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One-dimensional virus transport in porous media with time-dependent inactivation rate coefficients

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Abstract. A model for virus transport in one-dimensional, homogeneous, saturated porous media is developed, accounting for virus sorption and inactivation of liquid phase and adsorbed viruses with different time dependent rate coefficients. The virus inactivation process is represented by a pseudo first-order rate expression. The pseudo first-order approximation is shown to simulate available experimental data from three virus inactivation batch studies better than the frequently employed constant rate inactivation model. Model simulations indicated that the pseudo first-order approximation, compared to the constant inactivation, leads to extended survival of viruses and, consequently, more distant migration. Results from a parameter sensitivity analysis demonstrated that estimation of pseudo first-order inactivation rate coefficients from field observations requires data collection near the source of virus contamination during initial stages of virus transport.

Introduction

Viruses are submicron particles containing nucleic acids, either RNA or DNA, in a protein coat called a "capsid." Viruses may lose their infectivity owing to disruption of coat proteins and degradation of nucleic acid [*Gerba*, 1984]. This process is known as inactivation. In subsurface formations, inactivation is controlled mainly by the physicochemical characteristics of viruses (e.g., size, chemical composition, protein coat packaging) [*Yamagishi and Ozeki*, 1972] and external factors associated with geochemical heterogeneities of the porous medium (e.g., formation properties, temperature variations) [*Yates and Yates*, 1988]. Therefore the relatively complex nature of subsurface formations may lead to spatially as well as temporally variable virus inactivation.

Temporal variation of inactivation rate coefficients due to variabilities of virus characteristics has been observed in several experimental studies. *Parkinson and Huskey* [1971] and *Pollard and Solosko* [1971] noticed that bacteriophage λ and T_4 populations consist of two subpopulations with different resistances to heat (biphasic inactivation). They observed that the most sensitive viruses inactivate rapidly, while the remaining more resistant viruses undergo slower inactivation. Similarly, *Yamagishi and Ozeki* [1972] reported that the inactivation of bacteriophage λ exhibits two or more distinct phases (multiphasic inactivation), corresponding to subpopulations undergoing sequential inactivation with different inactivation rate coefficients. *Grant et al*. [1993] also observed multiphasic sequential inactivation of bacteriophage λ during batch experiments with and without the presence of sand.

Traditionally, models for virus transport through porous formations assume that the inactivation rate coefficients are constant [*Yates and Ouyang*, 1992; *Chrysikopoulos and Sim*, 1996]. It should be noted, however, that sequential inactivation of a virus population requires two or more discrete first-order rate coefficients, each governing a different inactivation phase [*Crane and Moore*, 1986]. For mathematical simplicity the multiphasic sequential inactivation can be approximated by a pseudo first-order expression with a time-dependent inactivation rate coefficient.

The present work introduces a model for one-dimensional virus transport in homogeneous, saturated porous media accounting for virus sorption and inactivation with time-dependent rate coefficients. The inactivation process is represented by a pseudo first-order expression with time-dependent rate coefficients determined from available experimental data. Model simulations are compared to the frequently considered case of constant inactivation rate coefficients. Furthermore, sensitivity analysis is conducted to evaluate the response of the virus transport model to inactivation rate fluctuations.

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Model Development

Governing Equations

The transient virus transport through one-dimensional, homogeneous, saturated porous media, accounting for virus adsorption and inactivation, is governed by the following partial differential equation [*Sim and Chrysikopoulos*, 1995]:

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

where C is the liquid phase virus concentration [M/L^3]; C^* is the adsorbed phase virus concentration (virus mass/solids mass) [M/M]; D is the hydrodynamic dispersion coefficient [L^2/t]; U is the average interstitial velocity [L/T]; λ and λ* are the time dependent inactivation rate coefficients of liquid phase and adsorbed phase viruses, respectively, [1/T]; ρ is the bulk density of the solid matrix [M/L^3]; θ is the porosity of the porous medium (liquid volume/aquifer volume) [L^3/L^3]; t is time [T]; and x is the spatial coordinate in the direction of flow [L]. The left-hand side of (1) consists of the accumulation

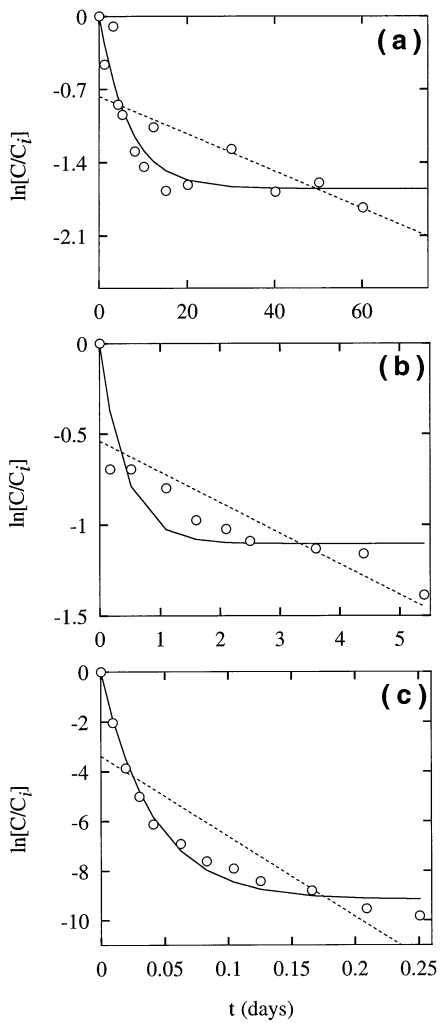

Figure 1. Batch inactivation experimental data (circles). (a) Poliovirus at 1°C with distilled water under anaerobic conditions adopted from *Hurst et al*. [1980]. (b) Bacteriophage λ at ≃15°C with 10 mM NaCl electrolyte and *p*H 7 under aerobic conditions adopted from *Grant et al*. [1993]. (c) Bacteriophage λ^{++} at 60°C with 10 mM $MgSO_4$ under aerobic conditions adopted from *Parkinson and Huskey* [1971]. Simulated concentration history is based on the pseudo first-order inactivation model (solid curves) and constant rate inactivation model (dashed lines).

terms, whereas the last two terms represent the inactivation of suspended and adsorbed viruses, respectively.

Assuming that the adsorption process consists of virus diffusion to the outer layer of a solid particle by nonequilibrium mass transfer and virus immobilization onto the solid particle while in equilibrium with the liquid phase virus concentration in the outer layer, also accounting for inactivation of adsorbed viruses, the accumulation of adsorbed viruses can be represented by the following mass balance equation:

$$\frac{\rho}{\theta}\frac{\partial C^*(t,x)}{\partial t} = k[C(t,x) - C_g(t,x)] - \lambda^*(t)\frac{\rho}{\theta}C^*(t,x), \tag{2}$$

where k is the mass transfer rate constant [1/T], and C_g is the liquid phase virus concentration in direct contact with solids [M/L^3]. Furthermore, it is assumed that the following linear equilibrium relationship is valid:

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

where K_d is the partition or distribution coefficient (liquid volume/solids mass) [L^3/M].

Time-Dependent Inactivation

In the present work, inactivation rate coefficients are considered to be time dependent, and, consequently, the inactivation of viruses in the liquid phase and solid phase are described by the modified first-order rate expressions

$$\frac{dC(t)}{dt} = -\lambda(t)C(t), \tag{4}$$

$$\frac{dC^*(t)}{dt} = -\lambda^*(t)C^*(t), \tag{5}$$

respectively, where the time-dependent inactivation rate coefficients of viruses in the respective phases are described by

$$\lambda(t) = \lambda_0 \exp(-\alpha t), \tag{6}$$

$$\lambda^*(t) = \lambda_0^* \exp(-\alpha^* t), \tag{7}$$

where λ_0 and λ_0^* are the initial inactivation rate coefficients of viruses in the respective phases [1/T], and α and α^* are the resistivity coefficients of viruses in the respective phases [1/T]. The magnitude of α is proportional to the resistivity of the dominant subpopulation, because the overall inactivation is controlled by the dominant subpopulation. The inactivation rate coefficients of viruses in the liquid phase are assumed to be twice as large as the coefficients of adsorbed viruses ($\lambda_0^* = \lambda_0/2$) [*Reddy et al*., 1981; *Yates and Ouyang*, 1992]. Furthermore, the resistivity coefficient of adsorbed viruses is considered to be equal to the resistivity coefficient of viruses in the liquid phase ($\alpha^* = \alpha$). Substituting (6) into (4) and solving the resulting expression subject to the initial condition $C(0) = C_i$, where C_i is the initial liquid phase virus concentration, yields

$$\ln\left[\frac{C(t)}{C_i}\right] = \frac{\lambda_0}{\alpha}[\exp(-\alpha t) - 1]. \tag{8}$$

The parameters λ_0 and α can be obtained by fitting (8) to existing experimental data.

Figure 1 presents virus inactivation experimental data fitted by both (8) and the constant inactivation rate model ($\lambda(t) = \lambda$). The estimated parameters for both inactivation rate models considered together with the corresponding residual sums of squared error (sse) are listed in Table 1. Clearly, the pseudo first-order inactivation rate model simulates the experimental data much better than the constant inactivation rate model, which fails to match the data at early and late times (see Figure 1). The slope of a tangent to a solid curve represents the inactivation rate coefficient at the particular time.

Initial and Boundary Conditions

The appropriate initial and boundary conditions for a semi-infinite, one-dimensional porous formation in the presence of a continuous source of viruses are [*Sim and Chrysikopoulos*, 1995]

$$C(0,x) = C^*(0,x) = 0, \tag{9}$$

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

$$\frac{\partial C(t,\infty)}{\partial x} = 0, \tag{11}$$

where C_0 is the source concentration. The condition (9) establishes that there are no initial liquid phase and adsorbed virus concentrations within the porous medium. The constant flux boundary condition (10) implies virus concentration discontinuity at the inlet. The downstream boundary condition (11) preserves concentration continuity for a semi-infinite system. The governing virus transport equation (1) in conjunction with the relationships (2), (3), (6), and (7) is solved numerically subject to initial/boundary conditions (9)–(11). The numerical solution is obtained by using the International Mathematics and Statistics, Inc., (IMSL) one-dimensional partial differential equation (PDE) solver MOLCH [*IMSL*, 1991].

Model Simulations

The effect of temporally variable inactivation on liquid phase virus concentration in saturated porous media is investigated by conducting model simulations. The fixed parameter values used for virus transport simulations are $D = 32.04$ cm^2/hr and $U = 5.04$ cm/hr [*Bales et al*., 1991]; $K_d = 2.08 \times 10^{-2}$ ml/mg [*Vilker*, 1981]; $k = 1.2$ hr^{-1} [*Vilker and Burge*, 1980]; $\rho = 1.5$ g/cm^3 [*Yates and Ouyang*, 1992]; and $\theta = 0.25$ [*Park et al*., 1992]. The pseudo first-order inactivation rate parameters are estimated from the experimental data collected by *Grant et al*. [1993] (see Figure 1b).

At early time the simulated concentration profile for the case of pseudo first-order inactivation is lower than the one for the case of constant inactivation rate (see Figure 2a), whereas at late time the concentration levels are reversed (see Figure 2b). The temporally variable inactivation allows rapid inactivation of the most sensitive subpopulations at early time and extended survival of the most resistive subpopulations at late time. Therefore viruses may remain infective in porous media for an extended period of time and thus travel farther downstream from the source.

Parameter Sensitivity Analysis

In order to investigate the response of the virus transport model to perturbations of the pseudo first-order inactivation parameters α and λ_0, a formal parameter sensitivity analysis is conducted. A parameter sensitivity coefficient that represents the degree of spatial and temporal change in the dependent variable (i.e., concentration) due to the fluctuation of a particular model parameter was obtained by differentiating the dependent variable with respect to the parameter of interest [*Koda et al*., 1979; *Knopman and Voss*, 1987].

Table 1. Estimated Virus Inactivation Parameters for Three Data Sets

Parameter	Poliovirus (1°C)*	Bacteriophage λ (≃15°C)†	Bacteriophage λ^{++} (60°C)‡
	Time Dependent Inactivation		
λ_0, d^{-1}	0.25	2.66	226.02
α, d^{-1}	0.42	2.41	24.65
sse	0.68	0.27	1.67
	Constant Inactivation		
λ, d^{-1}	0.018	0.17	32.28
sse	2.45	0.40	26.71

**Hurst et al*. [1980].
†*Grant et al*. [1993].
‡*Parkinson and Huskey* 1971].

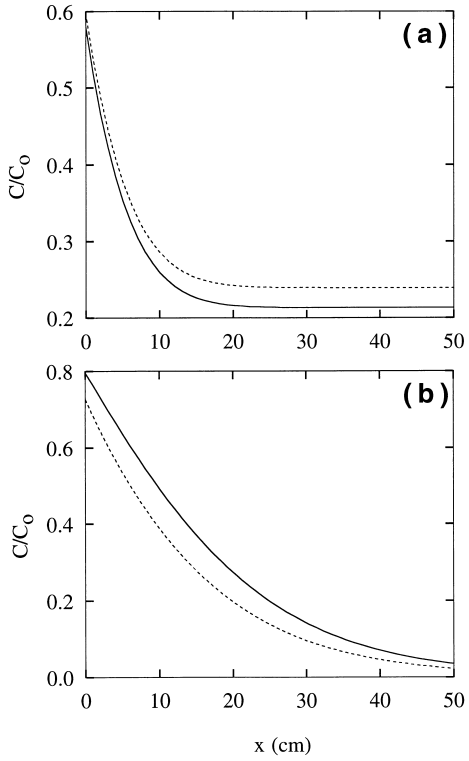

Figure 2. Liquid phase virus concentration snapshots for pseudo first-order (solid curves) or constant rate (dashed curves) inactivation at (a) $t = 0.05$ days and (b) $t = 10$ days.

The sensitivity coefficients with respect to the resistivity coefficient for liquid phase and adsorbed virus concentrations are given by

$$Z_\alpha(t,x) = \frac{\partial C(t,x)}{\partial \alpha}, \tag{12a}$$

$$Z^*_\alpha(t,x) = \frac{\partial C^*(t,x)}{\partial \alpha}, \tag{12b}$$

respectively. Differentiating the governing equations (1)–(3) with respect to α yields

$$\frac{\partial Z\alpha(t,x)}{\partial t} + \frac{\rho}{\theta}\frac{\partial Z^*_\alpha(t,x)}{\partial t} = D\frac{\partial^2 Z_\alpha}{\partial x^2} - U\frac{\partial Z_\alpha}{\partial x} + \lambda(t)[tC(t,x) - Z_\alpha(t,x)] - \lambda^*(t)\frac{\rho}{\theta}Z^*_\alpha(t,x), \tag{13}$$

$$\frac{\rho}{\theta}\frac{\partial Z^*_\alpha(t,x)}{\partial t} = k\left[Z_\alpha(t,x) - \frac{Z^*_\alpha(t,x)}{K_d}\right] - \lambda^*(t)\frac{\rho}{\theta}Z^*_\alpha(t,x). \tag{14}$$

Similarly, the corresponding initial and boundary conditions are obtained from (9)–(11) as follows:

$$Z_\alpha(0,x) = Z^*_\alpha(0,x) = 0, \tag{15}$$

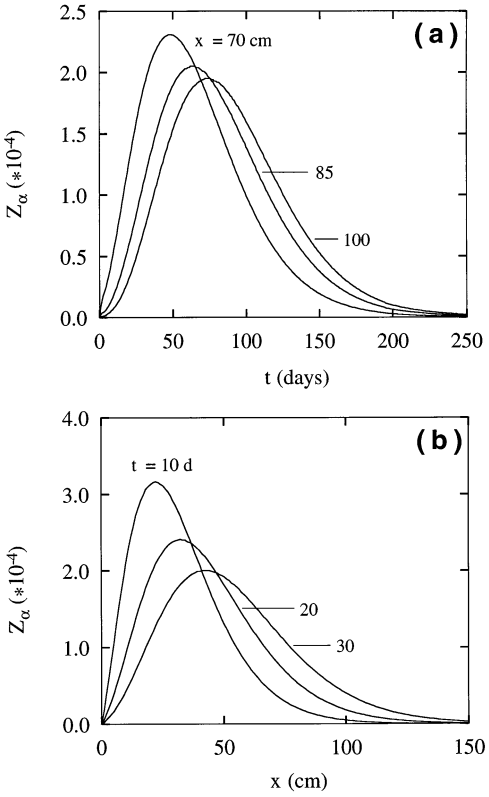

Figure 3. Variation of Z_α as a function of (a) time and (b) space.

$$- D\frac{\partial Z_\alpha(t, 0)}{\partial x} + UZ_\alpha(t, 0) = 0, \tag{16}$$

$$\frac{\partial Z_\alpha(t, \infty)}{\partial x} = 0. \tag{17}$$

The sensitivity coefficients Z_α and Z^*_α are evaluated by solving (13) and (14) subject to (15)–(17) together with the governing equations (1) and (2) subject to conditions (9)–(11). A numerical solution is obtained by MOLCH [*IMSL*, 1991].

The sensitivity coefficients with respect to the initial inactivation rate coefficient for liquid phase and adsorbed virus concentrations are given by

$$Z_{\lambda_0}(t, x) = \frac{\partial C(t, x)}{\partial \lambda_0}, \tag{18a}$$

$$Z^*_{\lambda_0}(t, x) = \frac{\partial C^*(t, x)}{\partial \lambda_0}, \tag{18b}$$

respectively. The desired partial differential equations are obtained in a fashion similar to the case of resistivity coefficient

$$\frac{\partial Z_{\lambda_0}(t, x)}{\partial t} + \frac{\rho}{\theta}\frac{\partial Z^*_{\lambda_0}(t, x)}{\partial t} = D\frac{\partial^2 Z_{\lambda_0}}{\partial x^2} - U\frac{\partial Z_{\lambda_0}}{\partial x}$$

$$-[\exp(-\alpha t)]C(t, x) - \lambda(t)Z_{\lambda_0}(t, x) - \lambda^*(t)\frac{\rho}{\theta}Z^*_{\lambda_0}(t, x), \tag{19}$$

$$\frac{\rho}{\theta}\frac{\partial Z^*_{\lambda_0}(t, x)}{\partial t} = k\left[Z_{\lambda_0}(t, x) - \frac{Z^*_{\lambda_0}(t, x)}{K_d}\right]$$

$$- \lambda^*(t)\frac{\rho}{\theta}Z^*_{\lambda_0}(t, x). \tag{20}$$

The sensitivity coefficients Z_{λ_0} and $Z^*_{\lambda_0}$ are evaluated by solving numerically (19) and (20) subject to conditions (15)–(17) with substitution of Z_α and Z^*_α by Z_{λ_0} and $Z^*_{\lambda_0}$, in conjunction with the governing equations (1) and (2) subject to conditions (9)–(11).

Figure 3a shows that at different locations within the one-dimensional porous medium, Z_α approaches a zero value with increasing time because the system reaches a steady state, where $C(t, x)$ is insensitive to variations in α. The impact of α on liquid phase virus concentration is most significant near the source. However, it should be noted that although the peak value of Z_α decreases with increasing distance from the source, the time interval over which α influences the liquid phase concentration increases with increasing distance from the source. The spatial variation of Z_α at different simulation times is illustrated in Figure 3b. It is observed that the peak value of Z_α progressively decreases with increasing time because the system approaches a steady state. Therefore liquid phase virus concentrations are highly sensitive to α near the source and during early stages of virus transport. The temporal and spatial variations of the sensitivity coefficient Z_{λ_0} exhibit similar trends to the ones observed for Z_α in Figure 3.

Summary

This study introduces a model for one-dimensional virus transport in saturated, homogeneous porous media. Unlike other virus transport models reported in the literature, this model accounts for inactivation of liquid phase and adsorbed viruses with different time-dependent inactivation rate coefficients which are represented by functional relationships determined from experimental data. The significant impact of temporally variable inactivation rate coefficients on virus transport was demonstrated by model simulations. It was concluded from a formal parameter sensitivity analysis that virus transport data collected in the vicinity of the source of contamination at early time are most reliable for estimation of inactivation rate coefficients.

The model is particularly useful for improving our understanding of virus inactivation processes in conjunction with studying virus transport through packed columns under controlled laboratory conditions. The applicability of this model to field investigations is limited to relatively homogeneous subsurface formations. The methodology of this work can provide a starting point for generalization to multidimensional virus transport in heterogeneous porous or fractured media.

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