p \rangle^\gamma \tag{4.12}$$

and note that for our unit cell calculations, or any homogeneous porous filter, we have

$$\mathbf{i} \cdot \langle \mathbf{v}_p \rangle = \mathbf{i} \cdot \langle \mathbf{v} \rangle = \langle \mathrm{v}_x \rangle \tag{4.13}$$

We can solve Eq. 4.12 in order to represent the change in particle concentration that takes place across a unit cell as

$$\frac{\langle n_p \rangle^\gamma \big|_{x=0} - \langle n_p \rangle^\gamma \big|_{x=\ell_\gamma}}{\langle n_p \rangle^\gamma \big|_{x=0}} = 1 - e^{-\frac{k_{eff}\,\ell_\gamma}{\langle v_x \rangle}} \tag{4.14}$$

It is convenient to define the left hand side of this result as the *cellular efficiency*, η_c , in order to distinguish it from the *single fiber efficiency*, and this leads to

$$\eta_c = 1 - e^{-\frac{k_{eff}\,\ell_\gamma}{\langle v_x \rangle}}, \qquad \textit{cellular efficiency} \tag{4.15}$$

The closure problems given by Eqs. 3.13 and 3.20 were solved using numerical methods similar to those described by Quintard and Whitaker (1993, 1994b). To illustrate the general nature of the solutions for the cellular efficiency, η_c, calculations were carried out for the parameters listed in Table 1.

Table 1 Physical Properties

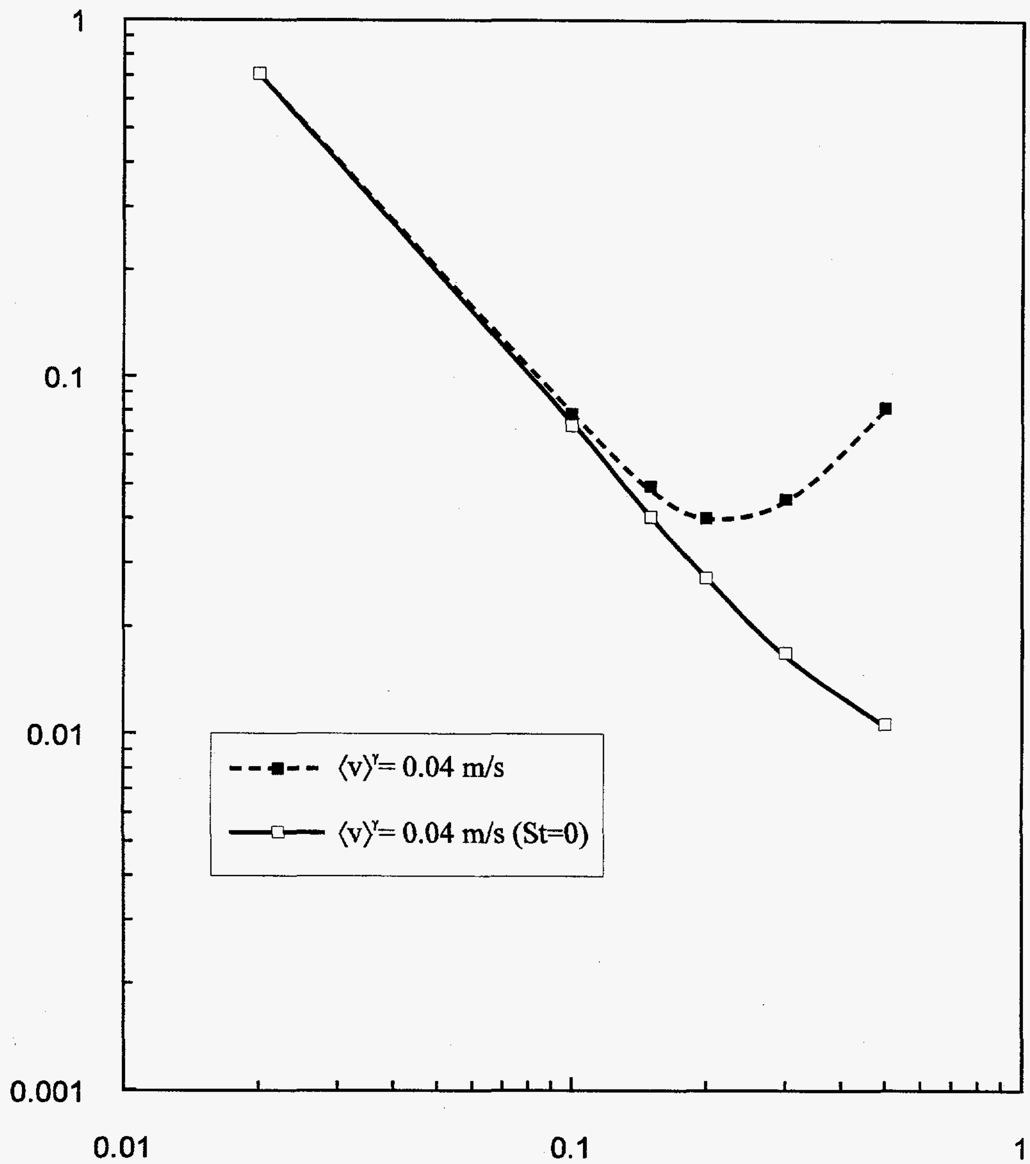
particle diameter	d_p = 0.5 and 0.1 μm
fiber diameter	$2a$ = 0.5 μm
particle density	ρ_p = 4.0 g/cm^3
temperature	283.0 K
viscosity	1.8×10^{-5} Pa s
porosity	ε_γ = 0.95

The illustrative values for η_c are shown in Figure 7 as a function of the particle diameter. The curve for $St = 0$ represents the purely diffusive case and does not exhibit a minimum. This means that the Smoluchowski equation for the particle concentration cannot be used to determine the most penetrating particle size. The curve for finite Stokes numbers, which range from 0^{-4} to almost 0^{-1}, indicates a minimum cellular efficiency for particle diameters on the order of 1 to 2 micrometers. In thinking about the results shown in Figure 7, one must be careful to remember that the additional convective transport represented by the coefficients **u** and **d** has been neglected along with the dispersive transport. Inclusion of these effects would change the values presented in Figure 7 but not the conclusion that the corrected Smoluchowski equation exhibits a minimum in the cellular efficiency as a function of particle diameter.

Although laboratory experiments are not available for the cellular efficiency, we can compare our results with the Brownian dynamics calculations of

Figure 7
Cellular Efficiency as a Function of Particle Diameter

($2a = 0.5$ μm, $\rho_p = 4000$ kg/m^3
T=283 K, μ=1.8 10^{-5} Pa s, ε_γ=0.95)



Tien and Ramarao (1992) and this comparison is illustrated in Figure 8 for a fiber diameter of 10 micrometers and a wide range of particle diameters. The conditions are representative of air flowing in a typical fibrous filter, and our calculated results resemble those of Tien and Ramarao but are lower by about a factor of two for particle diameters that are equal to or less than the diameter of the most penetrating particle. For larger particles the theory predicts values of η_c that are *significantly lower* than those determined by Brownian dynamics, and this would appear to indicate that the higher order terms in Eq. 1.7 need to be included in the theory. One should keep in mind that the boundary condition given by Eq. 2.2 is an *ad hoc* representation of the true physics of the particle capture process; however, it represents an *upperbound* in the particle capture process and therefore cannot be responsible for the differences observed in Figure 8.

Brownian dynamics calculations can be thought of as *numerical experiments* based on Eq. 1.2 and are therefore not subject to the errors associated with: (1) the derivation of Eq. 1.7, (2) the approximations contained in Eq. 1.8, and (3) the simplifications imposed on an analysis that began with Eq. 2.1 and ended with Eqs. 3.20. Because of this, any reliable continuum theory should agree with Brownian dynamics calculations. Since Tien and Ramarao used the Kuwabara unit cell to determine their velocity field, we should not expect their results to be exactly the same as those determined by the continuum particle transport equation. However, the velocity field generated from the Kuwabara unit cell is not greatly different from that calculated using the spatially periodic model shown in Figure 3. What is different between the two approaches is the boundary condition imposed at the entrance to the unit cell, and this difference is significant. In Brownian dynamics calculations the particles are uniformly distributed over the entrance to the unit cell, whereas this is not the case for the spatially periodic model shown in Figure 3. From Eqs 2.14 and 3.11, we have the following representation of the point particle concentration in terms of the closure variables

$$n_p = (1-s)\langle n_p\rangle^\gamma + \mathbf{b}\cdot\nabla\langle n_p\rangle^\gamma \tag{4.16}$$

If the gradient of $\langle n_p\rangle^\gamma$ is sufficiently small, we can extract an approximation that gives us the point particle concentration in terms of the average particle concentration. This is obviously given by

$$n_p \approx (1-s)\langle n_p\rangle^\gamma \tag{4.17}$$

and the calculated values of $(1-s)$ at $x=0$ are used to produce the results shown in Figure 9. The fully three-dimensional representation for the s-field is shown in Figure 10 where

Figure 7
Cellular Efficiency as a Function of Particle Diameter

($2a = 0.5$ µm, $\rho_p = 4000$ kg/m^3
T=283 K, μ=1.8 10^{-5} Pa s, ε_γ=0.95)

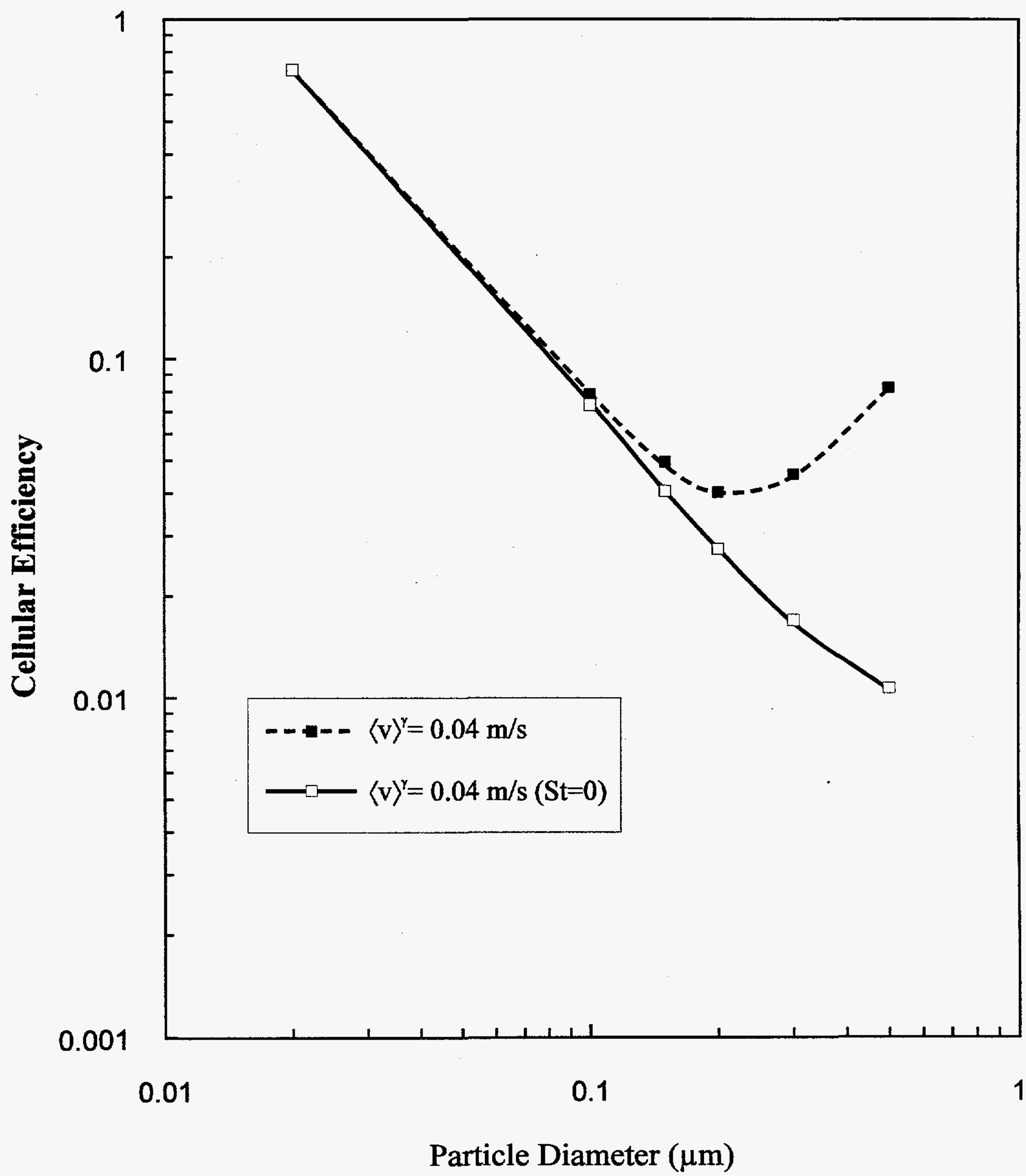


Figure 8
Comparison with the Cellular Efficiency from Brownian Dynamics

($2a = 10\ \mu m$, $\rho_p = 1000\ kg/m^3$
$T=278\ K$, $\mu=1.83\ 10^{-5}\ Pa\ s$, $\varepsilon_\gamma=0.9$)

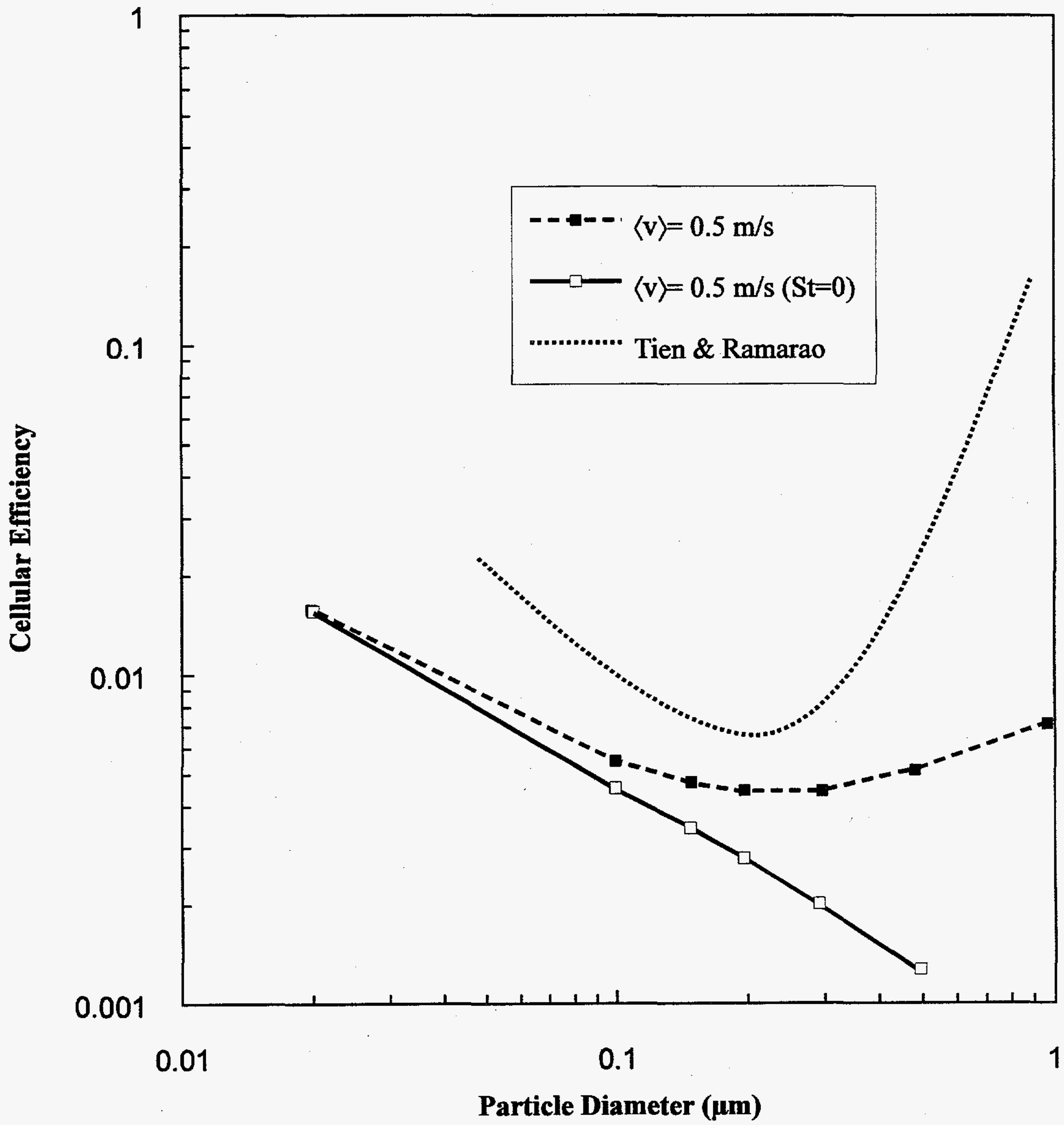


Figure 9
Concentration Profile at the Entrance to a Unit Cell in a Spatially Periodic Model of a Fibrous Filter

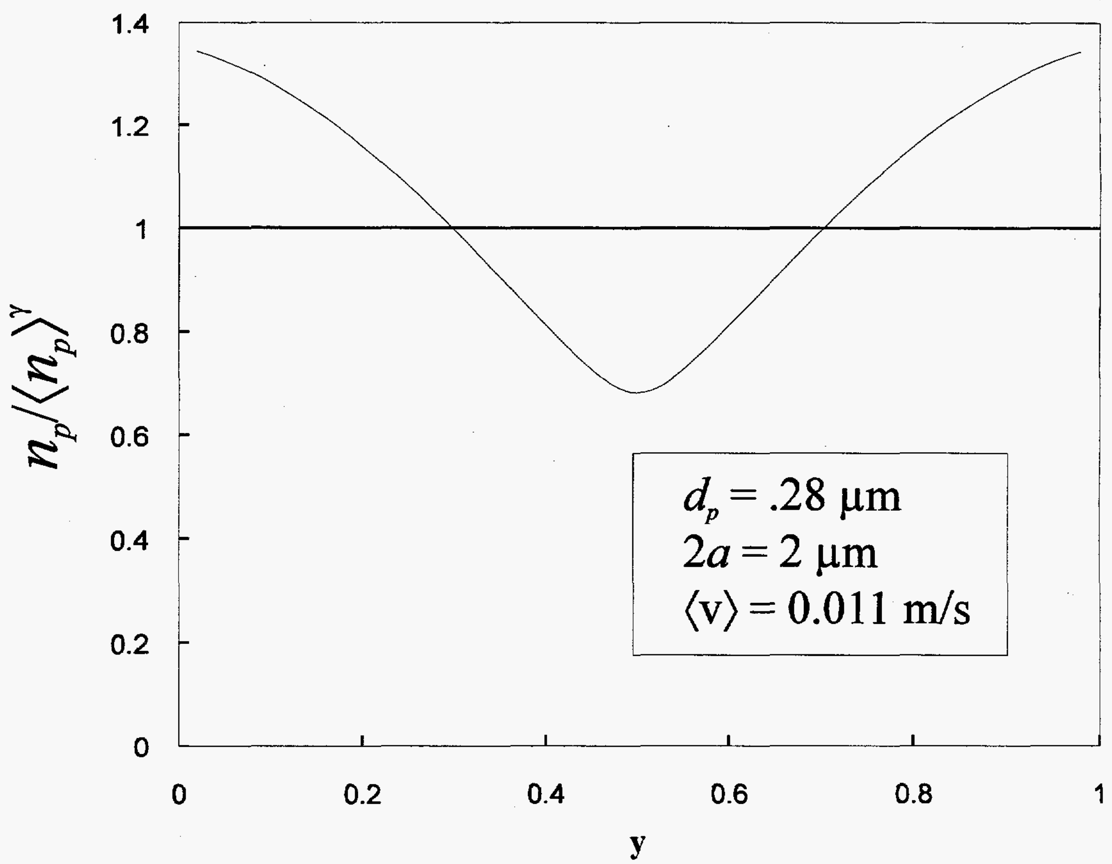
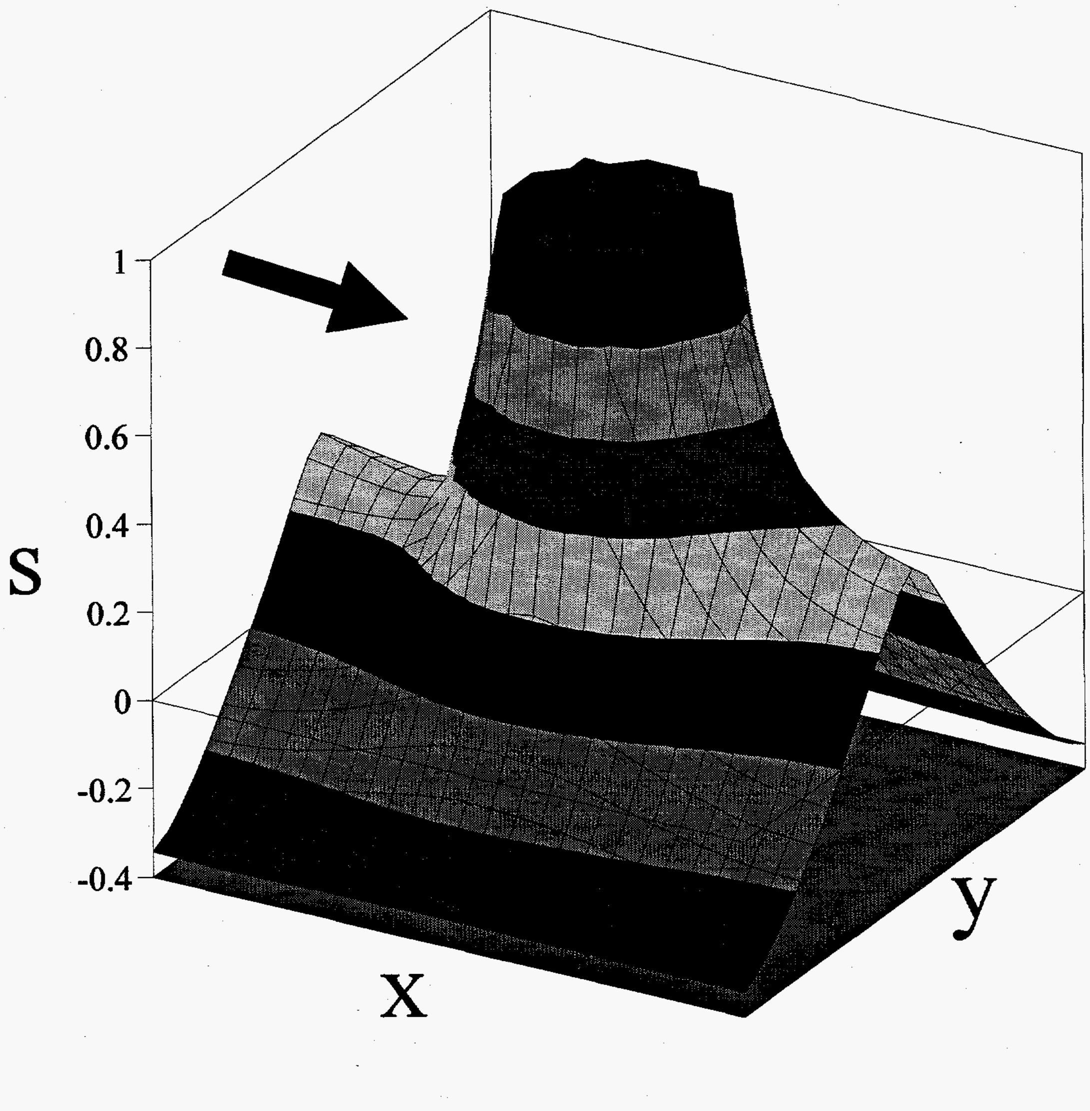


Figure 10

Three-Dimensional Representation of the *s*-Field



one can observe the non-uniformity at the entrances and exits in addition to the boundary layer that exits around the fiber. The variation of $n_p/\langle n_p \rangle^\gamma$ shown in Figure 9 is reminiscent of the velocity profile shown in Figure 4 and it represents a very significant particle concentration *nonuniformity* at the entrance (and at the exit by periodicity) of a unit cell. The fact that the concentration is *smaller* at the center of the entrance to the unit cell naturally gives rise to a rate of particle capture that is less than one would calculate using a uniform concentration at the entrance to the unit cell. We believe that this non-uniformity, that naturally arises because of the spatially periodic conditions associated with the model illustrated in Figure 3, is the source of the difference between the continuum theory and the Brownian dynamics calculations shown in Figure 8 for particle diameters that are equal to or less than the diameter of the most penetrating particle. For larger particle diameters, it is difficult to make a judgment since the rate of capture of larger particles will be very sensitive to the manner in which they are distributed over the entrance to the unit cell

In addition to comparing the theory with numerical experiments, it is important to compare our results with laboratory experiments. In Figure 11 we have shown a comparison with the work of Lee and Liu (1982) for an average velocity given by $\langle \mathrm{v} \rangle = 0.1 m/s$, and there one can see attractive agreement between theory and experiment for particle diameters that are equal to or smaller than the diameter of the most penetrating particle. For larger particles, the agreement diminishes and this would appear to confirm our suspicions concerning the importance of the higher order terms in Eq. 1.7. In Figure 12 the comparison between theory and experiment in shown for $\langle \mathrm{v} \rangle = 0.03 m/s$ and we agin see reasonable agreement for the smaller particles. The comparison for an even smaller velocity given by $\langle \mathrm{v} \rangle = 0.01 m/s$ is shown in Figure 13 and here it becomes apparent that the theory is less reliable at lower velocities. We have no explanation for this observation; however, one must remember that the model illustrated in Figure 3 cannot possibly capture all the characteristics of a real filter and further studies using more complex unit cells are certainly in order. In addition, the influence of local heterogeneities must be determined and that is the objective of a subsequent study.

5. **Conclusions**

In this work we have used the first correction to the Smoluchowski equation to describe the effects of particle inertia, and the resulting particle transport equation has been used to develop a local volume average transport equation that includes the effects of non-traditional convective transport, dispersion, and particle capture. A spatially periodic model of a fibrous filter has been used, along with two closure problems, to calculated the effective coefficients that appear in the volume averaged transport equation. This leads to a direct calculation of the cellular efficiency and results were determined for a unit cell containing a single fiber that is orthogonal to the mean flow field. The results are in reasonably good agreement with numerical experiments performed by means of Brownian dynamics and with laboratory experiments. Both

Figure 11
Comparison with Laboratory Experiments

($2a = 11\ \mu m$, $\rho_p = 1000\ kg/m^3$
$T = 278\ K$, $\mu = 1.83\ 10^{-5}\ Pa\ s$, $\varepsilon_\gamma = 0.849$)

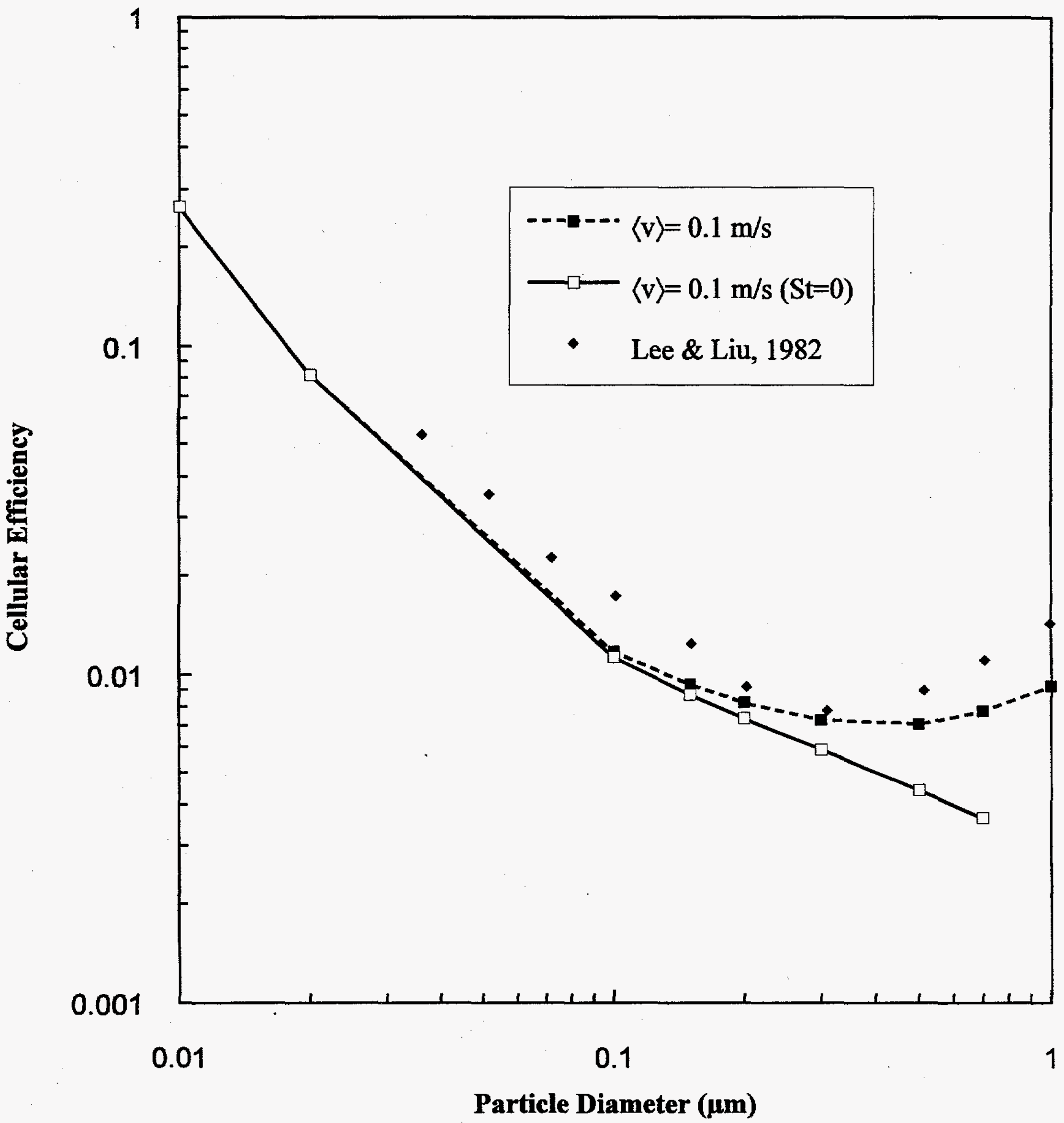


Figure 12
Comparison with Laboratory Experiments

($2a$ = 11 μm, ρ_p = 1000 kg/m^3
T=278 K, μ=1.83 10^{-5} Pa s, ε_γ=0.849)

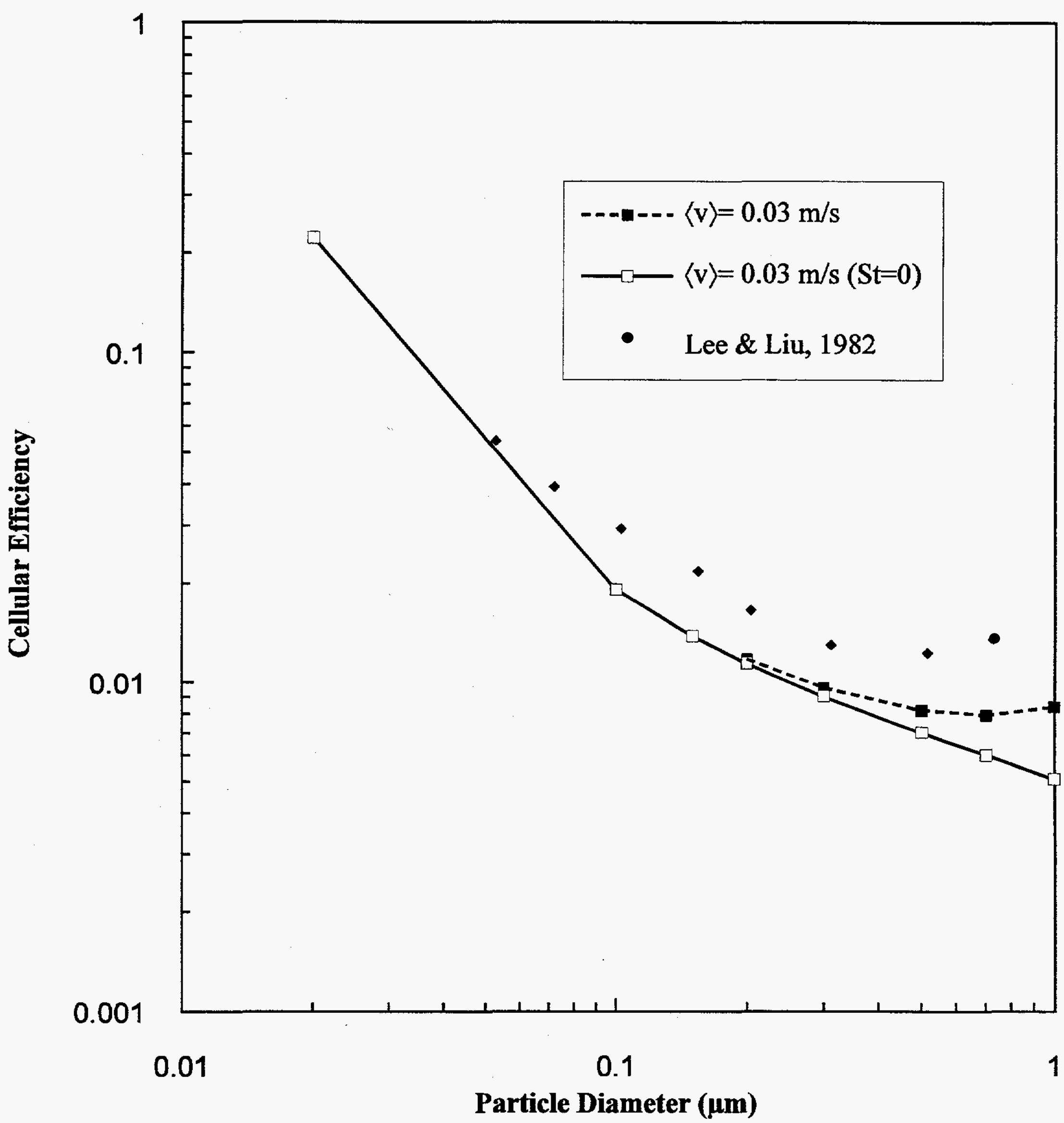
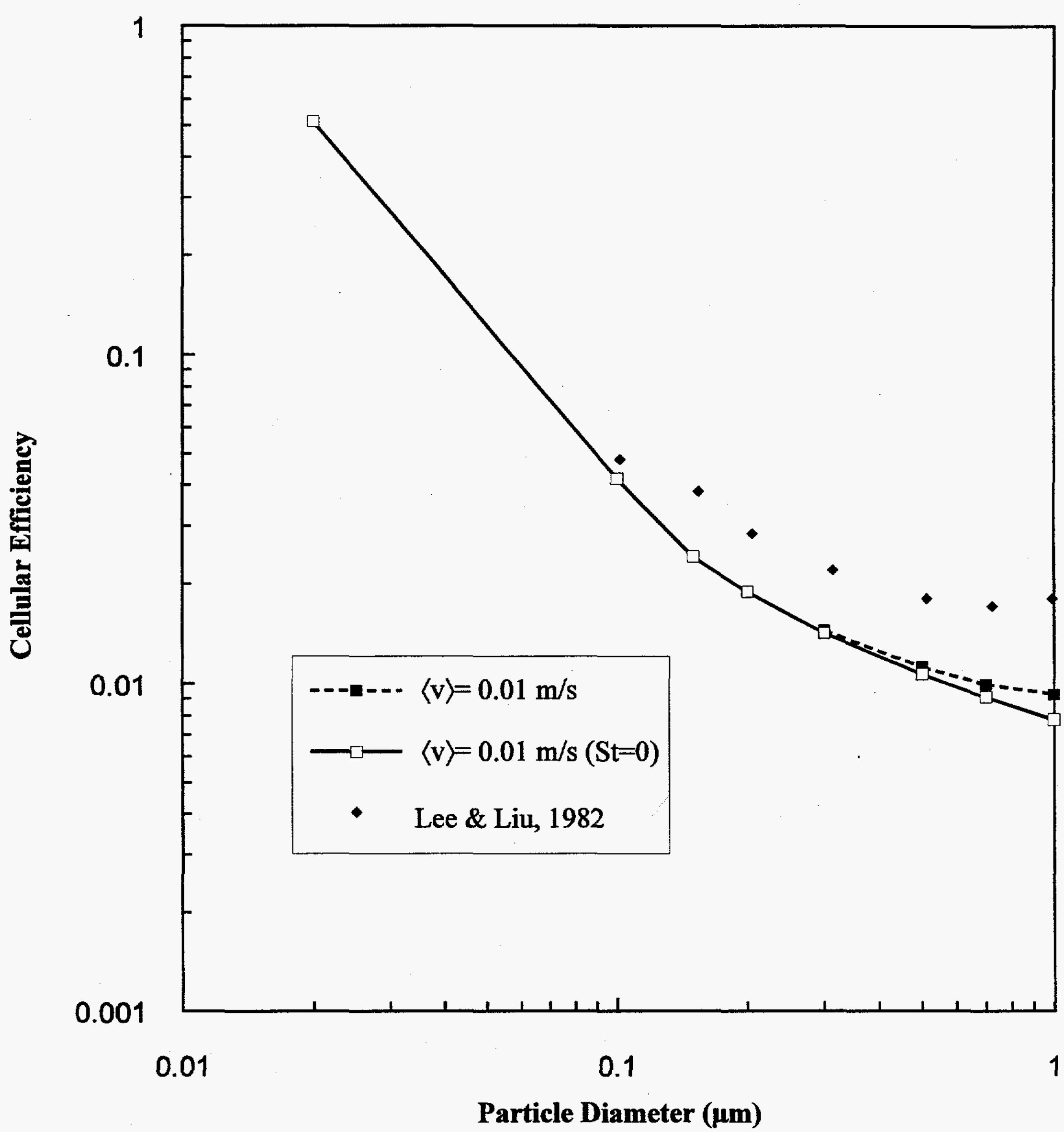


Figure 13
Comparison with Laboratory Experiments

($2a = 11\ \mu m$, $\rho_p = 1000\ kg/m^3$
$T=278\ K$, $\mu=1.83\ 10^{-5}\ Pa\ s$, $\varepsilon_\gamma=0.849$)



comparisons indicated that higher order corrections need to be included in the Smoluchowski equation in order to predict the behavior of large particles.

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7 Nomenclature

Roman Letters

a	fiber radius, m.
$A_{\gamma\sigma}$	area of the γ-σ interface contained within the averaging volume, m^2.
$\mathbf{b}$	a closure variable that maps $\nabla\langle n_p\rangle^\gamma$ onto $\tilde{n}_p$, m.
d_p	particle diameter, m.
$\mathbf{d}$	a velocity-like coefficient, m/s
D_p	Brownian diffusivity, m^2/s.
$\mathbf{D}^*$	dispersion tensor, m^2/s
$\mathbf{F}_r(t)$	Brownian or random force, N.
$\mathbf{g}$	gravity vector, m/s^2
k_{eff}	effective rate coefficient for particle capture, s^{-1}
ℓ_γ	characteristic length for a unit cell, m.
ℓ_σ	characteristic length for the σ-phase (= $2a$), m.
ℓ_i	i = 1, 2, and 3, lattice vectors, m.
L_ε	characteristic length for the porosity, m.
L_n	characteristic length for $\langle n_p\rangle^\gamma$, m.
L	generic characteristic length for volume averaged quantities, m.
m_p	mass of a particle, kg.
n_p	particle density, number/m^3.
$\tilde{n}_p$	spatial deviation of the particle of the particle, number/m^3.
$\langle n_p\rangle^\gamma$	intrinsic average particle concentration, number/m^3.
$\mathbf{n}_{\gamma\sigma}$	unit normal vector pointing from the γ-phase toward the σ-phase.
p	fluid pressure, N/m^2.
$\langle p\rangle^\gamma$	intrinsic average pressure in the γ-phase, m^3.
$\mathbf{r}$	position vector, m.
s	a closure variable that maps $\langle n_p\rangle^\gamma$ onto $\tilde{n}_p$.
St	$\langle v_x\rangle^\gamma \rho_p d_p^2 c_s / 18\mu\ell_\gamma$, the Stokes number.
t	time, s.

$\mathbf{u}$ a velocity-like coefficient, m/s.
averaging volume, m^3.
V_γ volume of the γ-phase contained in the averaging volume, m^3.
$\mathbf{v}$ fluid velocity vector, m/s.
$\langle \mathbf{v} \rangle^\gamma$ intrinsic average fluid velocity, m/s.
$\langle \mathbf{v} \rangle$ $\varepsilon_\gamma \langle \mathbf{v} \rangle^\gamma$, superficial average fluid velocity, m/s.
$\tilde{\mathbf{v}}$ $\mathbf{v} - \langle \mathbf{v} \rangle^\gamma$, spatial deviation fluid velocity, m/s.
$\mathbf{v}_p$ particle velocity, m/s.
$\overline{\mathbf{v}}_p$ deterministic particle velocity, m/s.
$\langle \mathbf{v}_p \rangle$ $\langle \mathbf{v} \rangle - \gamma^{-1} \langle \mathbf{v} \cdot \nabla \mathbf{v} \rangle$, inertia corrected volume average velocity for the particles, m/s.
$\mathbf{y}$ position vector relative to the centroid of the averaging volume, m.
$\mathbf{x}$ position vector locating the centroid of the averaging volume, m.

Greek Letters

μ fluid viscosity, Ns/m^2.
π 3.1416.......
ε_γ porosity.
γ $3\pi\mu d_p / m_p$, s^{-1}
ρ fluid density, kg/m^3.
ρ_p particle density, kg/m^3.

7. References

Anderson, T.B. and Jackson, R. 1967, A fluid mechanical description of fluidized beds, Ind. Engng. Chem. Fundam. **6**, 527 - 538.

Banks, D.O. and Kurowski, G.J. 1983, A perturbation method for the approximation of the inertial collection efficiency for fibrous filters with electrical enhancement, J. Aerosol Sci. **14**, 463 - 473.

Banks, D.O., Kurowski G.J., and Whitaker, S. 1991, Diffusion deposition on a fiber in non-transverse flow, Aerosol Sci. and Technol. **14**, 224-232.

Brenner, H. 1980, Dispersion resulting from flow through spatially periodic porous media, Trans. Roy. Soc. (London) **297**, 81 - 133.

Carbonell, R.G. and Whitaker, S., 1983, Dispersion in pulsed systems II: Theoretical developments for passive dispersion in porous media, Chem. Engng. Sci. **38**, 1795 - 1802.

Carbonell, R.G. and Whitaker, S. 1984, Heat and mass transfer in porous media, pages 123 - 198 in *Fundamentals of Transport Phenomena in Porous Media*, edited by J. Bear and M.Y. Corapcioglu, Martinus Nijhoff Publishers, Dordrecht, The Netherlands.

Crapiste, G.H., Rotstein, E. and Whitaker, S. 1986, A general closure scheme for the method of volume averaging, Chem. Engng. Sci. **41**, 227 - 235.

Cushman, J.H. 1990, *Dynamics of Fluids in Hierarchical* Porous Media, Academic Press, London.

de la Mora, J.F. and Rosner, D.E. 1981, Inertial deposition of particles revisited and extended: Eulerian approach to a traditionally lagrangian problem, PhysicoChem. Hydrody. **2**, 1 - 21.

de la Mora, J.F. and Rosner, D.E 1982, Effects of inertia on the diffusional deposition of small particles to spheres and cylinders at low Reynolds numbers, J. Fluid Mech. **125**, 379 - 395.

Edwards, D.A., Shapiro, M. and Brenner, H. 1993, Dispersion and reaction in two-dimensional model porous media, Phys. Fluids **A** 5(4), 837-848.

Eidsath, A.B., Carbonell, R.G., Whitaker, S., and Herrmann, L.R., 1983, Dispersion in pulsed systems III: Comparison between theory and experiments for packed beds, Chem. Engng. Sci. **38,** 1803 - 1816.

Gardiner, C.W. 1990, *Handbook of Stochastic Methods*, Springer-Verlag, New York.

Gray, W.G., 1975, A derivation of the equations for multiphase transport, Chem. Engng. Sci. **30**, 229 - 233.

Gupta, D. and Peters, M.H. 1985, A Brownian dynamics simulation of aerosol deposition onto spherical collectors, J. Coll. Inter. Sci. **104**, 375 - 389.

Gupta, D. and Peters, M.H. 1986, On the angular dependence of aerosol diffusional deposition onto spheres, J. Coll. Interf. Sci. **110**, 286 - 303.

Howes, F.A. and Whitaker, S., 1985, The spatial averaging theorem revisited, Chem. Engng. Sci. **40,** 1387 - 1392.

Kanaoka, C., Emi, H. and Tanthapanichakoon, W. 1983, Convective diffusional deposition and collection efficiency of aerosol on a dust-loaded fiber, AIChE J. **29**, 845 - 902.

Lee, H.L. 1979, Analysis of pseudo-continuum mass transfer in media with spatially periodic boundaries, Chem. Engng. Sci. **34**, 503 - 514.

Marle, C.M. 1967, Écoulements monophasique en milieu poreux, Rev. Inst. Français du Pétrole **22**(10), 1471 - 1509.

Mauri,R. 1991, Dispersion, convection, and reaction in porous media, Phys. Fluids **A3**, 743 - 756.

Nozad, I., Carbonell, R.G., and Whitaker, S. 1985, Heat conduction in multiphase systems I: Theory and experiment for two-phase systems, Chem. Engng. Sci. **40**, 843 - 855.

Ochoa, J.A., Stroeve, P. and Whitaker, S. 1986, Diffusion and reaction in cellular media, Chem. Engng. Sci. **41**, 2999 - 3013.

Peters, M.H. and Ying, R. 1991, Phase-space diffusion equations for single Brownian particle motion near surfaces, Chem. Eng. Comm. **108**, 165 - 185.

Plumb, O.A. and Whitaker, S., 1988, Dispersion in heterogeneous porous media I: Local volume averaging and large-scale averaging, Water Resour. Res. **24,** 913 - 926.

Plumb, O.A. and Whitaker, S., 1988, Dispersion in heterogeneous porous media II: Predictions for stratified and two-dimensional spatially periodic systems, Water Resour. Res. **24**, 927 - 938.

Plumb, O.A. and Whitaker, S., 1990, Diffusion, adsorption, and dispersion in porous media: Small-scale averaging and local volume averaging, Chapter V in *Dynamics of Fluids in Hierarchical Porous Media,* edited by J.H. Cushman, Academic Press, New York.

Plumb, O.A. and Whitaker, S., 1990, Diffusion, adsorption, and dispersion in heterogeneous porous media: The method of large-scale averaging, Chapter VI in *Dynamics of Fluids in Hierarchical Porous Media*, edited by J.H. Cushman, Academic Press, New York.

Quintard, M. and Whitaker, S., 1987, Écoulement monophasique en milieu poreux: Effet des hétérogénéités locales, J. Méc. théor. appl. **6**, 691 - 726.

Quintard, M. and Whitaker, S., 1988, Two-phase flow in heterogeneous porous media: The method of large-scale averaging, Transport in Porous Media **3**, 357 - 413.

Quintard, M. and Whitaker, S., 1990, Two-phase flow in heterogeneous porous media I: The influence of large spatial and temporal gradients, Transport in Porous Media **5**, 341 - 379.

Quintard, M. and Whitaker, S. 1990, Two-phase flow in heterogeneous porous media II: Numerical experiments for flow perpendicular to a stratified system, Transport in Porous Media **5**, 429 - 472.

Quintard, M. and Whitaker, S. 1993, One and two-equation models for transient diffusion processes in two-phase systems, pages 369 - 465 in *Advances in Heat Transfer*, Vol. 23, Academic Press, New York.

Quintard, M. and Whitaker, S. 1994a, Convection, dispersion, and interfacial transport of contaminants: Homogeneous porous media, Adv. Water Resour., in press.

Quintard, M. and Whitaker, S. 1994b, Transport in ordered and disordered porous media I: The cellular average and the use of weighting functions, Transport in Porous Media **14**, 163 - 177.

Quintard, M. and Whitaker, S. 1994b, Transport in ordered and disordered porous media II: Generalized volume averaging, Transport in Porous Media **14**, 179 - 206.

Quintard, M. and Whitaker, S. 1994b, Transport in ordered and disordered porous media III: Closure and comparison between theory and experiment, Transport in Porous Media, in press.

Quintard, M. and Whitaker, S. 1994b, Transport in ordered and disordered porous media IV: Computer generated porous media, Transport in Porous Media, in press.

Quintard, M. and Whitaker, S. 1994b, Transport in ordered and disordered porous media V: Geometrical results for two-dimensional systems, Transport in Porous Media, in press.

Ramarao, B.V. and Tien, C. 1991, Simulation of particle deposition in fluid flow, pages 191 - 246 in Transport Processes in Bubbles, Drops, and Particles, edited by R.P. Chhabra and D. DeKee, Hemisphere Pub. Co., New York

Ramarao, B.V., Tien, C., and Mohan, S 1994, Calculation of Single fiber efficiencies for interception and impaction with superposed Brownian motion, J. Aerosol Sci. **25**, 295 - 313.

Risken, H. 1989, *The Fokker-Planck Equation: Methods of Solution and Applications*, Springer-Verlag, New York.

Rubinstein, J.and Mauri, R. 1986, Dispersion and convection in periodic porous media, SIAM J. appl. Math. 46, 1018-1023.

Ruckenstein, E. and Prieve, D. 1973, Rate of deposition of Browian particles under the action of London and doble-layer forces, J. Chem. Soc. (London) Faraday Trans. **69**, 1522 - 1536.

Russel, W.B. 1981, Brownian motion of small particles suspended in liquids, *Ann, Rev. Fluid Mech.* **13**, 425 - 455.

Sahroui, M. and Kaviany, M. 1994, Slip and no-slip temperature boundary conditions at the interface of porous, plain media: Convection, Int. J. Heat Mass Trans. **37**, 1029 - 1044.

Sanchez-Palencia, E., 1980, Non-homogeneous media and vibration theory, *Lecture Notes in Physics* **127**, Springer-Verlag, New York.

Sangani, A.S. and Acrivos, A. 1982, Slow flow past periodic arrays of cylinders with application to heat transfer, Int. J. Multiphase Flow **8**, 193 - 206.

Skinner, J.L. and Wolynes, P.G., 1979, Derivation of Smoluchowski equations with corrections for Fokker-Planck and BGK collision models, Physica **96A**, 561 - 572.

Slattery, J.C. 1967, Flow of viscoelastic fluids through porous media, AIChE Journal **13**, 1066 - 1071.

Snyder, L.J. and Stewart, W.E. 1966, Velocity and pressure profiles for Newtonian creeping flow in regular packed beds of spheres, AIChE Journal **12**, 167 - 173.

Tien, C. *Granular Filtration of Aerosols and Hydrosols*, Butterworths, Boston.

Tien, C. and Ramarao, B.V., Simulation and calculation of diffusive aerosol deposition, report submitted to Dr. Werner Bergman, Subcontract B 160439, Lawrence Livermore National Laboratory.

Titulaer, U.M. 1978, A systematic solution procedure for the Fokker-Planck equation of a Browian partcile in the high-friction case, Physica **91A**, 321 - 344.

Whitaker, S. 1967, Diffusion and dispersion in porous media, AIChE Journal **13**,420 - 427.

Whitaker, S., 1986a, Transport processes with heterogeneous reaction, pages 1 to 94 in *Concepts and Design of Chemical Reactors,* edited by S. Whitaker and A.E. Cassano, Gordon and Breach Publishers, New York.

Whitaker, S., 1986b, Flow in porous media I: A theoretical derivation of Darcy's law, Transport in Porous Media **1,** 3 - 25.

Yeh, H-S. and Liu, B.Y.H. 1974, Aerosol filtration by fibrous filters I: Theoretical, Aerosol Science, **5**, 191 - 204.

Zanotti, F. and Carbonell, R.G. 1984, Development of transport equations for multiphase systems I: General development for two-phase systems, Chem. Engng. Sci. **39**, 263 - 278.

Zanotti, F. and Carbonell, R.G. 1984, Development of transport equations for multiphase systems II: Application to one-dimensional axisymmetric flows of two-phases, Chem. Engng. Sci. 39, 279 - 297.

Zanotti, F. and Carbonell, R.G. 1984, Development of transport equations for multiphase systems III: Application to heat transfer in packed beds, Chem. Engng. Sci. **39**, 299 - 311.

Zick, A.A. and Homsy, G.M. 1982, Stokes' flow through periodic arrays of spheres, J. Fluid Mech. **115**, 13 - 26.

APPENDIX A
ESTIMATES AND CONSTRAINTS

In order to develop the constraints associated with the volume averaged particle transport equation and with the closure problem, we need to estimate the spatial deviation particle concentration. In the boundary value problem for $\tilde{n}_p$ given by Eqs. 3.6 through 3.9 we see two *volumetric sources* in the transport equation, and one *surface source* in the interfacial boundary condition. It is these sources that give rise to non-zero values of $\tilde{n}_p$, and in order to estimate the magnitude of the spatial deviation particle concentration we simply need to estimate the contributions of these three sources. The surface source contribution is straightforward and is obviously given by

$$\tilde{n}_p\Big|_{\substack{surface\\source}} = \mathbf{O}\left(\langle n_p\rangle^\gamma\right) \tag{A1}$$

The estimate of the contribution due to the volume sources is based on the idea that the diffusive term in Eq. 3.6 is one of the dominant terms. It is easy to show that the diffusive term is the same order of magnitude as the capture term

$$\nabla\left(D_p\nabla\tilde{n}_p\right) = \mathbf{O}\left\{\frac{\varepsilon_\gamma^{-1}}{\mathscr{V}}\int_{A_{\gamma\sigma}}\mathbf{n}_{\gamma\sigma}D_p\nabla\tilde{n}_p dA\right\} \tag{A2}$$

and it may be more appealing to think of this latter term as the dominant term in a filtration process. The contribution of the first volume source is based on

$$\nabla\cdot\left(D_p\nabla\tilde{n}_p\right) = \mathbf{O}\left(\left[\tilde{\mathbf{v}}-\gamma^{-1}(\mathbf{v}\cdot\nabla\mathbf{v}-\langle\mathbf{v}\cdot\nabla\mathbf{v}\rangle^\gamma)\right]\nabla\langle n_p\rangle^\gamma\right) \tag{A3}$$

and in order to extract something comparable to Eq. A1 we need to estimate the gradients of both $\tilde{n}_p$ and $\langle n_p\rangle^\gamma$. In addition, we need to estimate the terms involving the velocity on the right hand side of Eq. A3. We begin by noting that the particle inertial contribution, in a general sense, will be constrained by

$$\gamma^{-1}\mathbf{v}\cdot\nabla\mathbf{v} \leq \mathbf{O}(\mathbf{v}) \tag{A4}$$

This leads to the simplification

$$\tilde{\mathbf{v}}-\gamma^{-1}(\mathbf{v}\cdot\nabla\mathbf{v}-\langle\mathbf{v}\cdot\nabla\mathbf{v}\rangle^\gamma) = \mathbf{O}(\tilde{\mathbf{v}}) \tag{A5}$$

and allows us to write Eq. A3 as

$$\nabla\cdot\left(D_p\nabla\tilde{n}_p\right) = \mathbf{O}\left(\langle\mathbf{v}\rangle^\gamma\nabla\langle n_p\rangle^\gamma\right) \tag{A6}$$

since $\tilde{\mathbf{v}}$ is on the order of $\langle\mathbf{v}\rangle^\gamma$ because of the no-slip condition given by Eq. 2.4. We now *define* a characteristic length for $\tilde{n}_p$ according to

$$\nabla\tilde{n}_p = \mathbf{O}\left(\frac{\tilde{n}_p}{\ell_{bl}}\right) \tag{A7}$$

Here one should think of ℓ_{bl} as the boundary layer thickness associated with the particle adsorption process (Banks et al., 1991) and we know that ℓ_{bl} is constrained by

$$\ell_{bl} < \ell_\gamma \tag{A8}$$

The characteristic length for $\langle n_p\rangle^\gamma$ is *defined* by

$$\nabla\langle n_p\rangle^\gamma = \mathbf{O}\left(\frac{\langle n_p\rangle^\gamma}{L_n}\right) \tag{A9}$$

and when this result, along with Eq. A7, is used in Eq. A6 we obtain the estimate

$$\tilde{n}_p\Big|_{\substack{volume\\source}} = \mathbf{O}\left[Pe\left(\frac{\ell_{bl}}{\ell_\sigma}\right)\left(\frac{\ell_{bl}}{L_n}\right)\langle n_p\rangle^\gamma\right] \tag{A10}$$

Here we have defined the Peclet number by

$$Pe = \frac{\ell_\sigma\langle\mathrm{v}\rangle^\gamma}{D_p} \tag{A11}$$

and since the boundary layer thickness can be estimated by (Banks, et al., 1991)

$$\ell_{bl} = \mathbf{O}\left(\frac{\ell_\sigma}{Pe^{\frac{1}{3}}}\right) \tag{A12}$$

we can express Eq. A10 as

$$\tilde{n}_p\Big|_{\substack{volume\\source}} = \mathbf{O}\left[Pe^{\frac{1}{3}}\left(\frac{\ell_\sigma}{L_n}\right)\langle n_p\rangle^\gamma\right] \tag{A13}$$

For typical values of Pe, ℓ_σ, and L_n this contribution to $\tilde{n}_p$ will be much less than that resulting from the surface source represented by Eq. A1. An examination of the second volume source in Eq. 3.6 leads one to a similar conclusion, thus we can represent our order of magnitude estimate of $\tilde{n}_p$ as

$$\tilde{n}_p = \mathbf{O}\left(\langle n_p \rangle^\gamma\right) \tag{A14}$$

One should be careful to note that while the volume sources in the closure problem can be neglected relative to the surface source, this is not necessarily true in the volume averaged transport equation. Simplification of these two equations must be carried out separately and one can not make the generic statement that a particular term is *small*.

At this point we are ready to return to the development in Sec. 2 and justify the simplification represented by Eq. 2.27. We begin by *defining* the length scale associated with the porosity by

$$\nabla \varepsilon_\gamma = \mathbf{O}\left(\frac{\varepsilon_\gamma}{L_\varepsilon}\right) \tag{A15}$$

and then estimating the left hand side of Eq. 2.27 as

$$(\nabla \varepsilon_\gamma) \cdot D_p \nabla \langle n_p \rangle^\gamma = \mathbf{O}\left(\frac{\varepsilon_\gamma D_p \langle n_p \rangle^\gamma}{L_\varepsilon L_n}\right) \tag{A16}$$

In order to estimate the right hand side of Eq. 2.27, we note that the area per unit volume can be expressed as

$$\frac{A_{\gamma\sigma}}{\mathscr{V}} = \mathbf{O}\left(\frac{\ell_\sigma^2}{\ell_\gamma^3}\right) \tag{A17}$$

and make use of Eqs. A7 and A14 to obtain

$$\frac{1}{\mathscr{V}} \int_{A_{\gamma\sigma}} \mathbf{n}_{\gamma\sigma} \cdot D_p \nabla \tilde{n}_p dA = \mathbf{O}\left(\frac{\ell_\sigma^2 D_p \langle n_p \rangle^\gamma}{\ell_\gamma^3 \ell_{bl}}\right) \tag{A18}$$

Use of Eqs. A16 and A18 in Eq. 2.27 leads to the inequality

$$\frac{\ell_\gamma^3 \ell_{bl}}{\ell_\sigma^2 L_\varepsilon L_n} \ll 1 \tag{A19}$$

and one can make use of Eq. A12 in order to express this result as

$$\frac{\ell_\gamma^3 Pe^{\frac{1}{3}}}{\ell_\sigma L_\varepsilon L_n} \ll 1 \tag{A20}$$

For homogeneous filters $L_\varepsilon \to \infty$ and this constraint is automatically satisfied. Real filters are heterogeneous, thus L_ε will have some finite value; however, representative values of the parameters that appear in Eq. A20 indicate that this constraint is generally satisfied for cases of practical interest.

APPENDIX B
EFFECTIVE COEFFICIENTS

In Sec. 4 we described the velocity fields that were calculated on the basis of the spatially periodic model shown in Figure 3, and we presented solutions for the effective capture coefficient, k_{eff}. Cellular efficiencies were calculated on the basis of Eq. 4.12 which neglects the contribution of the non-traditional "velocity-like" terms, **u** and **d**, and the dispersive transport characterized by the dispersion tensor, $\mathbf{D}^*$. The values of **u** and **d** are determined by Eqs. 3.25 and 3.26 and calculated values of the x-components of **u** and **d**, made dimensionless by the magnitude of $\langle \mathbf{v} \rangle$, are shown in Figures B1 and B2 as a function of the interstitial velocity for the conditions listed in Table 1. The closure problems were solved using numerical methods similar to those described in Quintard and Whitaker (1993, 1994b) The Stokes number is calculated on the basis of Eq. 4.5 and in Figures B1 and B2 we have shown results for the actual Stokes number based on the data listed in Table 1 and the interstitial velocity, along with results for which the Stokes number is arbitrarily set equal to zero. The results presented in Figures B1 and B2 indicate that **u** and **d** provide a contribution to the convective transport that is non-negligible, and should be taken into account in a thorough analysis of the filtration process. The values for a Stokes number of zero represent the purely diffusive case ($\gamma^{-1} = 0$ in Eq. 1.8) and the *increase* in the convective transport for this case occurs because the slow moving particles near the solid surface are removed thus leading to a larger overall velocity (Paine et al., 1983). For the finite Stokes numbers associated with the conditions listed in Table 1, we see that the *sign of the effect changes* for a velocity of about 0.05 m/s when $d_p = 0.1$ μm, and for a velocity of about 0.01 m/s when $d_p = 0.5$ μm. For the larger particle size the effect itself becomes very significant when the fluid velocity is greater that 0.01 m/s. The *diminished* convective transport at larger Stokes numbers presumably results from the fact that the high velocity particles are more effectively removed by inertial capture and this leads to a reduced convective transport. At the highest velocity represented in Figure B2, we see an abrupt change in the behavior of **u** and **d** and this may be due to numerical difficulties or due to the fact that at the larger Stokes numbers we need to include more of the terms in the expansion indicated in Eq. 1.7.

Figure B1
Velocity Coefficients as a Function of the Interstitial Fluid Velocity
$(d_p = 0.1\,\mu m, \quad 2a = 0.1\mu m)$

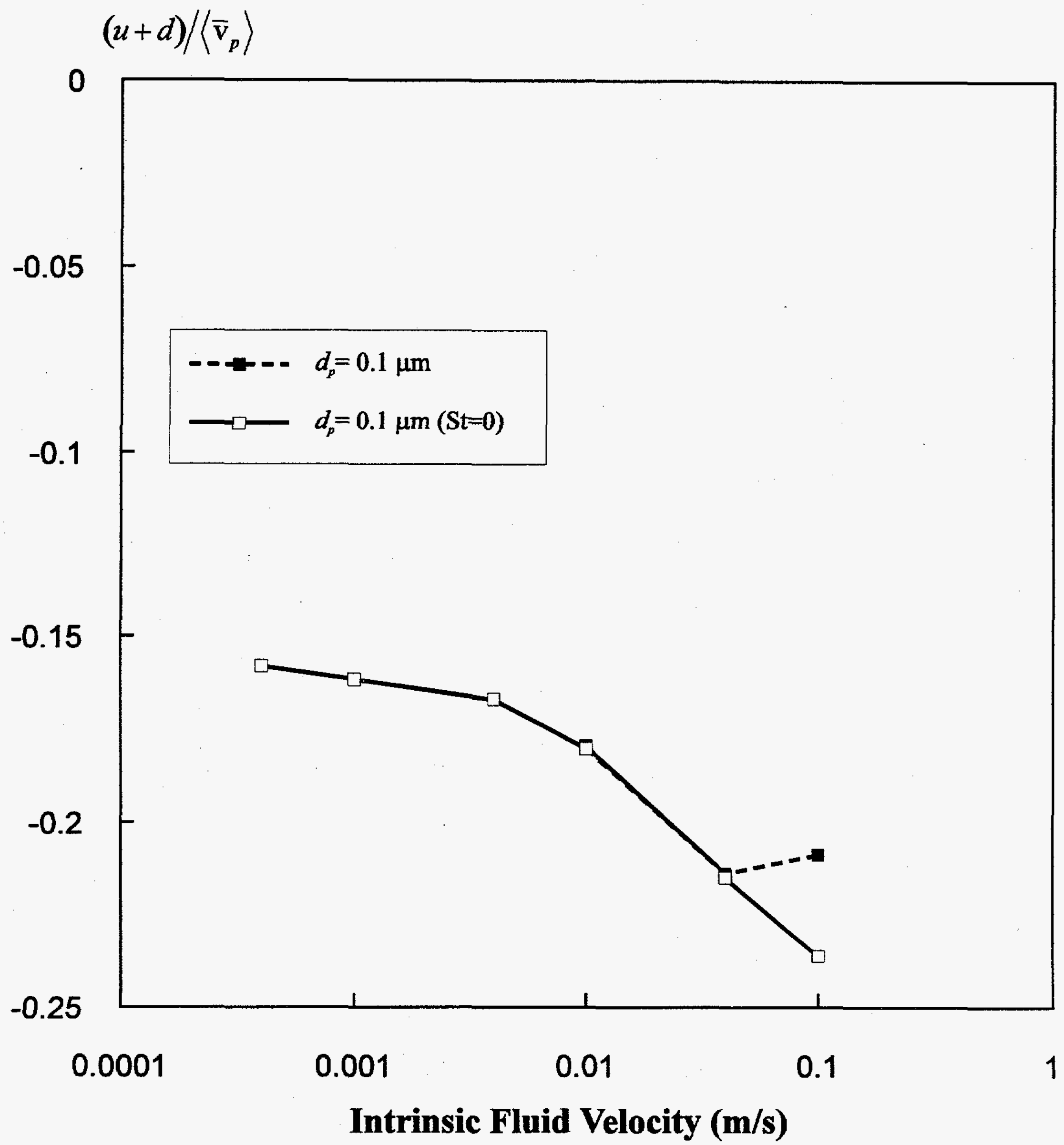
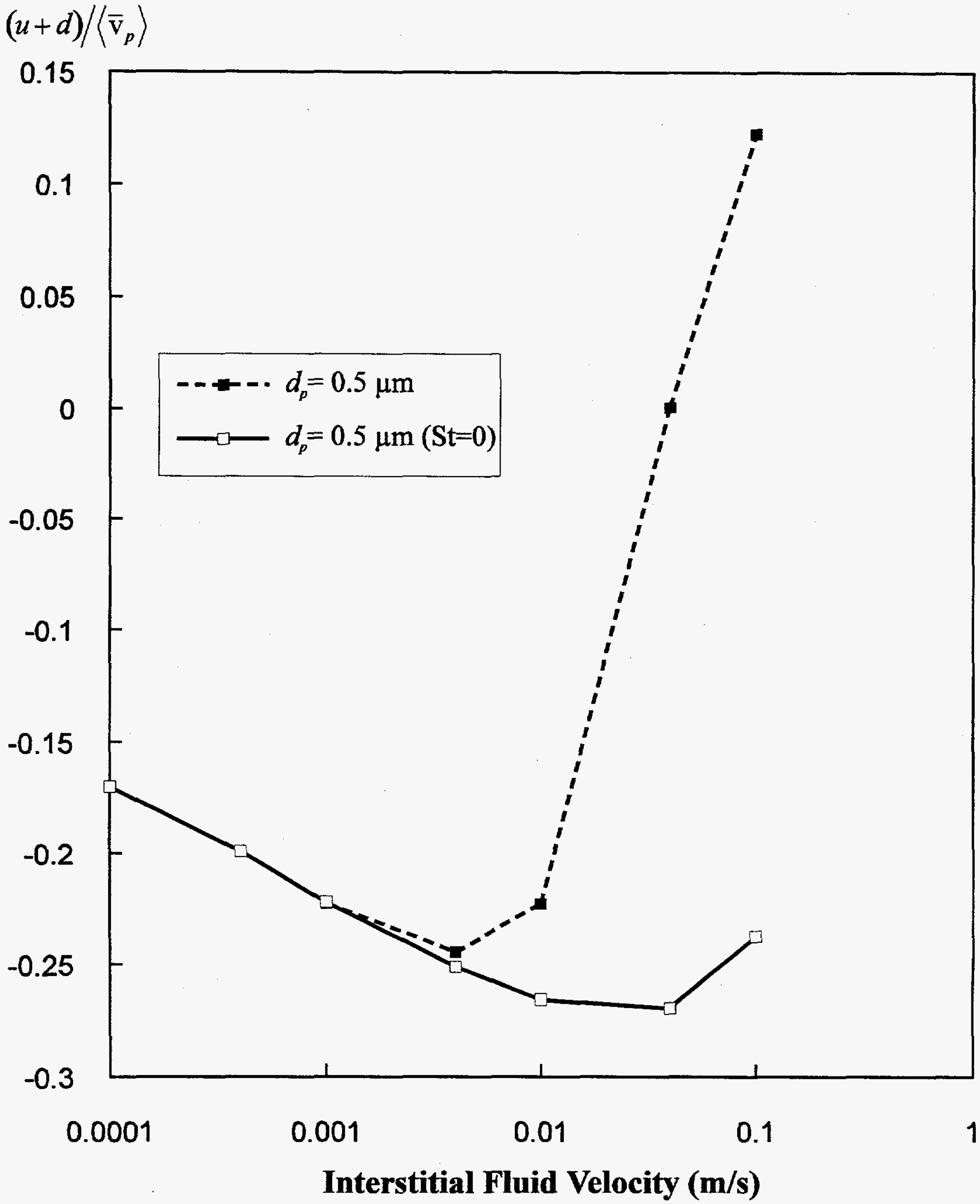


Figure B2
Velocity Coefficients as a Function of the Interstitial Fluid Velocity
(d_p = 0.5 μm, $2a$ = 0.5 μm)



In general, one can neglect dispersive transport relative to convective transport for many one-dimensional transport processes, and this is especially true for homogeneous media. In subsequent studies Eq. 4.6 will be used to develop a large-scale averaged transport equation for heterogeneous filters and in that case the dispersive transport may play an important role. Calculated results for the longitudinal dispersion coefficient, made dimensionless by the Brownian diffusivity, are shown in Figures B3 and B4 for the physical properties listed in Table 1 and a range of interstitial velocities. Once again, the case for $St = 0$ represents the diffusive case and the results are representative of the original calculations of Eidsath et al. (1983) and the more recent calculations of Sahroui. and Kaviany (1994) and of Quintard and Whitaker (1994b). For the finite Stokes numbers the dispersion coefficient diminishes slightly at the higher velocities for d_p = 0.5 μm and no influence of particle inertia is seen for d_p = 0.1 μm. The results shown in Figures B3 and B4 suggest that the classical dispersion coefficients obtained for species having negligible inertia represent an upperbound for the longitudinal dispersion of aerosol particles.

Figure B3
Longitudinal Dispersion Coefficients

(d_p = 0.5 μm, $2a$ = 0.5 μm, ρ_p = 4000 kg/m^3
T=283 K, μ=1.8 10^{-5} Pa s, ε_γ=0.95)

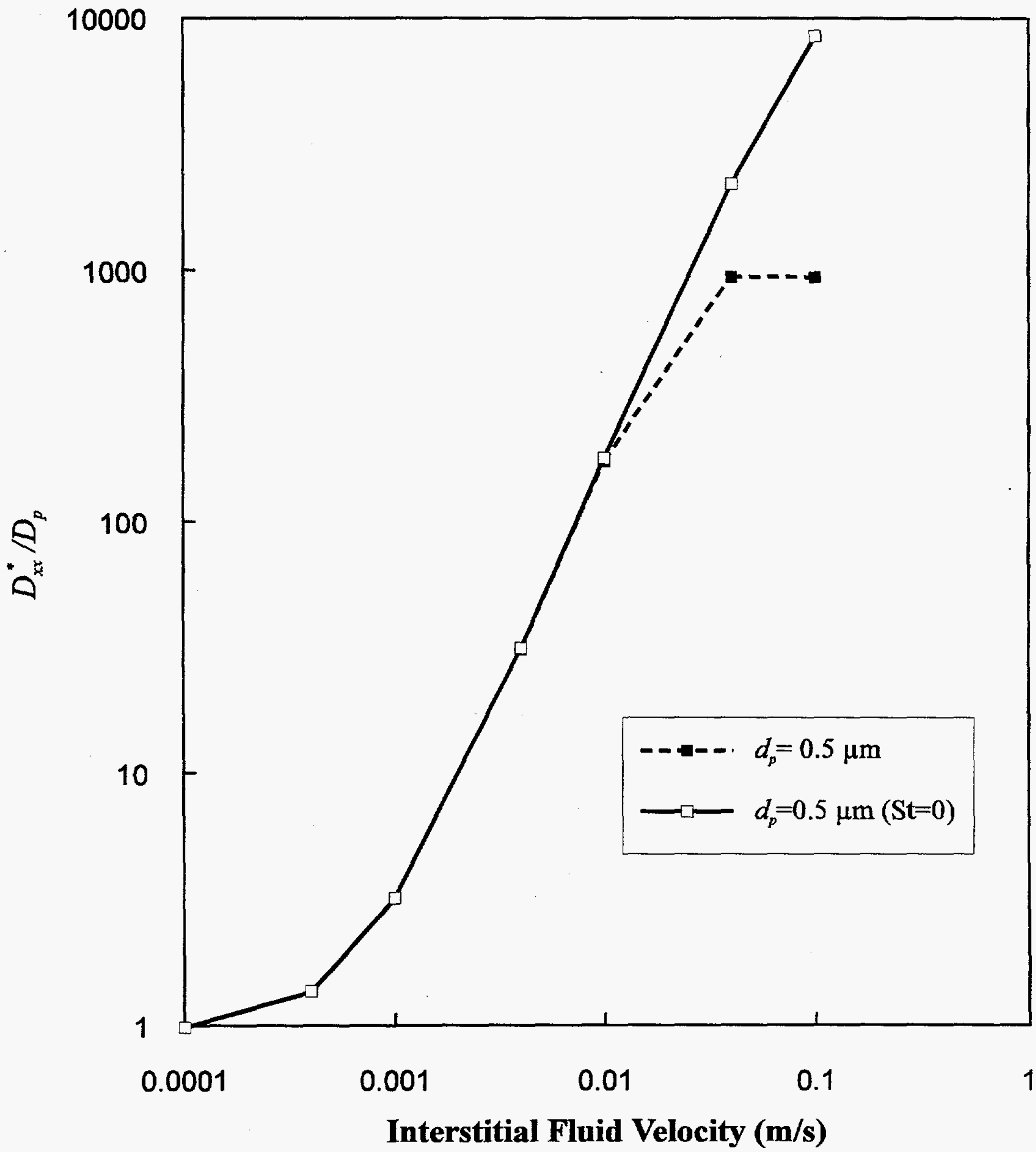


Figure B4
Longitudinal Dispersion Coefficients

(d_p = 0.1 μm, $2a$ = 0.1 μm, ρ_p = 4000 kg/m^3
T=283 K, μ=1.8 10^{-5} Pa s, ε_γ=0.95)