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One-dimensional virus transport in homogeneous porous media with time-dependent distribution coefficient

Constantinos V. Chrysikopoulos*, Youn Sim

Department of Civil and Environmental Engineering, University of California, Irvine, CA 92697, USA

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Abstract

A stochastic model for one-dimensional virus transport in homogeneous, saturated, semi-infinite porous media is developed. The model accounts for first-order inactivation of liquid-phase and adsorbed viruses with different inactivation rate constants, and time-dependent distribution coefficient. It is hypothesized that the virus adsorption process is described by a local equilibrium expression with a stochastic time-dependent distribution coefficient. A closed form analytical solution is obtained by the method of small perturbation or first-order approximation for a semi-infinite porous medium with a flux-type inlet boundary condition. The results from several simulations indicate that a time-dependent distribution coefficient results in an enhanced spreading of the liquid-phase virus concentration.

1. Introduction

Groundwater contamination by pathogenic bacteria and viruses has long been recognized as a serious hazard to human health (Keswick and Gerba, 1980). Most of the microorganisms in groundwater originate from human and animal sewage from nearby municipal wastewater discharges, septic tanks, sanitary landfills and agricultural practices. As microorganisms are released into the subsurface environment, they infiltrate through the vadose zone, and upon reaching the water table continue to migrate downstream (see Fig. 1). As groundwater is often consumed without prior conventional water treatment, or after inadequate treatment, it is necessary to understand fully the mechanisms governing the transport and fate of these microorganisms in groundwater systems so that the health risk owing to groundwater pollution by viruses can be evaluated. Mathematical models are frequently used as tools for prediction of the movement of viruses in the subsurface and

* Corresponding author.

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Notation

a	time correlation scale of K_d' (t)
A	defined in eqn (12)
B	defined in eqn (12)
C	liquid-phase virus concentration (suspended) $(M L^{-3})$
C_o	source concentration $(M L^{-3})$
C^*	sorbed virus concentration (virus mass/solid mass) $(M M^{-1})$
C'	zero mean random fluctuation of $< C >$ $(M L^{-3})$
D	hydrodynamic dispersion coefficient $(L^2 t^{-1})$
$\hat{D}$	defined in eqn (16)
erfc[x]	complementary error function, equal to $(2/\pi^{1/2}) \int_x^\infty \exp(-z^2)dz$
$E[]$	expectation operator
F	fundamental solution
G	defined in eqn (11)
h	defined in eqn (30)
H	defined in eqn (12)
K	hydraulic conductivity $(L t^{-1})$
K_d	partition or distribution coefficient $(L^3 M^{-1})$
K_d'	zero-mean random fluctuation of $\langle K_d \rangle$ $(L^3 M^{-1})$
Q	defined in eqn (27)
t	time (t)
U	average interstitial velocity $(L t^{-1})$
$\hat{U}$	defined in eqn (16b)
x	spatial coordinate in the longitudinal direction (L)
$\langle\rangle$	expected value: $E[]$
Greek letters	
γ	defined in eqn (13)
$\delta()$	Dirac delta function
ε	mathematical artifice (scalar)
θ	porosity (liquid volume/porous medium volume) $(L^3 L^{-3})$
κ	defined in eqn (23)
λ	inactivation rate constant of liquid-phase viruses (t^{-1})
λ^*	inactivation rate constant of adsorbed viruses (t^{-1})
$\hat{\lambda}$	defined in eqn (16)
Λ	defined in eqn (23)
ξ	dummy integration variable
ρ	bulk density of the solid matrix (solids mass/aquifer volume) $(M L^{-3})$
$\sigma^2_{K_d'}$	variance of K_d'
τ	dummy integration variable
ψ	defined in eqn (13)
ω	defined in eqn (23)

evaluation of long-term health risks, by determining safe distances between drinking water wells and sources of contamination (Yates et al., 1987).

Viruses are intracellular parasites that can be classified as colloid particles with size ranging from 0.02 μm to 0.3 μm (Brock and Madigan, 1991). They are generally negatively charged and vary widely in shape and chemical composition. A virus contains a nucleic acid, either DNA or RNA, which is surrounded by a protein coat (capsid) consisting of a number of protein molecules. These molecules are called capsomeres

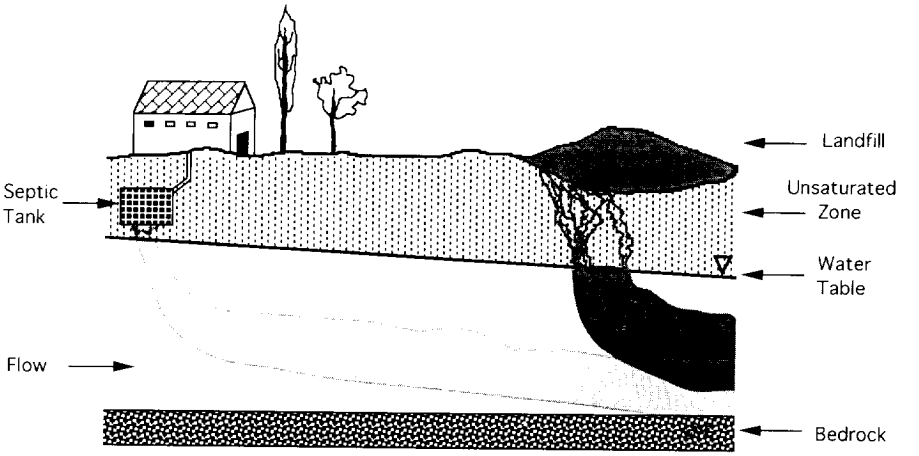

Fig. 1. Schematic illustration of sources and patterns of migration of microorganisms in the subsurface.

and they are arranged in a precise and highly repetitive pattern around nucleic acid. Because viruses do not have their own respiratory and biosynthetic functions, they reproduce inside other cells by a process called infection. Viruses are classified on the basis of the hosts they infect. The three major groups of viruses are animal viruses, plant viruses, and bacterial viruses (Brock and Madigan, 1991). The most common types of viruses found in groundwater which may infect human body are animal viruses such as adenovirus, coliphage, coxsackievirus, enterovirus, hepatitis, poliovirus and rotavirus (Gerba and Keswick, 1981; Yates and Yates, 1988).

The transport and fate of viruses in porous media are mainly governed by virus inactivation and adsorption onto the solid matrix (Vilker, 1981). Virus transport in porous media is distinguished from solute transport because viruses undergo considerably different inactivation and adsorption mechanisms. Inactivation of liquid-phase as well as sorbed or attached viruses is an irreversible sink mechanism, owing to disruption of coat proteins and degradation of the nucleic acid, which is commonly described by a first-order rate expression (Yates and Yates, 1988). Unlike the case of solute decay, experimental observations suggest the inactivation rate is smaller for attached than for liquid-phase viruses (Hurst et al., 1980; Gerba, 1984; Yates and Yates, 1988). Sobsey et al. (1980), Gerba (1984), and Yates et al. (1987) indicated that there exists a strong correlation between virus adsorption and inactivation. They showed that virus survival is prolonged for viruses adsorbed onto the solid matrix, because they are protected against disruption of coat protein and degradation of nucleic acid. Thus, inactivation rates of liquid-phase and attached viruses should not be assumed equal. The most important factor for virus inactivation in the subsurface is temperature (Yates and Ouyang, 1992). Viruses remain infective much longer at lower temperatures (1–8°C) than at higher temperatures (20–32°C) (Park et al., 1992). Therefore, near the top layer of an unsaturated subsurface formation, where considerable temperature fluctuations may occur, it is important to account for virus inactivation variation owing to temperature fluctuations. Although viruses may undergo sorption via physical adsorption, chemical adsorption or ion

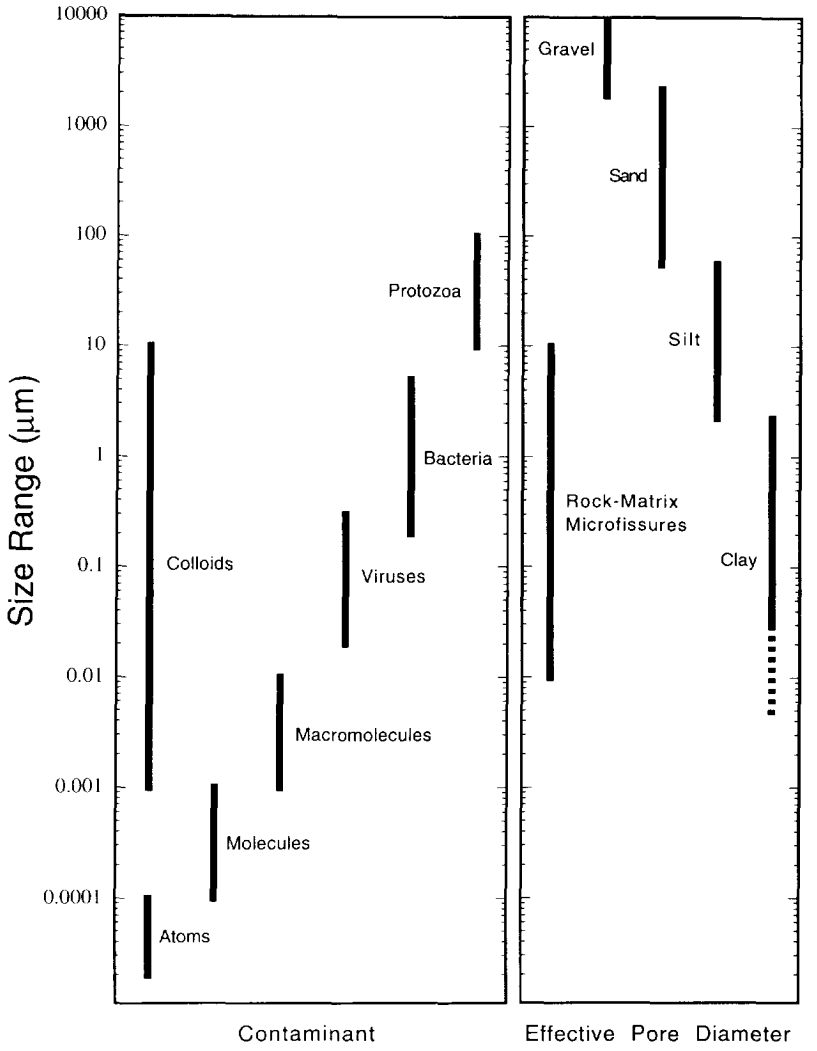

Fig. 2. Size ranges of contaminants present in groundwater and effective pore diameters of various porous media.

exchange in a fashion similar to solute adsorption, the major mechanism of virus attachment results from electrostatic double-layer interactions and Van der Waals forces (Teutsch et al., 1991). As illustrated in Fig. 2, viruses are larger than dissolved contaminants; however, it should be noted that viruses are at the lower end of the colloid size distribution. For this reason, virus adsorption is often described by either colloid filtration or solute sorption processes. For example, Sim and Chrysikopoulos (1995) derived analytical solutions to two deterministic mathematical models for virus transport in one-dimensional homogeneous, saturated porous media, accounting for first-order inactivation of liquid-phase and adsorbed viruses with different inactivation rate constants, and either nonequilibrium reversible virus adsorption (applicable to viruses behaving as

solutes) or virus filtration (suitable for viruses behaving as colloids). The size of viruses can be an important parameter if the porous medium is highly heterogeneous, where viruses may move faster than conservative tracers (Powelson and Gerba, 1994). This phenomenon is known as the pore size exclusion effect, caused by preferential transport of colloids through pores larger than their physical sizes. Consequently, pore size exclusion enhances colloid migration (Gerba et al., 1991; Powelson et al., 1993; Abdel-Salam and Chrysikopoulos, 1995).

Virus adsorption in homogeneous porous media is commonly described by an equilibrium adsorption relationship assuming an instantaneous equilibrium between viruses in the liquid-phase and onto the solid matrix. For the case of a linear isotherm (linear relationship between the amount of a virus in the liquid-phase and sorbed onto the solid matrix) the extent of equilibrium mass partitioning is often reflected by a constant partition coefficient or distribution coefficient, which is equal to the slope of the linear sorption isotherm.

Several mathematical models are available in the literature for virus transport in porous media, which account for a linear equilibrium adsorption with a constant distribution coefficient. Grosser (1984) employed a one-dimensional advection–dispersion equation to describe virus transport in homogeneous porous media under local equilibrium conditions assuming equal inactivation rates for both adsorbed and liquid-phase viruses. Tim and Mostaghimi (1991) developed a numerical model for water flow and virus transport in variably saturated formations assuming that virus adsorption is an equilibrium process, and virus inactivation is identical for adsorbed as well as liquid-phase viruses. Park et al. (1992) developed a semi-analytical–numerical model (VIRALT) for both steady-state and transient vertical virus transport in the unsaturated zone and along the flowlines in the saturated zone, accounting for equilibrium adsorption and inactivation. Matthess et al. (1988) presented a model accounting for equilibrium virus adsorption, inactivation, and filtration. Yates and Ouyang (1992) developed a one-dimensional numerical model (VIRTUS) that couples the flow of water, viruses, and heat in unsaturated porous media and accounts for equilibrium adsorption, filtration, and temperature-dependent inactivation.

Recent investigations suggest that the distribution coefficient for a contaminant in a physicochemically heterogeneous subsurface formation is not constant but exhibits temporal as well as spatial variability (Durant and Roberts, 1986; Bosma et al., 1993; Smith et al., 1993). Excluding the possibilities of mass transport limitations and solute transformation, this variability may be attributed to many factors, including grain size and surface area of adsorbent, chemical composition of groundwater, pH, redox potential, temperature, and solid to liquid ratio (Moody, 1982). Proteins (primary constituents of viruses) are also known to exhibit sorption variations with fluctuating external conditions (Norde, 1986). In principle, the results obtained from solutes and proteins can be extended to viruses. Therefore, it would be reasonable to consider virus adsorption as a time-dependent process, and consequently the distribution coefficient as a time-dependent parameter.

Time- and space-dependent adsorption has been observed experimentally and investigated theoretically in many solute transport studies. Chrysikopoulos et al. (1990) developed an analytical stochastic solute transport model for one-dimensional homogeneous

porous media to demonstrate that spatially variable retardation increases solute spreading. Kookana et al., 1992a,b) observed time-dependent pesticide sorption, where the variability was attributed to the physical heterogeneity of the soil medium. Chrysikopoulos et al., 1992a,b) employed the generalized Taylor–Aris–Brenner moment analysis to investigate the observed increase in solute spreading caused by the spatial variability of the retardation factor and the hydrodynamic parameters. Bellin et al. (1993) and Bosma et al. (1993) showed that spatially variable adsorption influences significantly the spreading and movement of reactive solutes in physicochemically heterogeneous porous formations. Burr et al. (1994) considered a three-dimensional numerical model for reactive and non-reactive solute transport in statistically anisotropic porous media with spatially variable distribution coefficient to show that the retardation factor increases with time and plume displacement distance.

The present work focuses on a virus transport model that accounts for virus inactivation and linear local equilibrium adsorption with a stochastic time-dependent distribution coefficient. The analytical solution is derived by the method of small perturbation (or first-order approximation), which has been employed in numerous groundwater (e.g. Bear and Dagan, 1964; Dagan, 1985) and solute transport investigations (e.g. Gelhar and Axness, 1983; Chrysikopoulos et al., 1990).

2. Model development

2.1. Transport model

The one-dimensional virus transport, in homogeneous, saturated, but geochemically heterogeneous porous media, accounting for virus adsorption and inactivation, is governed by the following partial differential equation (Sim and Chrysikopoulos, 1996):

$$\frac{\partial C(t,x)}{\partial t}+\frac{\rho}{\theta}\frac{\partial C^*(t,x)}{\partial t}=D\frac{\partial^2 C(t,x)}{\partial x^2}-U\frac{\partial C(t,x)}{\partial x}-\lambda C(t,x)-\lambda^*\frac{\rho}{\theta}C^*(t,x) \tag{1}$$

where C is the liquid-phase virus concentration, C^* is the mass of virus adsorbed on the solid matrix, D is the hydrodynamic dispersion coefficient, U is the average interstitial velocity, ρ is the bulk density of the solid matrix, λ is the inactivation rate constant of liquid-phase viruses, λ^* is the inactivation rate constant of adsorbed viruses, θ is the porosity of soil medium, and t is time. The left-hand side of Eq. (1) consists of the virus accumulation terms, and the last two terms on the right-hand side represent the inactivation of liquid-phase and adsorbed viruses, respectively.

The appropriate expression for time-dependent local equilibrium virus adsorption is

$$C^*(t,x)=K_d(t)C(t,x) \tag{2}$$

where $K_d(t)$ is the time-dependent distribution coefficient; then

$$\frac{\partial C^*(t,x)}{\partial t}=C(t,x)\frac{\partial K_d(t)}{\partial t}+K_d(t)\frac{\partial C(t,x)}{\partial t} \tag{3}$$

In view of Eq. (2) and Eq. (3), Eq. (1) can be written as

$$\left[1+\frac{\rho}{\theta}K_d(t)\right]\frac{\partial C(t,x)}{\partial t}+C(t,x)\frac{\rho}{\theta}\frac{\partial K_d(t)}{\partial t}=D\frac{\partial^2 C(t,x)}{\partial x^2}-U\frac{\partial C(t,x)}{\partial x}-\lambda C(t,x)$$
$$-\lambda^*\frac{\rho}{\theta}K_d(t)C(t,x) \tag{4}$$

For a one-dimensional, semi-infinite porous medium in the presence of a continuous source of viruses, the appropriate initial and boundary conditions are

$$C(0,x)=0 \tag{5a}$$

$$-D\frac{\partial C(t,0)}{\partial x}+UC(t,0)=UC_0 \tag{5b}$$

$$\frac{\partial C(t,\infty)}{\partial x}=0 \tag{5c}$$

where C_0 is the source concentration. The condition Eq. (5) establishes that there is no initial virus concentration within the one-dimensional porous medium. The constant flux boundary condition Eq. (5) implies virus concentration discontinuity at the inlet. The downstream boundary condition Eq. (5) preserves concentration continuity for a semi-infinite system.

The distribution coefficient and, consequently, the virus concentration are considered to be stochastic processes. In the absence of experimental evidence a conservative approach is to assume that the stochastic distribution coefficient is stationary with mean $\langle K_d\rangle = E[K_d(t)]$, where the angle brackets signify ensemble average or expected value over time of a random process. The concentration is both nonstationary and space dependent. Therefore, the liquid-phase virus concentration and the time-dependent distribution coefficient are expressed as

$$C(t,x)=\langle C\rangle(t,x)+C'(t,x) \tag{6a}$$

$$K_d(t)=\langle K_d\rangle+K_d'(t) \tag{6b}$$

where $\langle C\rangle(t,x)$ is the concentration mean, the prime signifies fluctuations in time, $E[C'(t,x)]=E[K_d'(t)]=0$. Substituting Eq. (6) and Eq. (6) into the governing Eq. (4) and conditions eqns (5), (5), () and (5) yields

$$\left(1+\frac{\rho}{\theta}[\langle K_d\rangle+K_d'(t)]\right)\left(\frac{\partial\langle C\rangle(t,x)}{\partial t}+\frac{\partial C'(t,x)}{\partial t}\right)$$
$$+\frac{\rho}{\theta}(\langle C\rangle(t,x)+C'(t,x))\left(\frac{\partial\langle K_d\rangle}{\partial t}+\frac{\partial K_d'(t)}{\partial t}\right)$$
$$=D\left(\frac{\partial^2\langle C\rangle(t,x)}{\partial x^2}+\frac{\partial^2 C'(t,x)}{\partial x^2}\right)-U\left(\frac{\partial\langle C\rangle(t,x)}{\partial x}+\frac{\partial C'(t,x)}{\partial x}\right)$$
$$-\lambda(\langle C\rangle(t,x)+C'(t,x))-\lambda^*\frac{\rho}{\theta}(\langle K_d\rangle+K_d'(t))(\langle C\rangle(t,x)+C'(t,x)) \tag{7}$$

$$\langle C\rangle(0,x)+C'(0,x)=0 \tag{8a}$$

$$-D\left(\frac{\partial\langle C\rangle(t,0)}{\partial x}+\frac{\partial C'(t,0)}{\partial x}\right)+U(\langle C\rangle(t,0)+C'(t,0))=UC_0 \tag{8b}$$

$$\frac{\partial\langle C\rangle(t,\infty)}{\partial x}+\frac{\partial C'(t,\infty)}{\partial x}=0 \tag{8c}$$

It should be noted that the derivative of $\langle K_d\rangle$ with respect to time is zero because $K_d(t)$ is stationary. Taking the ensemble averages of all terms in Eq. (7) and Eqs. (8a)–(8c) yields the stochastic partial differential equation and initial-boundary conditions for the ensemble averaged virus concentration

$$\begin{aligned}&\left[1+\frac{\rho}{\theta}\langle K_d\rangle\right]\frac{\partial\langle C\rangle(t,x)}{\partial t}+G(t,x)\\&=D\frac{\partial^2\langle C\rangle(t,x)}{\partial x^2}-U\frac{\partial\langle C\rangle(t,x)}{\partial x}-\left[\lambda+\lambda^*\frac{\rho}{\theta}\langle K_d\rangle\right]\langle C\rangle(t,x)\end{aligned} \tag{9}$$

$$\langle C\rangle(0,x)=0 \tag{10a}$$

$$-D\frac{\partial\langle C\rangle(t,0)}{\partial x}+U\langle C\rangle(t,0)=UC_0 \tag{10b}$$

$$\frac{\partial\langle C\rangle(t,\infty)}{\partial x}=0 \tag{10c}$$

where the function $G(t,x)$ represents the effect of stochastic fluctuations of the liquid-phase virus concentration and the distribution coefficient. This function is defined as follows:

$$G(t,x)=A(t,x)+B(t,x)+H(t,x) \tag{11}$$

$$A(t,x)=\frac{\rho}{\theta}\left\langle K_d'(t)\frac{\partial C'(t,x)}{\partial t}\right\rangle \tag{12a}$$

$$B(t,x)=\frac{\rho}{\theta}\left\langle C'(t,x)\frac{\partial K_d'(t)}{\partial t}\right\rangle \tag{12b}$$

$$H(t,x)=\lambda^*\frac{\rho}{\theta}\langle K_d'(t)C'(t,x)\rangle \tag{12c}$$

The desired solution of the transport model defined by Eq. (9) and Eqs (10a)–(10c) is the expression for the liquid-phase virus concentration mean $\langle C\rangle(t,x) = E[C(t,x)]$.

2.2. General solution

Assuming that $G(t,x)$ is a given function of t and x, Eq. (9) subject to conditions Eqs. (10a)–(10c) is solved analytically following the methods presented by Ito (1957a,b) and Chrysikopoulos et al. (1990), and is given by

$$\langle C\rangle(t,x) = \int_0^t d\tau \int_0^\infty F(t-\tau,x,\xi)\psi(\tau,\xi)d\xi + \int_0^t F(t-\tau,x,0)\gamma(\tau,0)d\tau \tag{13a}$$

where

$$\psi(t,x) = \frac{-G(t,x)}{\left[1+\frac{\rho}{\theta}\langle K_d\rangle\right]} \tag{13b}$$

$$\gamma(t,0) = \frac{UC_o}{\left[1+\frac{\rho}{\theta}\langle K_d\rangle\right]} \tag{13c}$$

and $F(t,x,\xi)$ is the fundamental solution of Eq. (9).

The fundamental solution, $F(t,x,\xi)$, is the solution to the homogeneous form of Eq. (9) subject to a point source initial condition and homogeneous boundary conditions as follows:

$$\frac{\partial F(t,x,\xi)}{\partial t} = \hat{D}\frac{\partial^2 F(t,x,\xi)}{\partial x^2} - \hat{U}\frac{\partial F(t,x,\xi)}{\partial x} - \hat{\lambda}F(t,x,\xi) \tag{14}$$

$$F(0,x,\xi) = \delta(x-\xi) \tag{15a}$$

$$-\hat{D}\frac{\partial F(t,0,\xi)}{\partial x} + \hat{U}F(t,0,\xi) = 0 \tag{15b}$$

$$\frac{\partial F(t,\infty,\xi)}{\partial x} = 0 \tag{15c}$$

where

$$\hat{D} = \frac{D}{\left[1+\frac{\rho}{\theta}\langle K_d\rangle\right]} \tag{16a}$$

$$\hat{U} = \frac{U}{\left[1+\frac{\rho}{\theta}\langle K_d\rangle\right]} \tag{16b}$$

$$\hat{\lambda} = \frac{\left[\lambda+\lambda^*\frac{\rho}{\theta}\langle K_d\rangle\right]}{\left[1+\frac{\rho}{\theta}\langle K_d\rangle\right]} \tag{16c}$$

Taking Laplace transforms of Eq. (14) and Eqs. (15a)–(15c) with respect to time variable t and space variable x, using the transformed boundary conditions and applying inverse

transformations yields the desired fundamental solution

$$F(t,x,\xi)=\left(\frac{1}{4\hat{D}\pi t}\right)^{1/2}\exp\left[\frac{\hat{U}(x-\xi)}{2\hat{D}}-\frac{(x-\xi)^2}{4\hat{D}t}-\left(\hat{\lambda}+\frac{\hat{U}^2}{4\hat{D}}\right)t\right]$$
$$+\left(\frac{1}{4\hat{D}\pi t}\right)^{1/2}\exp\left[\frac{\hat{U}(x-\xi)}{2\hat{D}}-\frac{(x+\xi)^2}{4\hat{D}t}-\left(\hat{\lambda}+\frac{\hat{U}^2}{4\hat{D}}\right)t\right] \tag{17}$$
$$-\left(\frac{\hat{U}}{2\hat{D}}\right)\exp\left[\frac{\hat{U}x}{\hat{D}}-\hat{\lambda}t\right]erfc\left[\left(\frac{\hat{U}^2t}{4\hat{D}}\right)^{1/2}+\frac{x+\xi}{(4\hat{D}t)^{1/2}}\right]$$

2.3. Derivation of $G(t,x)$

The nonhomogeneous term, $G(t,x)$, is evaluated by a small perturbation approximation. Assuming that the fluctuation terms in Eqs. (6a) and (6b) are small, a dimensionless scalar, ε, is introduced to keep track of the small terms. The mean values and fluctuations are expressed as follows:

$$\langle K_d\rangle=\varepsilon^0\langle K_{d0}\rangle \tag{18a}$$

$$K_d'(t)=\varepsilon^1 K_{d1}'(t) \tag{18b}$$

$$\langle C\rangle(t,x)=\varepsilon^0\langle C_0\rangle(t,x)+\varepsilon^1\langle C_1\rangle(t,x)+\cdots \tag{19a}$$

$$C'(t,x)=\varepsilon^1 C_1'(t,x)+\cdots \tag{19b}$$

where the subscript zero indicates zero-order terms, the subscript one first-order terms, etc. It should be noted that only a zero-order perturbation is performed for the mean distribution coefficient, because $K_d(t)$ is assumed stationary. Substituting Eqs. (18a) and (18b) and Eqs. (19a) and (19b) into the governing Eq. (7) yields

$$\left(1+\frac{\rho}{\theta}\left[\varepsilon^0\langle K_{d0}\rangle+\varepsilon^1 K_{d1}'(t)\right]\right)\left(\varepsilon^0\frac{\partial\langle C_0\rangle(t,x)}{\partial t}+\varepsilon^1\frac{\partial\langle C_1\rangle(t,x)}{\partial t}+\varepsilon^1\frac{\partial C_1'(t,x)}{\partial t}\right)$$
$$+\frac{\rho}{\theta}\left(\varepsilon^0\langle C_0\rangle(t,x)+\varepsilon^1\langle C_1\rangle(t,x)+\varepsilon^1 C_1'(t,x)\right)\left(\varepsilon^0\frac{\partial\langle K_{d0}\rangle}{\partial t}+\varepsilon^1\frac{\partial K_{d1}'(t)}{\partial t}\right)$$
$$=D\left(\varepsilon^0\frac{\partial^2\langle C_0\rangle(t,x)}{\partial x^2}+\varepsilon^1\frac{\partial^2\langle C_1\rangle(t,x)}{\partial x^2}+\varepsilon^1\frac{\partial^2 C_1'(t,x)}{\partial x^2}\right) \tag{20}$$
$$-U\left(\varepsilon^0\frac{\partial\langle C_0\rangle(t,x)}{\partial x}+\varepsilon^1\frac{\partial\langle C_1\rangle(t,x)}{\partial x}+\varepsilon^1\frac{\partial C_1'(t,x)}{\partial x}\right)$$
$$-\lambda\left(\varepsilon^0\langle C_0\rangle(t,x)+\varepsilon^1\langle C_1\rangle(t,x)+\varepsilon^1 C_1'(t,x)\right)$$
$$-\frac{\rho}{\theta}\lambda^*\left(\varepsilon^0\langle K_{d0}\rangle+\varepsilon^1 K_{d1}'(t)\right)\left(\varepsilon^0\langle C_0\rangle(t,x)+\varepsilon^1\langle C_1\rangle(t,x)+\varepsilon^1 C_1'(t,x)\right)$$

As the zero-order terms are linearly independent of the first-order terms, etc., the preceding equation must be satisfied separately for terms of each order. Equating coefficients of ε^0 into Eq. (20) yields

$$\left[1+\frac{\rho}{\theta}\langle K_{d0}\rangle\right]\frac{\partial\langle C_0\rangle(t,x)}{\partial t}=D\frac{\partial^2\langle C_0\rangle(t,x)}{\partial x^2}-U\frac{\partial\langle C_0\rangle(t,x)}{\partial x}-\lambda\langle C_0\rangle(t,x)-\frac{\rho}{\theta}\lambda^*\langle K_{d0}\rangle\langle C_0\rangle(t,x) \tag{21}$$

This is a deterministic advection–dispersion equation with sorption and decay terms. The analytical solution to this partial differential equation subject to initial and boundary conditions Eqs. (10a)–(10c) where $\langle C\rangle(t,x)$ is replaced by $\langle C_0\rangle(t,x)$, is easily obtained by Laplace transform techniques as

$$\begin{aligned}\langle C_0\rangle(t,x)=C_0\Bigg\{&\left[\frac{U}{U+\kappa}\right]\exp\left[\frac{x(U-\kappa)}{2D}\right]erfc\left[\frac{x\Lambda-\kappa t}{2(D\Lambda t)^{1/2}}\right]\\&+\left[\frac{U}{U-\kappa}\right]\exp\left[\frac{x(U+\kappa)}{2D}\right]erfc\left[\frac{x\Lambda+\kappa t}{2(D\Lambda t)^{1/2}}\right]\\&+\left[\frac{U^2}{2D\omega}\right]\exp\left[\frac{Ux}{D}-\frac{\omega t}{\Lambda}\right]erfc\left[\frac{x\Lambda+Ut}{2(D\Lambda t)^{1/2}}\right]\Bigg\}\end{aligned} \tag{22}$$

where the following substitutions have been employed:

$$\kappa=(U^2+4D\omega)^{1/2} \tag{23a}$$

$$\omega=\lambda+\lambda^*\frac{\rho}{\theta}\langle K_{d0}\rangle \tag{23b}$$

$$\Lambda=1+\frac{\rho}{\theta}\langle K_{d0}\rangle \tag{23c}$$

A slightly modified version of this solution can also be found in the compilation by Van Genuchten and Alves (1982, p. 61).

Equating coefficients of order ε^1 into Eq. (20) yields

$$\begin{aligned}&\frac{\rho}{\theta}K_{d1}'(t)\frac{\partial\langle C_0\rangle(t,x)}{\partial t}+\left[1+\frac{\rho}{\theta}\langle K_{d0}\rangle\right]\left(\frac{\partial\langle C_1\rangle(t,x)}{\partial t}+\frac{\partial C_1'(t,x)}{\partial t}\right)\\&=D\left(\frac{\partial^2\langle C_1\rangle(t,x)}{\partial x^2}+\frac{\partial^2 C_1'(t,x)}{\partial x^2}\right)-U\left(\frac{\partial\langle C_1\rangle(t,x)}{\partial x}+\frac{\partial C_1'(t,x)}{\partial x}\right)\\&\quad-\frac{\rho}{\theta}(\langle C_1\rangle(t,x)+C_1'(t,x))\frac{\partial\langle K_{d0}\rangle}{\partial t}-\frac{\rho}{\theta}\langle C_0\rangle(t,x)\frac{\partial K_{d1}'(t)}{\partial t}\\&\quad-\lambda(\langle C_1\rangle(t,x)+C_1'(t,x))-\frac{\rho}{\theta}\lambda^*\langle K_{d0}\rangle(\langle C_1\rangle(t,x)C_1'(t,x))\\&\qquad-\frac{\rho}{\theta}\lambda^*K_{d1}'(t)\langle C_0\rangle(t,x)\end{aligned} \tag{24}$$

Taking the expected values of all terms yields the following deterministic partial

differential equation:

$$\left[1+\frac{\rho}{\theta}\langle K_{d0}\rangle\right]\frac{\partial\langle C_1\rangle(t,x)}{\partial t}=D\frac{\partial^2\langle C_1\rangle(t,x)}{\partial x^2}-U\frac{\partial\langle C_1\rangle(t,x)}{\partial x}-\lambda\langle C_1\rangle(t,x)-\frac{\rho}{\theta}\lambda^*\langle K_{d0}\rangle\langle C_1\rangle(t,x) \tag{25}$$

which, subject to the homogeneous initial and boundary conditions obtained from Eqs. (5a)–(5c) by keeping first-order terms and taking expected values, has the trivial solution $\langle C_1\rangle(t,x) = 0$.

Subtracting Eq. (25) from Eq. (24) yields the following stochastic partial differential equation for the first-order fluctuation of liquid-phase virus concentration:

$$\begin{aligned}&\left[1+\frac{\rho}{\theta}\langle K_{d0}\rangle\right]\frac{\partial C_1'(t,x)}{\partial t}+Q(t,x)\\&=D\frac{\partial^2 C_1'(t,x)}{\partial x^2}-U\frac{\partial C_1'(t,x)}{\partial x}-\lambda C_1'(t,x)-\frac{\rho}{\theta}\lambda^*\langle K_{d0}\rangle C_1'(t,x)\end{aligned} \tag{26}$$

where the following substitution for the undetermined terms was employed:

$$Q(t,x)=\left[\frac{\rho}{\theta}K_{d1}'(t)\frac{\partial\langle C_0\rangle(t,x)}{\partial t}+\frac{\rho}{\theta}\langle C_0\rangle(t,x)\frac{\partial K_{d1}'(t)}{\partial t}+\frac{\rho}{\theta}\lambda^*K_{d1}'(t)\langle C_0\rangle(t,x)\right] \tag{27}$$

The appropriate initial and boundary conditions are

$$C_1'(0,x)=0 \tag{28a}$$

$$-D\frac{\partial C_1'(t,0)}{\partial x}+UC_1'(t,0)=0 \tag{28b}$$

$$\frac{\partial C_1'(t,\infty)}{\partial x}=0 \tag{28c}$$

Assuming that $Q(t,x)$ is a given function of t and x, the general solution of Eq. (26) subject to Eqs. (28a)–(28c) is given by (Ito, 1957a,b; Chrysikopoulos et al., 1990)

$$C_1'(t,x)=\int_0^t d\tau\int_0^\infty F(t-\tau,x,\xi)h(\tau,\xi)d\xi \tag{29}$$

where the fundamental solution $F(t,x,\tau)$ is presented in Eq. (17) (note that in view of Eq. (18a) $\langle K_d\rangle = \langle K_{d0}\rangle$), and

$$h(t,x)=\frac{-Q(t,x)}{\left[1+\frac{\rho}{\theta}\langle K_{d0}\rangle\right]} \tag{30}$$

It should be noted that the term $\partial\langle C_0\rangle(t,x)/\partial t$ in the definition of $Q(t,x)$, presented by Eq. (27), is obtained by differentiating Eq. (22) with respect to time to yield

$$\begin{aligned}\frac{\partial\langle C_0\rangle(t,x)}{\partial t} = C_o\Bigg\{&\left[\frac{U}{U+\kappa}\right]\left[\frac{x\Lambda+\kappa t}{2(D\Lambda\pi t^3)^{1/2}}\right]\exp\left[\frac{x(U-\kappa)}{2D}\right]\exp\left[\frac{-(x\Lambda-\kappa t)^2}{4D\Lambda t}\right]\\&+\left[\frac{U}{U-\kappa}\right]\left[\frac{x\Lambda-\kappa t}{2(D\Lambda\pi t^3)^{1/2}}\right]\exp\left[\frac{x(U+\kappa)}{2D}\right]\exp\left[\frac{-(x\Lambda+\kappa t)^2}{4D\Lambda t}\right]\\&+\left[\frac{U^2}{2D\omega}\right]\left[\frac{x\Lambda-Ut}{2(D\Lambda\pi t^3)^{1/2}}\right]\exp\left[\frac{Ux}{D}-\frac{\omega t}{\Lambda}\right]\exp\left[\frac{-(x\Lambda+Ut)^2}{4D\Lambda t}\right]\\&-\left[\frac{U^2}{2D\Lambda}\right]\exp\left[\frac{Ux}{D}-\frac{\omega t}{\Lambda}\right]erfc\left[\frac{x\Lambda+Ut}{2(D\Lambda t)^{1/2}}\right]\Bigg\}\end{aligned} \tag{31}$$

The first term, $A(t,x)$, of the desired function $G(t,x)$, defined in Eq. (11), can be obtained by employing Eqs. (18a) and (18b), Eqs. (19a) and (19b) and Eq. (29) into Eq. (12a) and using the linear property of the expectation operator to yield

$$\begin{aligned}A(t,x) &= \varepsilon^2\frac{\rho}{\theta}E\left[K_{d1}'(t)\frac{\partial}{\partial t}\left\{\int_0^t d\tau\int_0^\infty F(t-\tau,x,\xi)h(\tau,\xi)d\xi\right\}\right]\\&= \varepsilon^2\frac{\rho}{\theta}\int_0^t d\tau\int_0^\infty\frac{\partial F(t-\tau,x,\xi)}{\partial t}E[K_{d1}'(t)h(\tau,\xi)]d\xi-\varepsilon^2\frac{\rho}{\theta}E[K_{d1}'(t)h(t,x)]\end{aligned} \tag{32}$$

where Leibnitz's rule has been employed for differentiating the integral with respect to t. Similarly, the second component, $B(t,x)$, of the function $G(t,x)$ is given by

$$\begin{aligned}B(t,x) &= -\varepsilon^2\frac{\rho}{\theta}E\left[\frac{\partial K_{d1}'(t)}{\partial t}\int_0^t d\tau\int_0^\infty F(t-\tau,x,\xi)h(\tau,\xi)d\xi\right]\\&= -\varepsilon^2\frac{\rho}{\theta}\int_0^t d\tau\int_0^\infty F(t-\tau,x,\xi)E\left[\frac{\partial K_{d1}'(t)}{\partial t}h(\tau,\xi)\right]d\xi\end{aligned} \tag{33}$$

and the third component, $H(t,x)$, by

$$\begin{aligned}H(t,x) &= \varepsilon^2\frac{\rho}{\theta}\lambda^*E\left[K_{d1}'(t)\int_0^t d\tau\int_0^\infty F(t-\tau,x,\xi)h(\tau,\xi)d\xi\right]\\&= \varepsilon^2\frac{\rho}{\theta}\lambda^*\int_0^t d\tau\int_0^\infty F(t-\tau,x,\xi)E[K_{d1}'(t)h(\tau,\xi)]d\xi\end{aligned} \tag{34}$$

2.4. Evaluation of expected value terms

In view of Eq. (27) and Eq. (30), the averaged terms present in Eqs. (32)–(34) are

obtained as

$$E[K_{d1}'(t)h(\tau,\xi)] = -\frac{\rho}{\theta}\left[1+\frac{\rho}{\theta}\langle K_{d0}\rangle\right]^{-1}\left\{\frac{\partial\langle C_0\rangle(\tau,\xi)}{\partial\tau}E[K_{d1}'(t)K_{d1}'(\tau)] + \langle C_0\rangle(\tau,\xi)E\left[K_{d1}'(t)\frac{\partial K_{d1}'(\tau)}{\partial\tau}\right] + \lambda^*\langle C_0\rangle(\tau,\xi)E[K_{d1}'(t)K_{d1}'(\tau)]\right\} \tag{35}$$

$$E\left[\frac{\partial K_{d1}'(t)}{\partial t}h(\tau,\xi)\right] = -\frac{\rho}{\theta}\left[1+\frac{\rho}{\theta}\langle K_{d0}\rangle\right]^{-1}\left\{\frac{\partial\langle C_0\rangle(\tau,\xi)}{\partial\tau}E\left[\frac{\partial K_{d1}'(t)}{\partial t}K_{d1}'(\tau)\right] + \langle C_0\rangle(\tau,\xi)E\left[\frac{\partial K_{d1}'(t)}{\partial t}\frac{\partial K_{d1}'(\tau)}{\partial\tau}\right] + \lambda^*\langle C_0\rangle(\tau,\xi)E\left[\frac{\partial K_{d1}'(t)}{\partial t}K_{d1}'(\tau)\right]\right\} \tag{36}$$

It should be noted that the derived solution is quite general, because no assumptions are made about the time-dependent autocovariance function of the distribution coefficient fluctuations about its mean value. In this work, the frequently employed exponential autocovariance function (e.g. Agterberg, 1974; Chrysikopoulos et al., 1990; Bellin et al., 1993) is assumed to characterize the fluctuations of $K_d'(t)$, and is defined as

$$E[K_{d1}'(t)K_{d1}'(\tau)] = \sigma^2_{K_d'}\exp\left[\frac{-(t-\tau)}{a}\right] \quad (t \geq \tau) \tag{37}$$

where $\sigma^2_{K_d'}$ is the variance of $K_d'(t)$, and a is the time correlation scale of $K_d'(t)$. In view of Eq. (37), the remaining three covariance functions present in Eq. (35) and Eq. (36) are evaluated as

$$\begin{aligned} E\left[K_{d1}'(t)\frac{\partial K_{d1}'(\tau)}{\partial\tau}\right] &= \lim_{\Delta\tau\to 0} E\left[K_{d1}'(t)\left(\frac{K_{d1}'(\tau+\Delta\tau)-K_{d1}'(\tau)}{\Delta\tau}\right)\right] \\ &= \lim_{\Delta\tau\to 0}\frac{1}{\Delta\tau}E[K_{d1}'(t)K_{d1}'(\tau+\Delta\tau)-K_{d1}'(t)K_{d1}'(\tau)] \\ &= \frac{\partial}{\partial\tau}E[K_{d1}'(t)K_{d1}'(\tau)] = \sigma^2_{K_{d1}'}\frac{\partial}{\partial\tau}\exp\left[\frac{-(t-\tau)}{a}\right] \\ &= \frac{\sigma^2_{K_{d1}'}}{a}\exp\left[\frac{-(t-\tau)}{a}\right] = \frac{1}{a}E[K_{d1}'(t)K_{d1}'(\tau)] \end{aligned} \tag{38}$$

$$E\left[\frac{\partial K_{d1}'(t)}{\partial t}K_{d1}'(\tau)\right] = \frac{\partial}{\partial t}E[K_{d1}'(t)K_{d1}'(\tau)] = -\frac{1}{a}E[K_{d1}'(t)K_{d1}'(\tau)] \tag{39}$$

$$E\left[\frac{\partial K_{d1}'(t)}{\partial t}\frac{\partial K_{d1}'(\tau)}{\partial \tau}\right] = \frac{\partial}{\partial t}\frac{\partial}{\partial \tau}E[K_{d1}'(t)K_{d1}'(\tau)] = -\frac{1}{a^2}E[K_{d1}'(t)K_{d1}'(\tau)] \quad (40)$$

Now the desired analytical solution Eqs. (13a)–(13c) to the governing stochastic partial differential equation Eq. (9) subject to conditions Eqs. (10a)–(10c) is completed. The ensemble average $\langle C\rangle(t,x)$ is evaluated by substituting the derived expression for the fundamental solution $F(t,x,\xi)$ defined in Eq. (17), and the expression for $G(t,x)$ defined by Eq. (11) in conjunction with Eqs. (32–(40).

3. Model simulations and discussion

The effect of the time-dependent distribution coefficient on virus transport was investigated by presenting temporal and spatial distributions of the liquid-phase virus concentration for a variety of situations. For presentation purposes, calculated ensemble average concentrations were normalized by the source concentration. All integrals in the analytical solution (Eq. (13a), Eqs. (32)–(34)) were evaluated by the extended Simpson's rule (Press et al., 1992). Unless otherwise specified, breakthrough curves were predicted at a distance x = 40 cm downstream from the source. The fixed parameter values used for the model simulations are shown in Table 1.

For the special case where the inactivation of liquid-phase and adsorbed viruses is equal to zero ($\lambda = \lambda^* = 0$) and the distribution coefficient is constant ($K_d(t) = \langle K_d\rangle$), the derived solution Eqs. (13a)–(13c) is equivalent to the analytical solution of the well-known advection–dispersion equation with constant coefficients and flux-type inlet boundary conditions ($\langle C\rangle(t,x) = C(t,x)$) as presented by Lindstrom et al. (1967) and tabulated by Van Genuchten and Alves (1982, p. 10):

$$C(t,x) = \frac{C_0}{2}erfc\left[\frac{x-\hat{U}t}{2(\hat{D}t)^{1/2}}\right] + \left(\frac{\hat{U}^2 t}{\pi\hat{D}}\right)^{1/2}\exp\left[-\frac{(x-\hat{U}t)^2}{4\hat{D}t}\right]$$
$$-\frac{C_0}{2}\left(1+\frac{\hat{U}x}{\hat{D}}+\frac{\hat{U}^2 t}{\hat{D}}\right)\exp\left[\frac{\hat{U}x}{\hat{D}}\right]erfc\left[\frac{x+\hat{U}t}{2(\hat{D}t)^{1/2}}\right] \quad (41)$$

where $\hat{D}$ and $\hat{U}$ are defined in Eq. (16a) and Eq. (16b), respectively. For this particular case the analytical solution to the ensemble mean liquid-phase virus concentration (Eqs. (13a)–(13c)) was validated via comparison with Eq. (41). All concentrations were

Table 1
Model parameters for simulations

Parameter	Value
Dispersion coefficient	D = 1.6 cm^2 h^{-1}
Interstitial velocity	U = 1.0 cm h^{-1}
Bulk density	ρ = 1.5 g cm^{-3}
Porosity	θ = 0.25

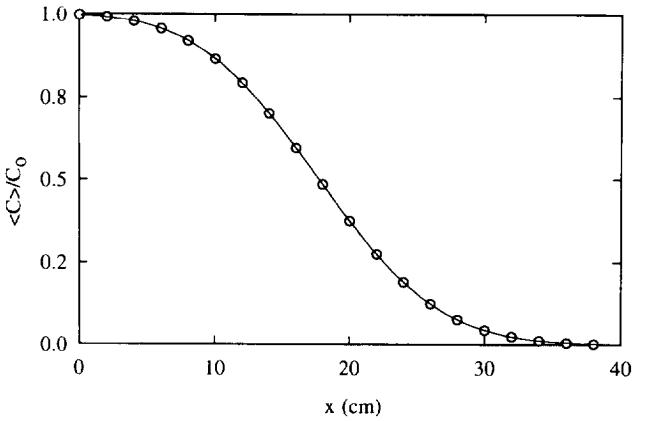

Fig. 3. Comparison between the stochastic model (Eq. (13), continuous line) and the deterministic model (Eq. (41), symbols) for the special case where $\sigma^2_{K_d'} = 0.0$ and $\lambda = \lambda^* = 0.0$ day^{-1}. (Here $t = 5$ days, and $\langle K_d \rangle = 1.0$ g cm^{-3}.)

conveniently expressed as dimensionless quantities ($\langle C \rangle(t,x)/C_0$). Clearly, Fig. 3 illustrates that the two simulations are virtually identical.

Snapshots (Fig. 4(a)) and breakthrough curves (Fig. 4(b)) created by the stochastic virus transport model Eqs. (13a)–(13c) were compared with the case of virus transport with constant distribution coefficient. Two different variances for $K_d'(t)$ were examined, including the zero-variance, which corresponds to the situation where the distribution coefficient is constant. It was shown that the temporally variable distribution coefficient leads to earlier breakthrough and enhanced spreading of the ensemble average liquid-phase virus concentration. The broadening of the predicted ensemble average liquid-phase virus concentration curves can also be interpreted as the result of an effective increase in the dispersive mass flux caused by the time-dependent distribution coefficient. This

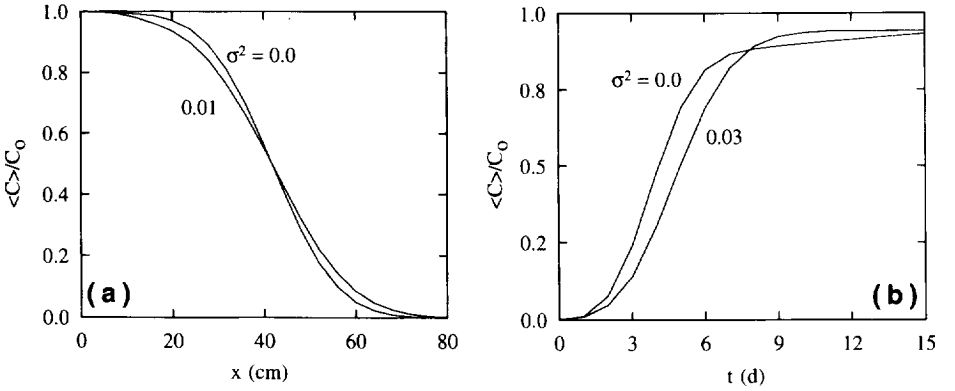

Fig. 4. Effect of the time-dependent distribution coefficient on (a) spatial and (b) temporal ensemble average normalized liquid-phase virus concentration profiles for two different variances of the fluctuating distribution coefficient. (Here $t = 5$ days, $\langle K_d \rangle = 0.33$ g cm^{-3}, $a = 0.5$ days; (a) $\sigma^2_{K_d'} = 0.01$, $\lambda = 0.004$ day^{-1}, and $\lambda^* = 0.002$ day^{-1}; (b) $\sigma^2_{K_d'} = 0.03$, $\lambda = 0.03$ days^{-1}, and $\lambda^* = 0.003$ days^{-1}.)

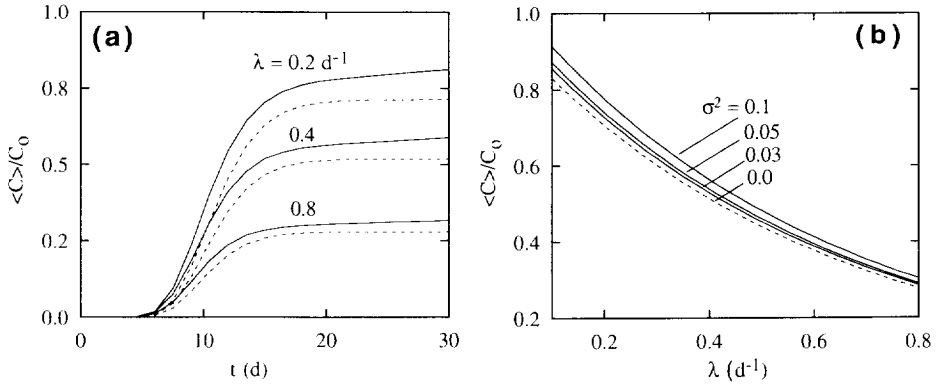

Fig. 5. Ensemble average normalized liquid-phase virus concentration profiles as a function of (a) time for several λ values with $\sigma^2_{K_d'} = 0.0$ (dashed lines) and $\sigma^2_{K_d'} = 0.01$ (continuous lines), and (b) inactivation rate constant of liquid-phase viruses for several values of $\sigma^2_{K_d'}$. (Here t = 20 days, $\langle K_d \rangle$ = 1.0 g cm^{-3}, a = 0.5 days, and λ^* = 0.001 days^{-1}.)

observation is analogous to the enhancement of solute spreading caused by a spatially variable retardation factor (Chrysikopoulos et al., 1990).

The effect of inactivation rate constant of liquid-phase viruses on the temporally distributed ensemble average normalized virus concentration is shown in Fig. 5(a). Three breakthrough curves, indicated by the continuous lines, were constructed for λ = 0.2, 0.4 and 0.8 day^{-1}, and they were compared with the case of invariable distribution coefficient, the dashed lines in Fig. 5(a). The effect of $\sigma^2_{K_d'}$ on liquid-phase virus concentration as a function of λ is illustrated in Fig. 5(b). The values of $\sigma^2_{K_d'}$ considered are: 0.0, 0.03, 0.05 and 0.1, where $\sigma^2_{K_d'} = 0.0$ corresponds to the case of constant $\langle K_d \rangle$. The concentration profiles suggest the intuitive result that the liquid-phase virus concentration decreases with increasing λ. Furthermore, it is illustrated that $K_d(t)$ leads to earlier breakthrough of the ensemble average liquid-phase virus concentration.

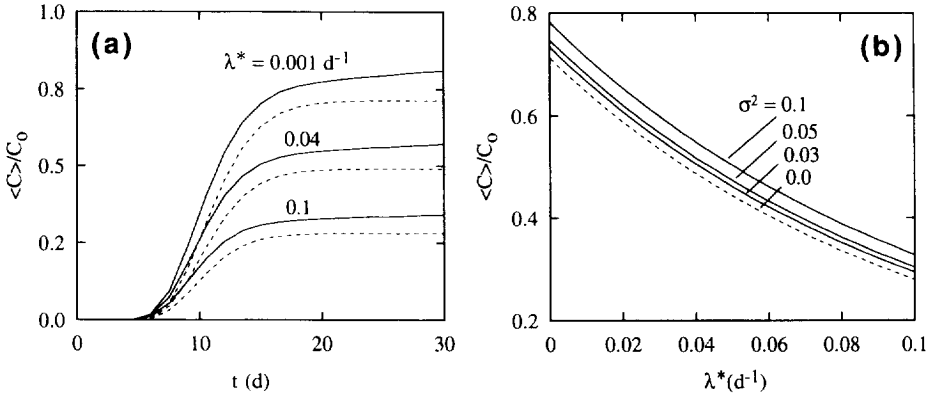

Fig. 6. Ensemble average normalized liquid-phase virus concentration profiles as a function of (a) time for several λ^* values with $\sigma^2_{K_d'} = 0.0$ (dashed lines) and $\sigma^2_{K_d'} = 0.01$ (continuous lines), and (b) inactivation rate constant of adsorbed viruses for several values of $\sigma^2_{K_d'}$. (Here t = 20 days, $\langle K_d \rangle$ = 1.0 g cm^{-3}, a = 0.5 days, and λ = 0.2 days^{-1}.)

The effect of inactivation rate constant of adsorbed viruses on the temporally distributed ensemble average normalized virus concentration is presented in Fig. 6(a), and the effect of $\sigma^2_{K_d'}$ on liquid-phase virus concentration as a function of λ^* is shown in Fig. 6(b). It is evident that the normalized ensemble average liquid-phase virus concentration decreases with increasing λ^*, and that $K_d(t)$ appears to cause a faster virus breakthrough.

The effect of the time correlation scale of $K_d'(t)$ on spatial and temporal normalized liquid-phase ensemble average virus concentration distributions is illustrated in Fig. 7(a) and Fig. 7(b), respectively. Two correlation scales, 0.5 and 1.0 days, were considered. The concentration profiles were conveniently compared with the case of constant distribution coefficient ($\sigma^2_{K_d'} = 0$), which is represented by the dashed lines. It is shown that the smaller the time correlation scale the earlier the breakthrough of the liquid-phase virus concentration. The correlation scale represents the influence period of K_d variation (Journel and Huijbregts, 1978). Therefore, the smaller the time correlation scale, the higher the variability in the K_d fluctuations and consequently the greater the spreading of the liquid-phase virus concentration.

The validity of the local equilibrium assumption, neglecting rate limitations, may be questioned. It is, however, employed in this study in the interest of mathematical simplicity. Furthermore, the physical significance of the simulations presented may be criticized because, at present, there are no experimental data available in the literature to support the validity of the exponential autocovariance function used. However, it should be noted that the fundamental results of this work will not be affected qualitatively if another autocovariance function had been employed.

4. Summary

A virus transport model was developed for one-dimensional, homogeneous, saturated, but geochemically heterogeneous porous media. The model accounts for first-order inactivation of liquid-phase and adsorbed viruses with different inactivation rate constants,

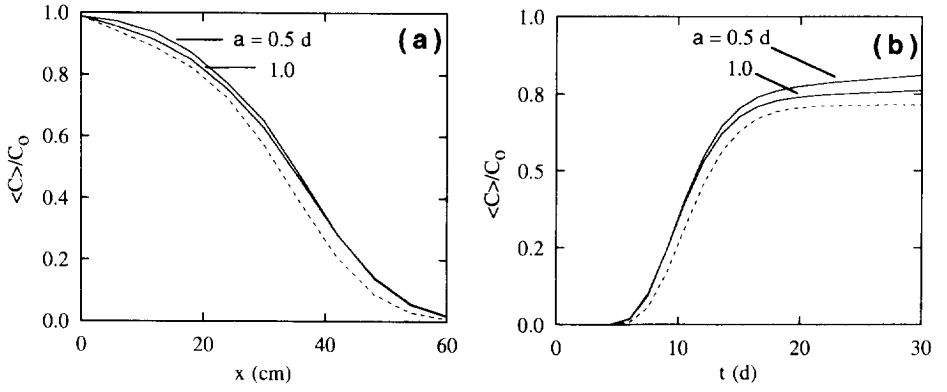

Fig. 7. Effect of the time correlation scale of $K_d'(t)$ on (a) spatial and (b) temporal ensemble average normalized liquid-phase virus concentration profiles. The dashed lines represent the situation where the distribution coefficient is constant. (Here $\langle K_d \rangle = 1.0$ g cm^{-3}, $\sigma^2_{K_d'} = 0.1$, $\lambda = 0.2$ day^{-1}, and $\lambda^* = 0.001$ day^{-1}.)

and local equilibrium adsorption described by a stochastic time-dependent distribution coefficient. The governing transport equation was solved analytically, for a semi-infinite porous medium with a flux-type inlet boundary condition. The analytical solution to the governing stochastic partial differential Eq. (9) subject to conditions Eqs. (10a)–(10c) is given by Eqs. (13a)–(13c), where the expressions for $G(t,x)$ and $F(t,x,\xi)$ are defined in Eq. (11) and Eq. (17), respectively. The analytical small-perturbation solution is based on the average of individual realizations of the temporal fluctuations of $K_d'(t)$, and it is general enough so that any autocovariance function for the time-dependent fluctuations of the distribution coefficient can be employed.

The effect of temporally variable distribution coefficient on spatial and temporal virus concentration profiles was investigated. It was shown that the time-dependent distribution coefficient results in an enhanced spreading of the liquid-phase virus concentration.

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