Journal of Contaminant Hydrology 35 (1999) 429–440

Analytical solutions for multiple species reactive transport in multiple dimensions

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Received 1 October 1997; accepted 15 July 1998

Abstract

Many numerical computer codes used to simulate multi-species reactive transport and biodegradation have been developed in recent years. Such numerical codes must be validated by comparison of the numerical solutions with an analytical solution. In this paper, a method for deriving analytical solutions of the partial differential equations describing multiple species multi-dimensional transport with first-order sequential reactions is presented. Although others have developed specific solutions of multi-species transport equations, here a more general analytical approach, capable of describing any number of reactive species in multiple dimensions is derived. A substitution method is used to transform the multi-species reactive transport problem to one that can be solved using previously published single-species solutions for various initial and boundary conditions. One- and three-dimensional examples are presented to illustrate the steps involved in extending single-species solutions to a four-species system with sequential first-order reactions. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Analytical solution; Biodegradation; Transport; Multi-species; First-order reaction

1. Introduction

Many numerical computer codes used to simulate multi-species reactive transport and biodegradation have been developed in recent years (Molz et al., 1986; Rifai and Bedient, 1990; Clement et al., 1998). Such numerical codes must be validated by

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PII: S0169-7722(98)00105-3

comparing the numerical solutions against an analytical solution. In most instances, simple first-order sequential degradation models can be used to accomplish this validation process (Clement et al., 1998). Sequential first-order reaction models have also been used to describe reactive transport in many groundwater contamination problems. For example, during denitrification, nitrate reacts to nitrite, and subsequently to ammonia or nitrogen gas. The specific rates of these reactions have been determined previously (Germon, 1983; Hooker et al., 1994). Similarly, reductive anaerobic degradation of PCE (tetrachloroethylene) to TCE (trichloroethylene), to DCE (dichloroethylene), and to VC (vinyl chloride) may be modeled using a sequential first-order degradation kinetic model (Wiedemeier et al., 1997).

Determination of analytical solutions of solute transport problems usually involve complex mathematical manipulation in order to derive inverse Laplace transforms (Cho, 1970; Bear, 1979; van Genuchten, 1985; Lunn et al., 1996). Cho (1970) derived an analytical solution to a three-species chain with a simple boundary condition. van Genuchten (1985) derived analytical solutions for a four species chain. Lunn et al. (1996) used Fourier sine transforms instead of Laplace transforms to derive an analytical solution of three-species transport in an one-dimensional soil column, considering adsorption of the first species. However, these analytical solutions are limited to one dimension with specific initial and boundary conditions. The 1-D assumption limits the application of the solution. Moreover, even though the solution of Lunn et al. (1996) is relatively simpler than the solution of Cho (1970), it is difficult to implement it as a general computer code.

Progress has been made in single species transport with first-order reaction in multiple dimensions. After Bear (1972) developed the solution of single species transport with first-order decay in one dimension, Wilson and Miller (1978) and Domenico (1987), respectively, developed its two- and three-dimensional analytical solutions. Recently, the solution of Domenico (1987) was used to evaluate natural attenuation in field scale systems (Newell et al., 1996).

In this paper, a general method is developed to derive analytical solutions of any number of species with first-order sequential degradation in multiple dimensions. The solution is based on existing analytical solutions of single-species transport with first-order reaction which are available for various initial and boundary conditions (Wilson and Miller, 1978; Bear, 1979; Domenico, 1987). Using the proposed method, any single species solution can be directly extended to describe the reactive transport of any number of species that are degrading sequentially with a series of first-order reactions.

2. The multiple species transport equations

The basic equation used to describe single species transport in porous media is the mass balance equation (Bear, 1979):

$$\frac{\partial c}{\partial t} - D_x \frac{\partial^2 c}{\partial x^2} - D_y \frac{\partial^2 c}{\partial y^2} - D_z \frac{\partial^2 c}{\partial z^2} + v \frac{\partial c}{\partial x} = f, \tag{1}$$

where c is the species concentration $[ML^{-3}]$; x, y, z are distance $[L]$; t is time $[T]$; D is constant hydrodynamic dispersion coefficient $[L^2T^{-1}]$; v is constant flow velocity $[LT^{-1}]$; and f is the reaction rate $[ML^{-3}T^{-1}]$.

When the reaction is assumed to be first-order, Eq. (1) can be written as:

$$\frac{\partial c}{\partial t} - D_x\frac{\partial^2 c}{\partial x^2} - D_y\frac{\partial^2 c}{\partial y^2} - D_z\frac{\partial^2 c}{\partial z^2} + v\frac{\partial c}{\partial x} = -kc, \tag{2}$$

where k is the first-order reaction rate $[T^{-1}]$. Several analytical solutions are available for Eq. (2) with various initial and boundary conditions (Bear, 1979).

The above equation can be extended to describe reactive transport of multiple species. For example, anaerobic degradation of PCE may be modeled as a sequential reaction chain with each species degrading in a first-order fashion (Wiedemeier et al., 1997; Clement et al., 1998):

$$c_1 \to c_2 \to \cdots c_i \to \cdots \to c_n,$$

where c_i is the species concentration in the ith generation. Species i, which is produced by reaction of Species $i-1$, also reacts to produce Species $i+1$ which in turn reacts to produce Species $i+2$. Such a sequential reactive transport system can be described using a general multi-species transport equation of the form:

$$\frac{\partial c_i}{\partial t} - D_x\frac{\partial^2 c_i}{\partial x^2} - D_y\frac{\partial^2 c_i}{\partial y^2} - D_z\frac{\partial^2 c_i}{\partial z^2} + v\frac{\partial c_i}{\partial x} = k_{i-1}c_{i-1} - k_i c_i, \tag{3}$$

$$\forall i = 1,2,\dots,n,$$

where i is the index of species, k_i, $i = 1,2,\dots,n$, are constant first-order reaction rates $[T^{-1}]$, $k_o = 0.0$, and n is the number of species. Assume stoichiometry of the above reaction is such that one mole of product is produced from one mole of reactant.

3. Solution method

Various solutions for any single-species reactive transport problem, of the form of Eq. (2), are readily available in the literature. Here we present a new substitution method to transform Eq. (3) to the same form as Eq. (2), so that the single-species solutions can be used to derive solutions of any number of coupled multiple species transport equations.

To accomplish this, we first define a set of auxiliary variables as:

$$a_i = c_i + \sum_{j=1}^{i-1}\prod_{l=j}^{i-1}\frac{k_l}{k_l - k_i}c_j, \tag{4}$$

$$\forall i = 2,3,\dots,n.$$

Differentiating Eq. (4) with respect to time, the reactive transport equations for auxiliary variables from Species 2 to Species n can be simplified as:

$$\frac{\partial a_i}{\partial t} - D_x \frac{\partial^2 a_i}{\partial x^2} - D_y \frac{\partial^2 a_i}{\partial y^2} - D_z \frac{\partial^2 a_i}{\partial z^2} + v \frac{\partial a_i}{\partial x} = -k_i a_i, \tag{5}$$
$$\forall i = 2,3,\dots,n,$$

After reducing to this form, any previously derived analytical solutions for single-species transport with first-order reaction can be directly applied for predicting multiple species transport. Care must be taken to also appropriately transform all the initial and boundary conditions. The mathematical steps involved in reducing Eqs. (3)–(5), using the auxiliary variable transformation in Eq. (4), are included in Appendix A. Numerical implementation, showing all the mathematical steps involved in reducing a multi-species equation to a single-species form, is illustrated in Section 4.

4. Numerical implementation

In order to illustrate the numerical implementation of the substitution procedure for solving the multiple species transport problem with successive first-order degradation terms, we start from an example of a single-species transport in a semi-infinite column. The transport system is defined as (Bear, 1979):

$$\begin{aligned} &\frac{\partial c}{\partial t} - D\frac{\partial^2 c}{\partial x^2} + v\frac{\partial c}{\partial x} = -kc \\ &c(x,0) = 0 \qquad x \geq 0 \\ &c(0,t) = c_o \qquad t > 0 \\ &c(\infty,t) = 0 \qquad t > 0, \end{aligned} \tag{6}$$

where the column $(x > 0)$, initially at the species concentration $c = 0$, is connected to a reservoir at $(x = 0)$ containing the species solution at a constant concentration c_o. The flow in the column is maintained at a constant specific discharge $q = v\phi$ in the $+x$ direction, and ϕ is a constant porosity.

By applying the Laplace transform, the solution of Eq. (6) is obtained as (Bear, 1972, 1979):

$$\begin{aligned} c(x,t) = \frac{c_o}{2}\exp\left(\frac{vx}{2D}\right)\Bigg[&\exp(-\beta x)\,\mathrm{erfc}\,\frac{x - (v^2 + 4kD)^{1/2}t}{2(Dt)^{1/2}} \\ &+ \exp(\beta x)\,\mathrm{erfc}\,\frac{x + (v^2 + 4kD)^{1/2}t}{2(Dt)^{1/2}}\Bigg], \end{aligned} \tag{7}$$

where

$$\beta = \left(v^2/4D^2 + k/D\right)^{1/2},$$

and

$$\mathrm{erfc}(x) = 1 - \mathrm{erf}(x) = \frac{2}{\sqrt{\pi}}\int_x^\infty \exp(-\tau^2)\,d\tau.$$

Since hydraulic parameters are all assumed to be constant, the solution of Eq. (7) is only a function of the boundary concentration, c_o, and the reaction rate constant, k.

Since Eq. (6) is exactly the same as Eq. (5), in a one-dimensional form, solution Eq. (7) can be applied to Eq. (5) with an appropriate definition of the auxiliary variable. The general form of the solution, in terms of the auxiliary variables, can written as:

$$a_i(x,t) = F(a_{io},k_i), \qquad \forall i = 1,2,\ldots,n \tag{8}$$

where F represents the functional form of Eq. (7), a_{io} is used instead of c_o, and k_i is used instead of k. The value of a_{io} can be computed from c_o using the expression:

$$a_{io} = c_{io} + \sum_{j=1}^{i-1}\prod_{l=j}^{i-1}\frac{k_l}{k_l - k_i}c_{jo}, \qquad \forall i = 1,2,\ldots,n. \tag{9}$$

After solving the transport Eq. (5), in terms of the auxiliary variable, a_i, the concentration of desired species, c_i, can be determined from the following expression:

$$c_i = a_i - \sum_{j=1}^{i-1}\prod_{l=j}^{i-1}\frac{k_l}{k_l - k_i}c_j, \qquad \forall i = 1,2,\ldots,n. \tag{10}$$

To implement this solution procedure, a set of Fortran subroutines, incorporating different published analytical solutions, for simulating single-species transport with first-order reaction was developed. Based on the problem specific initial and boundary conditions, the required subroutine is first called and the solution is computed in terms of auxiliary variables. Later, real concentration values are calculated by a recursive substitution process using Eq. (10).

5. Adsorption consideration

The analytical solutions discussed so far ignore adsorption reactions. When linear adsorption is assumed and if reactions occur only in the aqueous phase, then the reactive transport equations can be written as:

$$R\frac{\partial c_i}{\partial t} - D_x\frac{\partial^2 c_i}{\partial x^2} - D_y\frac{\partial^2 c_i}{\partial y^2} - D_z\frac{\partial^2 c_i}{\partial z^2} + v\frac{\partial c_i}{\partial x} = k_{i-1}c_{i-1} - k_i c_i, \tag{11}$$
$$\forall i = 1,2,\ldots,n,$$

where R is the retardation factor for all species and $k_o = 0$.

If Eq. (11) is divided by retardation factor, R, it becomes identical to the basic continuity equation without considering adsorption,

$$\frac{\partial c_i}{\partial t} - D'_x\frac{\partial^2 c_i}{\partial x^2} - D'_y\frac{\partial^2 c_i}{\partial y^2} - D'_z\frac{\partial^2 c_i}{\partial z^2} + v'\frac{\partial c_i}{\partial x} = k'_{i-1}c_{i-1} - k'_i c_i, \tag{12}$$

where $D'_x = D_x/R$, $D'_y = D_y/R$, $D'_z = D_z/R$, $v' = v/R$, and $k' = k/R$.

Therefore, the solution method derived above is applicable also when sorption is considered. Note that the flow velocity, the dispersion coefficients, and reaction rates should be transformed accordingly. Instead of Eq. (8), the solution of the auxiliary variables, a_i, $\forall i = 1,2,\ldots,n$, with linear adsorption, is

$$a_i(x,t) = F(a_{io},k'_i,D',v'). \tag{13}$$

The presented transformation procedure is valid for deriving multi-species analytical solutions only when the retardation factor is identical for all the transported species.

6. Analysis and application

6.1. Three-species reactive transport in a one-dimensional soil column-comparison with Lunn et al. (1996)

Cho (1970) and Lunn et al. (1996) simulated three nitrogen species (ammonium, nitrite, and nitrate) transport in a one-dimensional soil column. Following the example of Lunn et al. (1996), we used the same column geometry and defined system parameters as shown in Table 1. The retardation factor for all the species were assumed to be unity while solving the problem using the presented solution strategy. Similar conditions were also simulated using Lunn et al.'s solution.

Initial concentrations of all three species were assumed to be zero throughout the column. A constant boundary condition with $c_1 = 1.0$, $c_2 = 0.0$, and $c_3 = 0.0$ was assumed at the inlet boundary, and a free-boundary condition was assumed at the exit boundary. This one-dimensional soil column is described by the differential equation and associated boundary condition given in Eq. (6).

Fig. 1 shows both the solutions derived by Lunn et al. (1996) and the new solution derived here after 400 h when $v = 0.2$ cm h^{-1}. The results from these two solutions are also compared when $v = 0.1$ cm h^{-1}, $v = 0.3$ cm h^{-1}, $v = 0.4$ cm h^{-1}, $v = 0.5$ cm h^{-1}, and $v = 1.0$ cm h^{-1} at 400 h. The maximum concentration difference between two model solutions is 3.79×10^{-5}. The computations were performed using single precision Microsoft FORTRAN PowerStation 4.0 on a 100 MHz Pentium computer. The CPU time required for the model of Lunn et al. (1996) was 84.733 s. while that required for substitution model derived here was 73.385 s. Hence, the solution can be considered identical although the substitution method derived here requires some 15% less CPU time.

6.2. Four-species reactive transport in a one-dimensional soil column

In this section, the solution for a four-species transport coupled by sequential degradation of a contaminant and its daughter products is illustrated in field scale. Four partial differential equations coupled by first-order sequential reaction terms were solved

Table 1
System parameters used in three-species chain problem

Dispersion coefficient	D	0.18	cm^2 h^{-1}
Velocity	v	0.1, 0.2, 0.4	cm h^{-1}
Decay rate of Species 1	k_1	0.05	h^{-1}
Decay rate of Species 2	k_2	0.03	h^{-1}
Decay rate of Species 3	k_3	0.02	h^{-1}

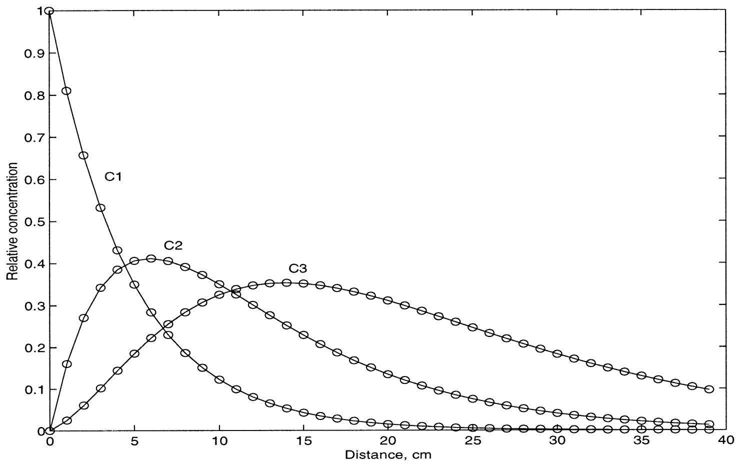

Fig. 1. Comparison of the newly derived solution with the solution from the Lunn et al. (1996) model when $v = 0.2$ cm h^{-1} $t = 400$ h. The solid line represents the newly derived solution and the circle is from the Lunn et al. model.

using the analytical solution technique described above. To be consistent with the three-species case in Section 6.1, the similar initial and boundary conditions were used. Therefore, the solution subroutine which was used to calculate Eq. (7) was also used in this example problem. The first-order reaction rates for the four species were assumed to be 0.05 d^{-1}, 0.02 d^{-1}, 0.01 d^{-1}, 0.005 d^{-1}. Other parameters used in this example are $v = 0.2$ and 0.5 m d^{-1}; $D = 0.3$ m^2 h^{-1}; $R_1 = R_2 = R_3 = R_4 = 1.0$.

Figs. 2 and 3 shows the concentration profiles of these four species after 100 d, 200 d, 300 d, and 400 d, respectively, when $v = 0.2$ m d^{-1} and $v = 0.5$ m d^{-1}.

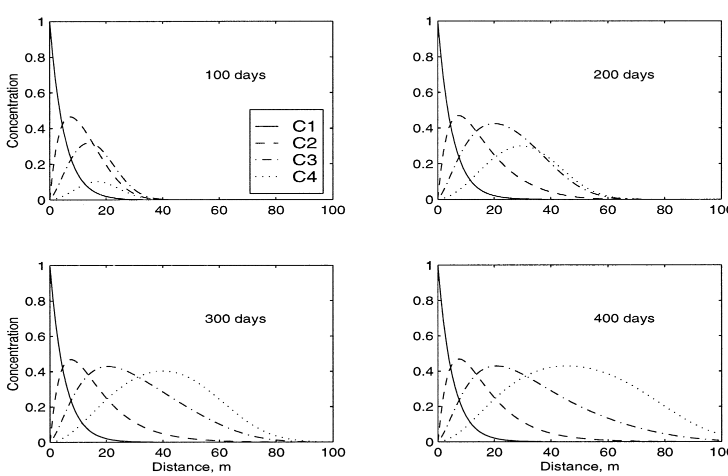

Fig. 2. Concentration profiles of the four-species for one-dimensional reactive transport after 100 d, 200 d, 300 d, and 400 d. $v = 0.2$ m d^{-1}.

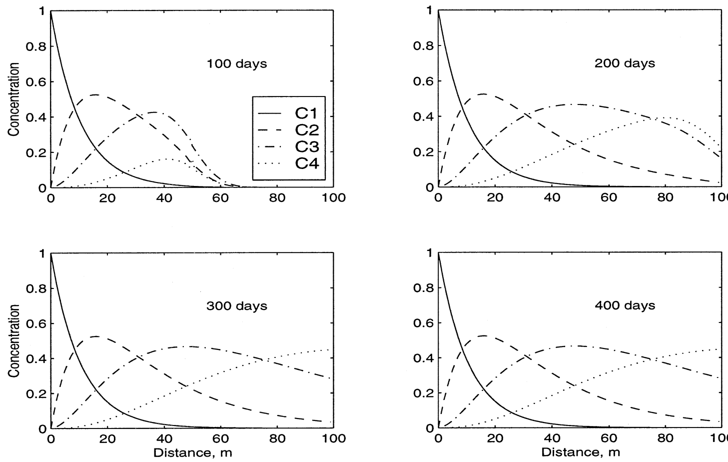

Fig. 3. Concentration profiles of the four-species for one-dimensional reactive transport after 100 d, 200 d, 300 d, and 400 d. $v = 0.5$ m d^{-1}.

6.3. Four-species reactive transport in a three-dimensional aquifer

Domenico (1987) derived an analytical solution of a single-species transport with first-order reaction in three dimensions as:

$$c(x,y,z,t) = \frac{c_o}{8}\exp\left\{\frac{x\left[1-(1+4k\alpha_x/v)^{1/2}\right]}{2\alpha_x}\right\}$$
$$\times \operatorname{erfc}\left\{\frac{x-vt(1+4k\alpha_x/v)^{1/2}}{2(\alpha_x vt)^{1/2}}\right\}$$
$$\times\left\{\operatorname{erf}\frac{y+Y/2}{2(\alpha_y x)^{1/2}} - \operatorname{erf}\frac{y-Y/2}{2(\alpha_y x)^{1/2}}\right\}$$
$$\times\left\{\operatorname{erf}\frac{z+Z/2}{2(\alpha_z x)^{1/2}} - \operatorname{erf}\frac{z-Z/2}{2(\alpha_z x)^{1/2}}\right\} \tag{14}$$

where Y and Z are the source dimensions $[L]$, and α_x, α_y, α_z are, respectively, longitudinal, transverse, and vertical dispersivities $[L]$. Based on this solution, the solution of four species reactive transport in three dimensions was derived using the method described above.

To demonstrate the application, the solution of four species transport was examined in a three dimensional aquifer (100 m $\times$ 41 m $\times$ 25 m). The assumed first-order reaction rates were the same as those used in the one-dimensional case. Other parameters are: $v = 0.2$ m d^{-1}; $\alpha_x = 1.5$ m; $\alpha_y = 0.3\alpha_x$; $\alpha_z = 0.1\alpha_x$; $Y = 11.0$ m $Z = 5$ m.

Fig. 4 shows the four species concentrations after 400 days. The concentration contours are presented, respectively on the horizontal plane where $z = 13.0$ m and

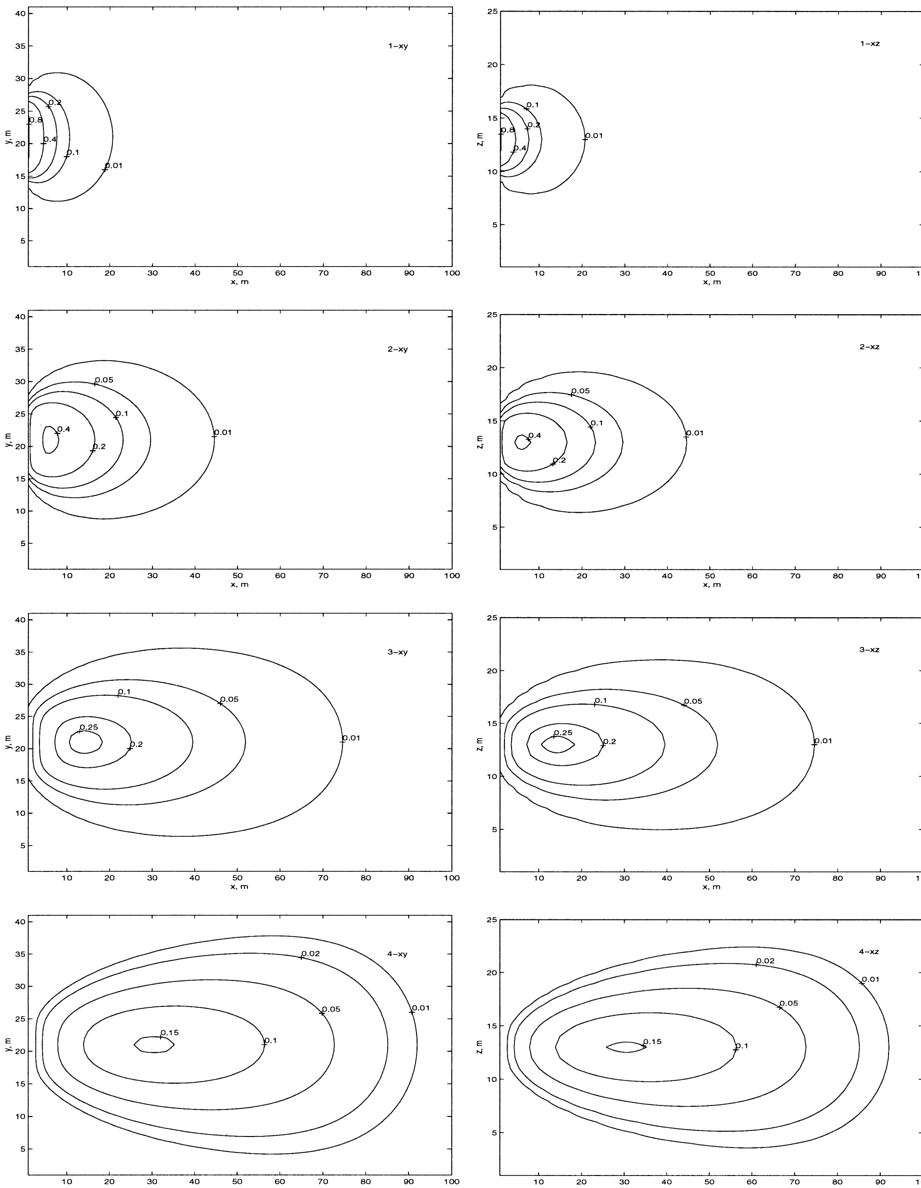

Fig. 4. Concentration profiles of four-species sequential reaction problem. *i-xy* and *i-xz*, $\forall i = 1,2,3,4$, represent concentration contours of ith species on xy $(z = 13$ m$)$ and xz $(y = 21$ m$)$ planes, respectively.

vertical plane where $y = 21.0$ m. Obviously, this transformation method provides a powerful tool for describing analytical solutions for multi-dimensional, multi-species reactive transport problems.

7. Conclusion

A new substitution method has been derived that allows development of analytical solutions for multiple species transport with sequential first-order reactions. The method can be used to derive new analytical solutions for multi-species problems based on available analytical solutions for single species transport with first-order reaction under different initial and boundary conditions.

The solution for a single radioactive tracer decay in a semi-infinite column (Bear, 1979, page 268) was used to derive the solution for three-species transport with first-order reaction. The results of the analytical solution, derived using substitution solution technique is found to be identical to those obtained using the solution of Lunn et al. (1996).

To further demonstrate that the method can be used to describe multi-dimensional, multi-species problems, solutions to a four species, one-dimensional reactive problem and a four-species three-dimensional reactive problem, are obtained. These examples demonstrated that the substitution method can be easily used to obtain solutions to any multi-species and multi-dimensional problems with first-order reaction kinetics. As such, this method provides a powerful tool for solving multi-species reactive transport problems.

Appendix A. Detailed derivation for Eq. (5)

Define

$$SCL = \frac{\partial}{\partial t} - D_x \frac{\partial^2}{\partial x^2} - D_y \frac{\partial^2}{\partial y^2} - D_z \frac{\partial^2}{\partial z^2} + v \frac{\partial}{\partial x}, \tag{A.1}$$

then, the mass balance Eq. (3) for species i is written

$$SCL(c_i) = k_{i-1} c_{i-1} - k_i c_i. \tag{A.2}$$

Using the definition of auxiliary variables,

$$a_i = c_i + \sum_{j=1}^{i-1} \prod_{l=j}^{i-1} \frac{k_l}{k_l - k_i} c_j, \tag{A.3}$$

we have

$$\begin{aligned} SCL(a_i) &= SCL(c_i) + \sum_{j=1}^{i-1} \prod_{l=j}^{i-1} \frac{k_l}{k_l - k_i} SCL(c_j) \\ &= k_{i-1} c_{i-1} - k_i c_i + \sum_{j=1}^{i-1} \prod_{l=j}^{i-1} \frac{k_l}{k_l - k_i} \left[k_{j-1} c_{j-1} - k_j c_j \right]. \end{aligned} \tag{A.4}$$

Combining terms with common factor c_s,

$$-\prod_{l=s}^{i-1} \frac{k_l}{k_l - k_i} k_s c_s, \qquad j = s \tag{A.5}$$

$$\prod_{l=s+1}^{i-1} \frac{k_l}{k_l - k_i} k_s c_s, \qquad j = s+1 \tag{A.6}$$

the sum

$$-\frac{\prod_{l=s}^{i-1} k_l}{\prod_{l=s}^{i-1}(k_l-k_i)} k_s c_s + (k_s-k_i)\frac{\prod_{l=s+1}^{i-1} k_l}{\prod_{l=s}^{i-1}(k_l-k_i)} k_s c_s = -k_i \prod_{l=s}^{i-1}\frac{k_l}{k_l-k_i} c_s. \tag{A.7}$$

Similarly, the last term with c_{i-1} can be combined with the first term in Eq. (A.4), $k_{i-1}c_{i-1}$,

$$\frac{k_{i-1}}{k_{i-1}-k_i}(-k_{i-1}c_{i-1}) + k_{i-1}c_{i-1} = \frac{-k_i k_{i-1}}{k_{i-1}-k_i} c_{i-1}.$$

Therefore,

$$SCL(a_i) = -k_i\left[c_i + \sum_{j=1}^{i-1}\prod_{l=j}^{i-1}\frac{k_l}{k_l-k_i} c_j\right] = -k_i a_i. \tag{A.8}$$

Recalling Eq. (A.1),

$$\frac{\partial a_i}{\partial t} - D_x\frac{\partial^2 a_i}{\partial x^2} - D_y\frac{\partial^2 a_i}{\partial y^2} - D_z\frac{\partial^2 a_i}{\partial z^2} + v\frac{\partial a_i}{\partial x} = -k_i a_i. \tag{A.9}$$

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